



Compact Blue-Green Lasers

**William P. Risk
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COMPACT BLUE-GREEN LASERS

This book describes the theory and practical implementation of three techniques for the generation of blue-green light: nonlinear frequency conversion of infrared lasers, upconversion lasers, and wide-bandgap semiconductor diode lasers.

The book begins with a discussion of the various applications that have driven the development of compact sources of blue-green light. Part 1 then describes approaches to blue-green light generation that exploit second-order nonlinear optics, including single-pass, intracavity, resonator-enhanced and guided-wave second harmonic generation. Part 2, concerned with upconversion lasers, investigates how the energy of multiple red or infrared photons can be combined to directly pump blue-green laser transitions. The physical basis of this approach is thoroughly discussed and both bulk-optic and fiber-optic implementations are described. Part 3 describes wide-bandgap blue-green semiconductor diode lasers, implemented in both II–VI and III–V materials. The concluding chapter reflects on the progress in developing these lasers and using them in practical applications such as high-density data storage, color displays, reprographics, and biomedical technology.

Compact Blue-Green Lasers provides the first comprehensive, unified treatment of this subject and is suitable for use as an introductory textbook for graduate-level courses or as a reference for academics and professionals in optics, applied physics, and electrical engineering.

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Preface

Since the mid-1980s, the development of practical, powerful sources of coherent visible light has received intense interest and concentrated activity. This interest and activity was fueled by twin circumstances: the realization of powerful, efficient infrared laser diodes and the emergence of numerous applications that required compact visible sources. The availability of these infrared lasers affected the development of visible sources in two ways: It stimulated the investigation of techniques for efficiently converting the infrared output of these lasers to the visible portion of the spectrum and it encouraged the hope that the fabrication techniques themselves might be adapted to make similar devices working at shorter wavelengths.

Within the visible spectrum the blue-green wavelength region has demanded – and received – special attention. The demonstration of powerful red diode lasers followed relatively soon after the development of their infrared counterparts – in contrast, the extension to shorter blue-green wavelengths has required decades of wrestling with the idiosyncrasies of wide-bandgap materials systems. The first blue-green diode lasers were not successfully demonstrated until 1991, and it has only been within the past year or two that long-lived devices with output powers of tens of milliwatts have been achieved.

As this field emerged and began to grow, it quickly became evident that it would necessarily be a very multi-disciplinary one. On one hand, a variety of approaches were being pursued in order to generate blue-green light. The three main ones – nonlinear frequency conversion, upconversion lasers, blue-green semiconductor lasers – are the focus of this book. The common goal of developing laser devices capable of emitting as much as several watts in the 400–550-nm spectral range brought together experts in nonlinear optical materials, diode-pumped solid-state lasers, guided-wave optics, rare-earth spectroscopy, semiconductor material processing and laser diode device physics. On the other hand, the range of applications for such devices attracted experts from such diverse fields as biomedical

engineering, display science and technology, optical data storage, and undersea communications.

Capturing this broad range of both approach and application in a book of reasonable length has been challenging, as has been writing clearly for readers that we expect will come to this book from a wide variety of disciplines and backgrounds. In the interest of clarity, we have included some material introducing and explaining basic concepts of nonlinear optics, rare-earth spectroscopy, and semiconductor device physics. Some readers will already be completely familiar with this material and may wish to skip directly to sections that explain in greater depth the application of these basic principles to specific approaches for generating blue-green light. Other readers may appreciate a brief refresher in some of these concepts – the reader who is fully conversant with nonlinear optics, rare-earth spectroscopy, and semiconductor device physics is probably a rare creature! Still other readers may wish to consider some of these basic ideas in greater depth – for these, we have recommended where possible other books that treat these subjects and have also made available some supplementary material on the Cambridge University Press website at <http://publishing.cambridge.org/resources/0521623189>.

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1

The need for compact blue-green lasers

1.1 A SHORT HISTORICAL OVERVIEW

For years after its invention in 1961, the laser was described as a remarkable tool in search of an application. However, by the late 1970s and early 1980s, a variety of applications had emerged that were limited in their implementation by lack of a suitable laser. The common thread running through these applications was the need for a powerful, compact, rugged, inexpensive source of light in the blue-green portion of the spectrum. The details varied greatly, depending on the application: some required tunability, some a fixed wavelength; some required a miniscule amount of optical power, others a great deal; some required continuous-wave (cw) oscillation, others rapid modulation.

In many of these applications, gas lasers – such as argon-ion or helium-cadmium lasers – were initially used to provide blue-green light, and in some cases were incorporated into commercial products; however, they could not satisfy the requirements of every application. The lasing wavelengths available from these lasers are fixed by the atomic transitions of the gas species, and some applications required a laser wavelength that is simply not available from an argon-ion or helium-cadmium laser. Other applications required a degree of tunability that is unavailable from a gas laser. In many of them, the limited lifetime, mechanical fragility, and relatively large size of gas lasers was a problem.

At about the same time, new options for generation of blue-green radiation began to appear, due to developments in other areas of laser science and technology. The development of highly efficient, high-power semiconductor diode lasers at wavelengths around 810 nm opened up the possibility of diode-pumping solid-state lasers, such as those based on neodymium-doped crystals and glasses. New and improved nonlinear materials made it practical to apply second-harmonic generation to the infrared outputs of these diode-pumped solid-state lasers to generate wavelengths in the blue-green regions of the spectrum. Demonstrations in

1986 of compact green sources based on intracavity frequency doubling of diode-pumped neodymium lasers by researchers at Spectra-Physics and Stanford University sparked tremendous interest in sources based on this approach. This interest has led to commercially-available diode-pumped green sources with powers of several watts and, more recently, blue sources with powers of several milliwatts.

Rather than pump a neodymium laser, why not simply use nonlinear optics to frequency double the output of an infrared semiconductor laser directly? The reason has been, until fairly recently, that high-power semiconductor diode lasers have had rather broad spectral distributions and rather poor spatial beam quality. While these characteristics did not prevent the use of these diode lasers as pumps for solid-state lasers, they did inhibit their use for direct nonlinear frequency conversion, in which the spectral and spatial mode properties of the infrared source are much more critical. As the spatial and spectral mode properties of high-power semiconductor diode lasers have improved, however, the same techniques of nonlinear frequency conversion have been applied to direct frequency-doubling of these devices, and efficient blue and green sources have been demonstrated. In some cases, resonant enhancement and guided-wave geometries have been used to increase the efficiencies of these nonlinear interactions.

An alternative approach to blue-green light generation using infrared sources is the so-called "upconversion laser". In a standard laser, energy conservation requires that the energy of an absorbed pump photon be greater than the energy of an emitted laser photon; hence the pump wavelength must be shorter than the lasing wavelength. In upconversion lasers, the energy from two or more pump photons is combined to excite the lasing transition; thus the pump wavelength can be longer than the lasing wavelength, so that, for example, infrared light can be used to directly pump a green laser. Although upconversion lasing was demonstrated in 1971 by Johnson and Guggenheim (1971), the field remained largely dormant for several years because flashlamp pumping of such lasers was inefficient. Experiments conducted at IBM in 1986 which demonstrated efficient laser pumping of upconversion lasers revived interest in the field. These initial experiments used bulk rare-earth-doped crystals and had to be performed at cryogenic temperatures, but they demonstrated the feasibility of these devices, including the fact that they could be efficiently pumped with laser diodes. Later, efficient room-temperature operation was achieved using optical fibers doped with rare-earth elements.

Perhaps the most direct and attractive way to generate blue and green light is to use a semiconductor diode laser. Semiconductor laser devices are efficient, small, robust, rugged, and powerful. However, in order to generate blue-green radiation, semiconductors with bandgaps of ~ 3 eV must be used. Suitable materials systems include II–VI semiconductors such as ZnS and ZnSe, and wide-gap III–V materials such as GaN. The growth of thin films of these semiconductors suitable

for device fabrication has proven to be an extremely difficult challenge. However, breakthroughs in the growth of appropriately-doped films in both material systems has allowed the demonstration of light-emitting diodes (LEDs) and, more recently, lasers in both material systems. However, despite rapid progress, demonstration of continuous-wave (cw) operation at room temperature with powers and lifetimes comparable to infrared semiconductor lasers has not yet been achieved, and more development is required before these lasers can be used in the applications cited above.

1.2 APPLICATIONS FOR COMPACT BLUE-GREEN LASERS

One of the factors that has made the field of compact blue-green lasers interesting and vibrant is its diversity in both the variety of technical approaches used to produce them and the wide range of applications for which they have been sought. The specialized topical meetings that sprang up in the early 1990s in response to the intense interest and activity in this field (such as the Optical Society of America's Topical Meeting on Compact Blue-Green Lasers, held in 1992, 1993, and 1994) brought together researchers from such disparate fields as submarine communications and DNA sequencing. In this section, we review some of the principal applications for which blue-green lasers have been sought, and the requirements placed on the lasers by these uses.

1.2.1 Optical data storage

The terms "optical data storage" and "optical recording" have been used to refer to a variety of different approaches for recording and retrieving information using optical methods, including those based on such exotic phenomena as persistent spectral hole burning (Lenth *et al.*, 1986). However, these terms usually refer to somewhat more mundane systems that read data from (and, in some cases, write data to) spinning disks in a fashion analogous to magnetic disk drives (Figure 1.1).

In optical data storage systems, a bit is stored on the disk by altering some physical characteristic of the disk in a tiny spot. This alteration can be done once, as in the case of read-only disks (such as audio CDs and CD-ROMs), or it can be done repeatedly, as in the case of rewritable disks (such as those based on magneto-optic or phase-change media). To read back the information stored on an optical disk, a focused laser beam is scanned over these spots and the light reflected from the disk is detected. The physical characteristic that was altered to record a bit must produce a corresponding change in some optical property of the reflected beam. In audio CDs and CD-ROMs, data are impressed upon a plastic disk in the form of tiny pits stamped into the disk by the manufacturer. The depth of these pits is one-fourth

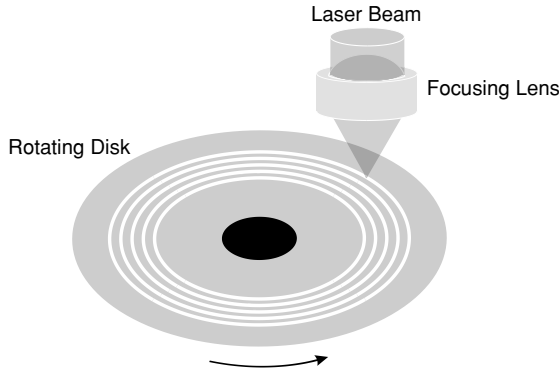


Figure 1.1: Optical data storage system.

the laser wavelength, so that when the beam is scanned over the pit, the portion reflected from the bottom of the pit travels an additional half-wavelength compared with the light reflected from the surface of the disk and is therefore 180° out-of-phase with it; thus, the amplitude of the reflected beam is diminished due to destructive interference. In “magneto-optic” media, data are recorded by using the laser beam as a heater: the focused laser spot heats the magnetic material above the Curie temperature, and the presence of an applied magnetic field causes the magnetization of the medium to reverse in the heated region. When the heating is removed and the material cools below the Curie temperature, that reversed magnetization is “frozen in”. The data can be read back by exploiting the fact that the polarization of light reflected from the disk in these materials depends on the orientation of the magnetic domain (the “polar Kerr effect”). In “phase-change” material, data are recorded by using the focused laser beam to melt the material locally and induce a phase transition from what was originally a crystalline structure to an amorphous one. Data are read back by exploiting the fact that the amorphous state of the material has a different reflectivity than the crystalline state.

In order to write a small mark and be able to read it back accurately, the laser beam must be focused to as small a spot as possible. A gaussian beam can be focused by a lens to a diffraction-limited spot with a diameter d of

$$d \simeq \frac{\lambda}{NA}$$

where λ is the wavelength and NA is the numerical aperture of the lens. Therefore, one way to achieve a smaller spot size is to reduce the wavelength. Halving the wavelength from that of a GaAlAs laser diode at 860 nm to that of a blue laser at 430 nm would cut the spot size in half, and could quadruple the storage density. In addition, for a given rotation rate of the disk, the data rate could be increased by a

factor of 2, since the marks can be placed twice as close together. An additional motivation to pursue development of blue-green lasers for magneto-optic storage was the discovery of garnet-based recording materials that exhibit better performance in the blue-green regions of the spectrum than do their counterparts designed for use in the infrared (Eppler and Kryder, 1995).

Using a blue-green laser in an optical storage system places severe demands upon its performance (Kozlovsky, 1995). In the magneto-optic approach, the power required is comparable with that demanded of the infrared diode lasers used for optical storage (~ 40 mW) (Asthana, 1994). This may seem counterintuitive – one might expect that since the beam is focused to a smaller spot, less power would be required to produce the same temperature increase for writing. This statement is true as far as it goes; however, when reading data back with a blue beam, there are fewer photons per milliwatt than would be present in an infrared beam, which leads to increased noise. In order to obtain an adequate signal-to-noise ratio, the recording medium must be de-sensitized so that a higher readback power can be used without erasing the data. Thus, something like 2–6 mW is required for reading and 40–50 mW are required for writing. For focusing to a small spot, the wavefront aberration of the blue beam must be less than 0.05 wavelengths. The noise of the blue beam must be low: < -110 dBc (decibels below carrier) for magneto-optic storage, where differential detection is used, and < -135 dBc for phase change and CD-ROM, where single-ended detection is used. The laser must have a long lifetime, ideally as long as the lifetime of the drive itself (perhaps 100 000 hours mean-time-between-failures). Finally, the laser must be inexpensive.

1.2.2 Reprographics

Reprographic applications use lasers in a fashion similar to optical data storage – the laser is focused to a small spot and used to make a mark on some medium. Here, however, the medium is the photoconductor of a laser printer, or photographic film or paper. Except in certain specialized applications (for example, writing on microfilm), reprographics does not require as small a spot size as optical data storage. A laser printer with 2400 dpi resolution requires that the laser beam be focused to only a $10\text{ }\mu\text{m}$ spot, a size that can be achieved easily using a red or near-infrared diode laser. However, this $10\text{ }\mu\text{m}$ spot size must be maintained as the beam is scanned rapidly over a page several centimeters wide. Decreasing the wavelength for a particular spot size relaxes the design requirements of the optical system by reducing the numerical aperture required to form a spot of the desired size and by increasing the depth-of-field.

In color reprographics, lasers can be used to expose photographic paper or film (Owens, 1992). The considerations just described for laser printers also apply here.

In addition, the wavelengths of the lasers must be chosen to provide correct exposure for existing photographic media. For photographic films, wavelengths of 430 nm, 550 nm, and 650 nm (blue, green, red) are desired. For photographic papers, wavelengths of 470 nm, 550 nm, and 700 nm are preferred. Powers of a few milliwatts are needed, along with good beam quality, low noise, and high stability. The ability to directly modulate the laser at frequencies up to 50 MHz is desirable.

1.2.3 Color displays

Blue-green lasers have also been sought for use in color displays. At present, the most popular type of color display device is the cathode ray tube (CRT) used in computer monitors and color televisions. In CRTs, colors are synthesized through the superposition of three primary colors – red, green, and blue – generated by an electron beam striking one of three corresponding phosphors. The combination of these red, green, and blue emissions in various proportions creates the other colors visible on the screen. A similar approach has been proposed for laser-based displays, in which three separate lasers would provide red, green, and blue primary colors that could be combined to project full-color images on a large screen (Figure 1.2). Each laser could be raster-scanned across the screen, or could remain stationary and be used to illuminate an “image gate”, such as motion picture film or a spatial light modulator containing the image to be projected.

Lasers are attractive light sources for display applications because of their high brightness and complete color saturation. The brightness of a laser (power emitted per unit area per unit solid angle) can be very high due to the directionality of

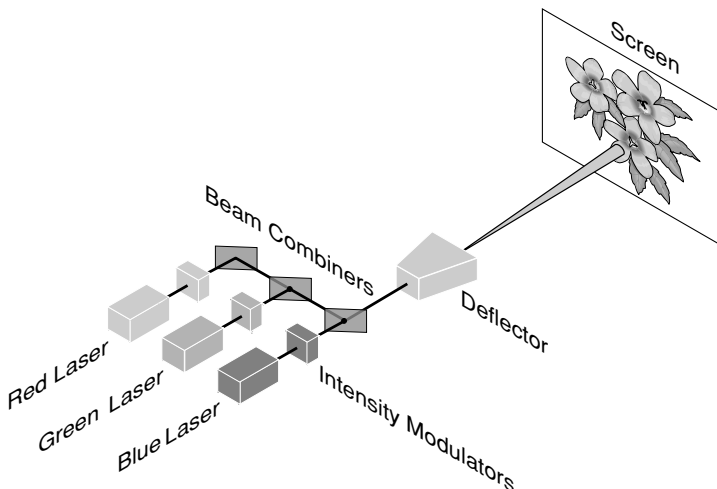


Figure 1.2: Laser-based color projection display.

the beam. This high brightness leads to high efficiency for a laser-based projector, since most of the generated optical power can be directed by appropriate optics to illuminate the screen or image gate. In contrast, a conventional motion picture projector uses an incandescent bulb that emits light into a 4π -steradian solid angle, most of which never reaches the screen. The ability of a laser to concentrate the emitted light into a confined solid angle provides an efficiency advantage over competing technologies (Glenn and Dixon, 1993).

Another advantage of laser-based displays is improved color saturation. In conventional CRT displays, the light emitted by the phosphor is not spectrally pure; the spectral bandwidth of the emission may be several nanometers. In the language of color theory, the red, green, and blue colors emitted by these phosphors are not “fully saturated”: that is, the primary colors are not the “bluest blue”, “greenest green”, or “reddest red” that the eye can perceive, but appear somewhat washed out by the addition of white. As a result, a CRT cannot reproduce the entire range of colors perceptible to human vision, and in particular, cannot produce fully saturated colors. The range of colors that can be produced by addition of primaries can be depicted by the “CIE chromaticity diagram” (Figure 1.3). In this diagram, fully saturated colors (monochromatic light waves of a specified wavelength) correspond to points around the periphery. White corresponds to a point in the interior of the

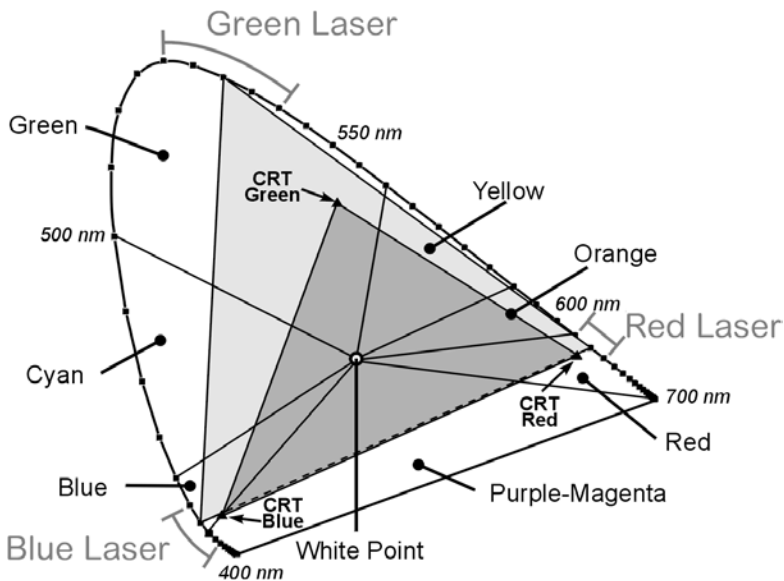


Figure 1.3: CIE diagram showing the color space spanned by CRT phosphors (dark shading) and the color space which could be spanned in a color display using monochromatic red, green, and blue lasers to generate the primary colors (lighter shading). The primary colors for each system fall at the corners of the triangles, as indicated.

diagram. If one draws a line from the “white point” out to a particular color on the periphery, the points along that line represent various saturation levels of the same color; for example, fully saturated green corresponds to a point on the periphery, and points along the line correspond to increasingly paler shades of green as one moves toward the white point. If one plots the points corresponding to three primary colors on such a diagram, the range of colors that can be synthesized by combining these primaries corresponds to the interior of the triangle connecting the three primary points. Figure 1.3 shows the points corresponding to the primary colors of a standard color CRT monitor. Although the red CRT phosphor is nearly saturated, the blue and green phosphors are considerably less so. Thus, while a CRT monitor can produce well-saturated reds, it is difficult to achieve well-saturated blues and greens. A laser-based color display produces primary colors that are fully saturated (that is, spectrally-pure monochromatic waves); thus, the range of colors that can be produced is greater and the colors themselves are richer than in a CRT. In order for the primary colors to appear to human vision as true blues, greens, and reds, they must fall within the wavelength ranges depicted in Figure 1.3: $605 \text{ nm} \pm 5 \text{ nm}$ for red, $530 \text{ nm} \pm 10 \text{ nm}$ for green, and $470 \text{ nm} \pm 10 \text{ nm}$ for blue (Glenn and Dixon, 1993). The power required varies depending upon the size of the screen, but ranges from approximately 1 W per color for a 10-ft $\times$ 16-ft screen to 20 mW per color for a 16-in CRT-like display (Valley and Ansely, 1997).

1.2.4 Submarine communications

Communication with a submerged vessel has been another important application driving the development of blue-green lasers. Naval forces would like to be able to send messages to their submarines without requiring them to rise from their operating depth and risk detection by an enemy (Figure 1.4). Ideally, such a communications system would be able to send a signal through seawater to a great depth and to transmit information at a rapid rate. Electromagnetic radiation can penetrate seawater to a significant depth only at extremely low frequencies (ELFs) ($\lesssim 100 \text{ Hz}$) or in the blue-green portion of the optical spectrum, where minimum attenuation (the “Jerlov Minimum”) occurs for wavelengths between 400 nm and 500 nm (Figure 1.5). Although ELF systems have been built and used to send messages to submarines, systems using ELFs also have extremely low data rates, and in practice, only extremely short messages can be sent. Transmitting with blue-green wavelengths could make it possible to send messages to great depth with much higher data rate than with ELF. However simple this may sound in principle, the development of such a system has presented such great technical challenges that it has been described as “the most complex communications system known to man” (Painter, 1989).

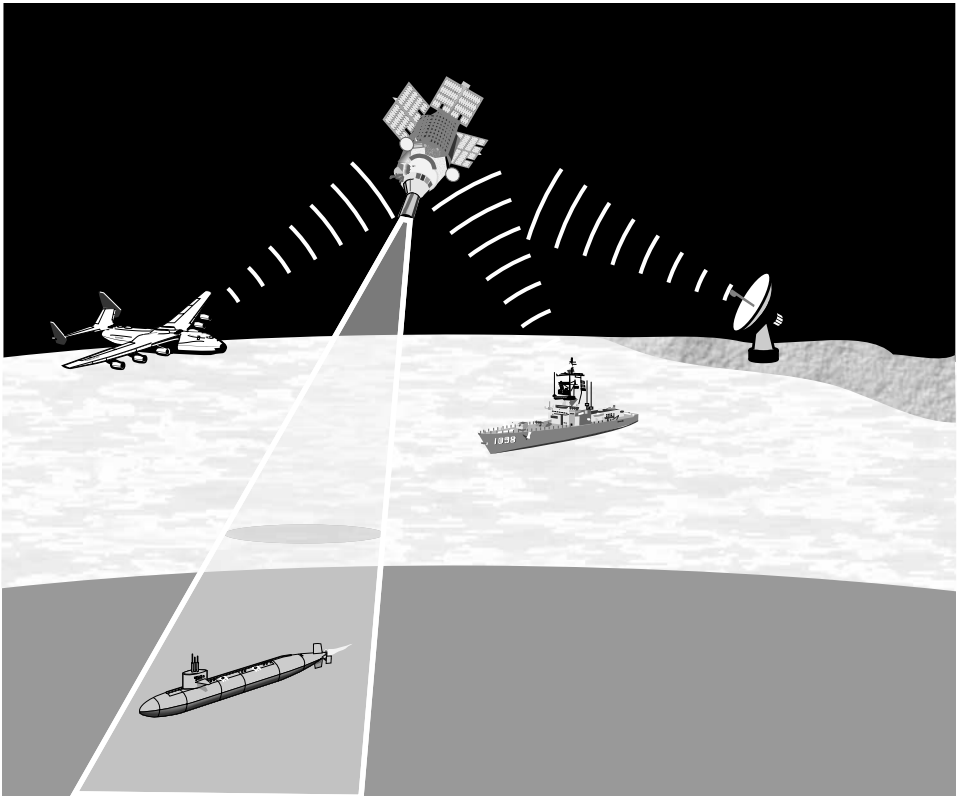


Figure 1.4: Signals are sent by conventional radio from a surface ship, ground station, or aircraft to an orbiting satellite. A blue-green laser aboard the satellite then relays the message to a submerged submarine. (Adapted with permission from Painter (1989).)

What are these challenges? Even at the Jerlov Minimum, the attenuation of seawater is not negligible, and the signal reaching a submerged vessel may be quite weak, requiring the receiver aboard the submarine to be very sensitive. This sensitivity introduces an additional complication: sunlight contains a significant blue-green component which can also penetrate the ocean and introduce noise into the received signal. One way to solve this problem is to exploit the difference between the very narrow spectral width of the blue-green laser and the much broader spectral distribution of sunlight. An optical filter with a sufficiently narrow passband can transmit most of the blue-green laser photons to the detector while rejecting most of the solar photons. In addition to a narrow passband, such a filter must have a wide field-of-view. Photons transmitted from a satellite to a submarine may pass through cloud layers that introduce scattering, and are further scattered during passage through the sea, so that they may impinge upon the submarine from a variety of

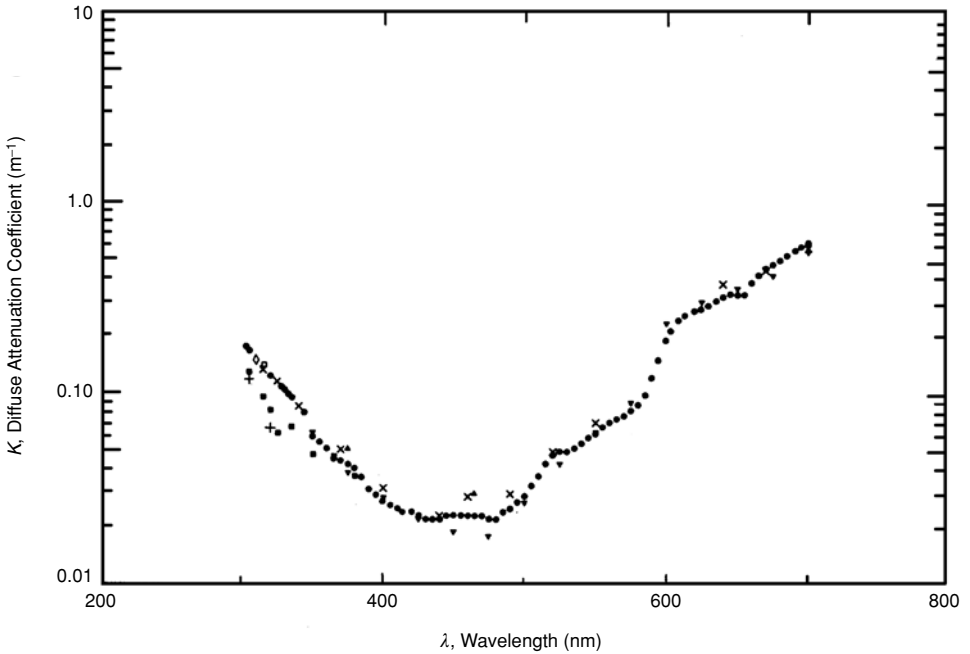


Figure 1.5: Attenuation of seawater over the blue-green portion of the optical spectrum, showing the “Jerlov Minimum” near 450 nm. Various points correspond to measurements by different authors. [Reprinted by permission from Smith and Baker (1981).]

angles. Simultaneously meeting both these requirements – narrow passband and wide field-of-view – is difficult.

The most successful approach devised to meet this challenge is the “atomic resonance filter” or “ARF” (also called “QLORD” – “quantum-limited optical resonance detector”), which has the narrow passband and wide field-of-view required for submarine communications (Gelbwachs, 1988). The ratio of the spectral width $\Delta\lambda$ to center wavelength λ_0 of the passband in these filters can be $\Delta\lambda/\lambda_0 \simeq 10^{-6}$. Thus, for a center wavelength $\lambda_0 \sim 500$ nm, the width of the passband can be as narrow as ~ 0.005 Å (Marling *et al.*, 1979)! An ARF based on cesium vapor is particularly suited to submarine communications and has been pursued for this purpose. The operation of the cesium ARF can be understood from Figure 1.6. A conventional filter (e.g., colored glass such as BG-18) allows only blue-green light to enter the cesium cell. In the cesium vapor, light at 456 nm or 459 nm is absorbed to excite population from the $6s$ level to the $7p$ level. This population subsequently decays nonradiatively to the $6p$ level, through either the $7s$ or $5d$ levels. When the $6p$ population relaxes back to the ground state, infrared photons at 852 nm or 894 nm are emitted. Another conventional filter (such as RG-715 glass) permits only infrared radiation to impinge upon the detector. Since there is no overlap in the passbands of the two conventional filters, no light would reach the detector if the cesium cell

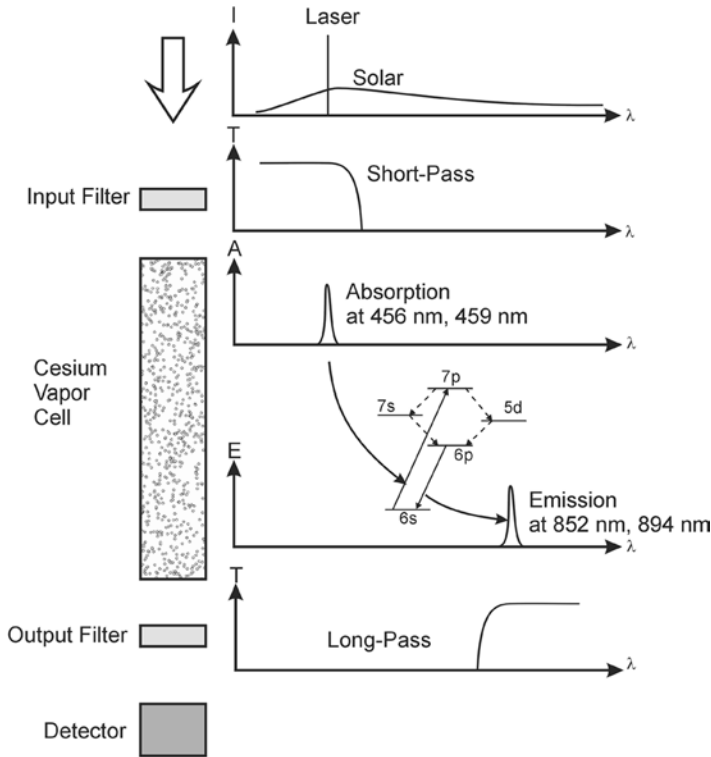


Figure 1.6: Operation of the cesium ARF. I: intensity of incident light, T: transmission of filter, A: absorption of cesium cells, E: emission of cesium cell.

were not present. Thus, the only photons that can impinge upon the detector are those that are converted in wavelength through absorption and reemission by the cesium cell. Since the linewidth of the atomic transition is very narrow, the ARF can have the very narrow passband required for rejection of the solar background and reception of the blue-green laser signal.

However, the same factors which make the cesium filter advantageous for use in submarine communications place stringent requirements upon the blue-green laser. The blue-green laser must be tuned precisely to excite the $6s-7p$ transition; thus, the wavelength must fall within a window approximately $0.01 \text{ \AA}$ wide near 456 nm or 459 nm. The narrow spectral width of this transition requires that the laser linewidth be less than $\lesssim 1 \text{ GHz}$ and be stable to the same degree (Leslie, 1995). The required power is in the kilowatt range (*Laser Focus World*, 1980).

The transmission characteristics of seawater also dictate the use of a blue-green laser for a related application: high-resolution optical imaging of the ocean floor (MacDonald *et al.*, 1995). Here, a blue-green laser carried by a moving submarine is scanned across the seafloor perpendicular to the line of travel and the reflected light is collected to create one line of the image. As the submarine moves forward, successive line scans are collected and used to build up a two-dimensional image.

Table 1.1. *Ions and wavelengths of interest for laser cooling of trapped ions*

Ion	Wavelength	References
Ca	397 nm, 442.7 nm	Urabe <i>et al.</i> (1992), Hayasaka <i>et al.</i> (1994), Beverini <i>et al.</i> (1996)
H	243 nm	Zimmermann <i>et al.</i> (1995)
Mg	383 nm	Beverini <i>et al.</i> (1996)
Pb	368.3 nm	Tamm (1993)
Rb	421 nm	Hemmerich <i>et al.</i> (1990)
Sr	422 nm	Barwood <i>et al.</i> (1992), Barwood <i>et al.</i> (1993)
Yb	369.5 nm	Tamm (1993)

This technique can provide detailed maps of geographical features of the seafloor, or of man-made features such as pipelines.

1.2.5 Spectroscopic applications

Laser cooling of trapped ions is of interest as the basis for optical frequency standards (Itano, 1991). In this approach, an ion is held in an electromagnetic trap and cooled using radiation pressure from a laser slightly detuned from an absorption transition. Several ions that have been proposed for this application require a ultra-violet or blue laser for excitation of the relevant transition (Table 1.1). Convenient sources in the 300–500 nm spectral range are therefore useful for spectroscopy and laser pumping of such transitions. In most cases, relatively modest powers are required (at most, a few milliwatts), but the blue output must be tunable.

A spectroscopic use of blue-green lasers with great immediate practical application is *in situ* process control of physical vapor deposition (PVD). A number of technologically important materials are deposited in thin films from a vapor state, using techniques such as evaporation and sputtering. The deposition rate of these materials is typically measured using a quartz crystal monitor or ion gauge. However, these techniques are not well suited to the deposition requirements of many modern, technologically-important materials. For example, traditional rate-monitoring techniques are inadequate for deposition of superconducting films, in which a high background pressure of oxygen is required, and for co-deposition of composite or alloy films, in which it is necessary to simultaneously monitor and control the flux of more than one species. In addition, these monitoring techniques require physically placing a sensor within the deposition chamber. Since the sensor must not obscure the target, or otherwise interfere with deposition of materials on it, it necessarily cannot directly measure the characteristics of the flux incident upon the substrate.

An alternative technique for monitoring the evaporation flux is atomic absorption spectroscopy. In this approach, a laser beam is passed through the atomic vapor and

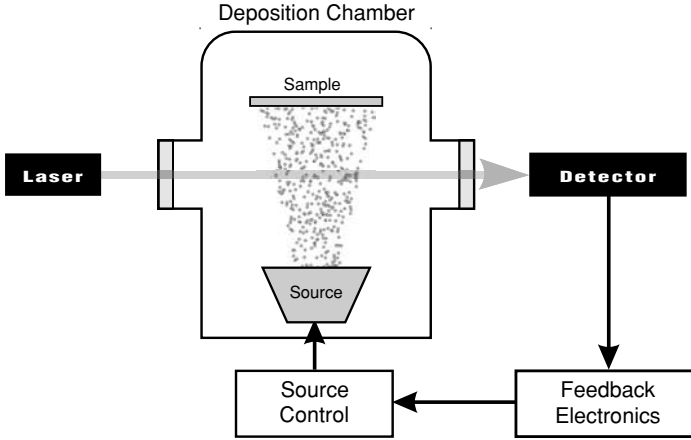


Figure 1.7: A tunable blue-green laser is used for monitoring and controlling the deposition rate in a PVD system.

is tuned to an absorption line (Figure 1.7). The beam is attenuated by absorption in the vapor and the emerging intensity is given by Beer's law, $I = I_0 e^{-n\sigma l}$, where I_0 is the initial intensity, I is the output intensity, n is the density of the evaporant, σ is the absorption cross-section, and l is the path length. By measuring the effect of the atomic vapor on the intensity of the transmitted laser beam, the evaporation rate can be determined. In addition, this information can be fed back to the vapor source and used for closed-loop control of the evaporation rate. Unlike other techniques for monitoring evaporation rate, atomic absorption spectroscopy is noninvasive, can be used with unusual deposition geometries, is highly sensitive and species selective, and can probe both the spatial and velocity distribution of the evaporant.

Atomic absorption spectroscopy can be implemented using hollow cathode lamps; however, these lamps have a number of unattractive features, including relatively short lifetime (~ 1000 h), low intensity, and broad spectral width (Benerofe *et al.*, 1994). The use of a laser instead of a hollow cathode lamp offers several advantages: long lifetime, high intensity, and narrow spectral width can be obtained. Because the linewidth of the laser can be very narrow, Doppler broadening of the evaporated flux can be investigated. Sophisticated spectroscopic techniques, such as frequency-modulation spectroscopy (Bjorklund, 1980) or nonlinear spectroscopy (Levenson, 1982) can be employed.

There are several technologically-important materials that are deposited by PVD and that have absorption lines in the blue-green portion of the spectrum, including: aluminum (394 nm) (Wang *et al.*, 1996), titanium (391 nm) (Galanti *et al.*, 1996), yttrium (408 nm) (Wang *et al.*, 1995), tungsten (385 nm), and gallium (403 nm). Again, the laser must be tunable over the relevant absorption line and powers on the order of a milliwatt are required.

1.2.6 Biotechnology

Blue-green lasers also have uses in the field of biotechnology (Trainor, 1990). One of the most important applications is flow cytometry, in which cells that have certain properties are counted or measured as they are forced to flow by a detector. Cells with the desired property can be detected by attaching to them a fluorescent molecule, or “fluorophore.” The flowing cells are forced to pass through the focused beam of a laser that is tuned to excite the fluorophore, and the presence of the cell can be detected by looking for the associated fluorescence.

One particularly important example of flow cytometry is DNA sequencing. The objective of this procedure is to determine the sequence of the four nucleotides (adenosine, cytosine, guanosine, and thymidine) that encode genetic information in the molecular structure of DNA. Such sequences are important for understanding and diagnosing human genetic disorders and for examining forensic evidence in criminal cases. In one technique for DNA sequencing, a portion of the code is determined by creating a series of replicas of a particular section of the DNA molecule. Each replica starts at the same point in the sequence, but differs in length from the other replicas by one nucleotide (Figure 1.8). Thus, if the replicas can be sorted by length and if the terminal nucleotide can be determined, the DNA sequence can be deduced.

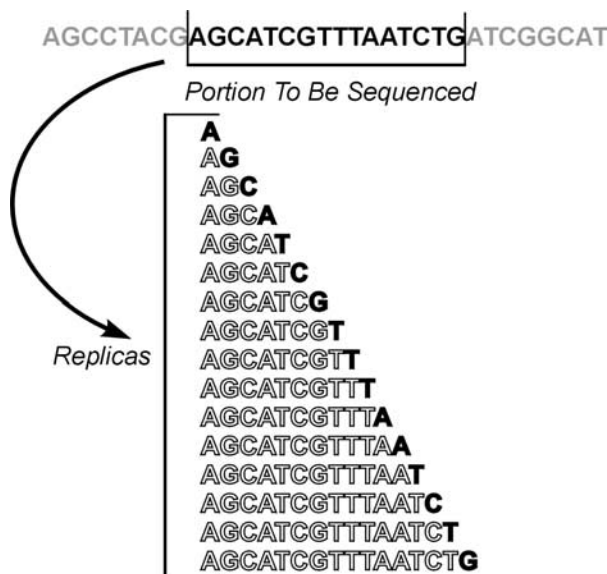


Figure 1.8: A portion of the DNA sequence is determined by making a series of replicas, each of which differs by one nucleotide in length. If the replicas can be sorted by length, and the terminating nucleotide can be determined, then that portion of the DNA sequence can be reconstructed.

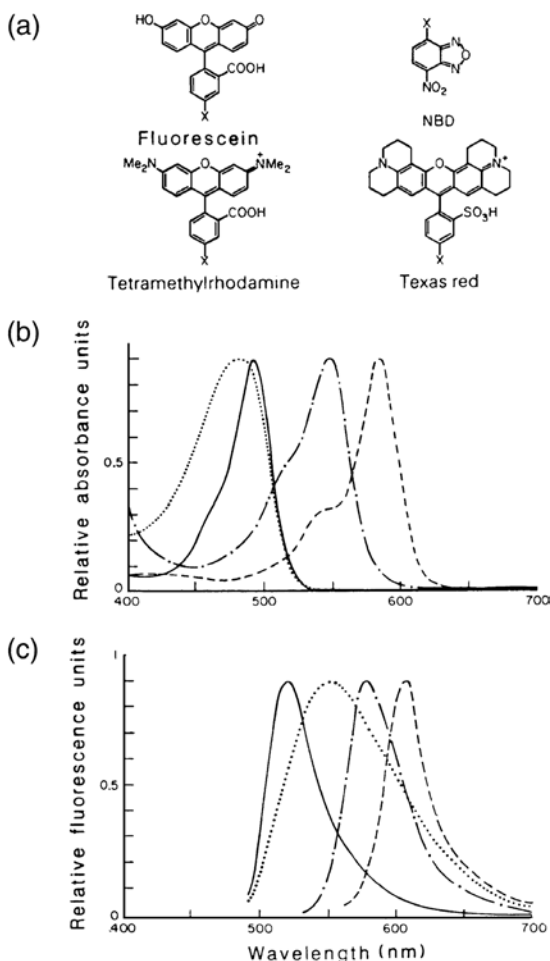


Figure 1.9: (a) Four dyes used for tagging DNA nucleotides. (b) The corresponding absorption spectra. Solid curve: fluorescein; dotted curve: NBD; dashed curve: Texas Red; dot-dash curve: tetramethylrhodamine. (c) The corresponding fluorescence spectra. Same line types as in (b). (Reprinted with permission from Smith *et al.* (1986). Copyright: Macmillan Magazines Limited.)

The terminating nucleotide of each replica can be determined by “tagging” the replica with one of four fluorophores, each fluorophore marking the presence of one of the four possible nucleotides. The fluorophores are chosen such that their emission peaks are sufficiently separated to be easily resolved and identified. Four common fluorescent molecules used for this purpose are shown in Figure 1.9, together with their absorption and emission spectra (Smith *et al.*, 1986). All of these dyes can be excited using blue-green wavelengths.

The replicas can be sorted by length by forcing them to diffuse through an electrophoresis cell. Longer replicas move more slowly than short ones; thus, the

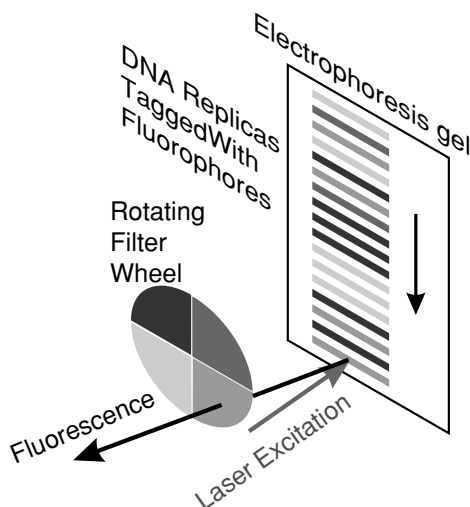


Figure 1.10: DNA replicas are sorted by diffusion through an electrophoresis cell. The nucleotide terminating each replica is tagged with a fluorophore. As each replica reaches the end of the gel, excitation from a blue-green laser excites emission from the fluorophore. The fluorophore and corresponding nucleotide are identified by analyzing the fluorescent emission through a filter wheel. (Adapted with permission from Trainor (1990). Copyright: American Chemical Society.)

replicas become spatially separated as they pass through the cell, spreading out like runners of different speeds in a race (Figure 1.10). This spatial separation corresponds to a temporal one; each replica crosses the “finish line” at a different time. This spatial/temporal separation is remarkably linear with respect to the length of the replica, and replicas that differ by only one nucleotide in length can be easily distinguished. As each replica reaches the end of the gel, it passes through a focused blue-green laser beam. The blue-green photons are absorbed by the fluorophore, which then fluoresces, emitting photons at its own characteristic wavelength. The fluorophore can then be identified by analyzing the spectral content of the emitted photons; this can usually be done simply with a filter wheel divided into four sections, each of which passes only the signal associated with one particular fluorophore.

A great deal of development and engineering has gone into DNA sequencers based on this technology, and several commercial products are available that use argon-ion lasers as the source of blue-green light. Hence, for compatibility with existing products, manufacturers of DNA sequencers and other flow cytometry instrumentation seek compact blue-green lasers operating at wavelengths of 488 nm and 514 nm. The powers required can be as low as 10 mW and as high as several watts, depending on the details of the application (Sklar, 1992).

1.3 BLUE-GREEN AND BEYOND

In addition to the representative applications discussed above, there are numerous others – including straightforward, but commercially important ones (such as CD mastering (Kaneda and Kubota, 1997) and semiconductor wafer inspection) and rather exotic ones (such as calibration of neutrino detectors (Kitchin and Newcomer, 1994)). These all exploit some property of blue-green light in much the same way as those applications we have already discussed.

In addition, the technologies explored for compact blue-green light generation border on other fields where identical ideas are employed, even if generation of blue-green light is not the primary objective. For example, efficient nonlinear frequency-doubling of near-infrared light in resonators (Pereira *et al.*, 1988) and waveguides (Anderson *et al.*, 1995) has been of great interest in the production and study of squeezed states. The same ideas used in upconversion lasers have been applied to three-dimensional displays based on exciting visible fluorescence by absorption of two infrared photons in a volume element defined by the intersection of two diode laser beams in a cube of rare-earth-doped glass (Downing *et al.*, 1996). The development of efficient blue LEDs based on II–VI and III–V semiconductors has been both a precursor to the development of lasers based on the same technology and an important end in itself, as there are many applications for which LEDs are preferred over lasers.

Finally, the topics discussed in the remainder of this book – the basics of second-order nonlinear processes, materials for second-harmonic generation and sum-frequency mixing, diode-pumping and intracavity frequency doubling of solid-state lasers, nonlinear frequency conversion in resonators and waveguides, rare-earth spectroscopy and the physics of upconversion lasers, and fundamentals of semiconductor diode lasers – are relevant to the generation of wavelengths other than blue-green ones. There is great interest in compact sources of light in portions of the visible spectrum other than the blue-green, and in the generation of wavelengths longer than $\sim 1\ \mu\text{m}$ using near-infrared semiconductor and diode-pumped solid-state lasers as the starting point. Many of the basic ideas used for upconversion of near-infrared light to shorter wavelengths (particularly those based on nonlinear optics) can also be used for downconversion to longer wavelengths. The fundamentals outlined in the remainder of this book should establish a sound basis for understanding these devices as well.

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Part 1

Blue-green lasers based on nonlinear frequency conversion

2

Fundamentals of nonlinear frequency upconversion

2.1 INTRODUCTION

Blue-green light can be generated by using nonlinear crystals to “upconvert” the infrared wavelengths produced by high-power semiconductor diode lasers. In second-harmonic generation (SHG), a single infrared laser with frequency ω_1 is passed through a nonlinear crystal and blue-green light emerges with frequency $2\omega_1$. In sum-frequency generation (SFG), two infrared lasers with frequencies ω_1 and ω_2 are combined in the crystal; the generated blue-green beam then has frequency $\omega_1 + \omega_2$. These “second-order” nonlinear effects are relatively weak, yet it is still possible to use them to generate blue-green radiation at power levels suitable for the applications described in Chapter 1. In fact, of the three basic approaches to blue-green light generation discussed in this book, nonlinear frequency upconversion has so far been the most extensively developed and the most prolific in spawning commercial blue-green laser products.

The inherent weakness of these nonlinear effects has forced researchers and laser engineers to explore a variety of techniques for enhancing the efficiency of these interactions. In Chapters 3–6, we will discuss these different approaches, which include such things as intracavity frequency-doubling, resonant enhancement, and guided-wave interactions. However, all of these different embodiments exploit the same basic nonlinear interactions, and this chapter is devoted to explaining the essential nature of those processes. In it, we will give a qualitative explanation of the physical process underlying SHG and SFG, we will present some of the basic equations necessary for understanding and designing blue-green lasers based on these effects, we will discuss techniques for providing “phasematching”, which we will see is a crucial requirement for efficient generation of blue-green light, and we will examine some of the nonlinear materials that can be used for frequency conversion of near-infrared light.

Limited space will not permit a full exposition of the principles of nonlinear optics – for that, the interested reader is referred to texts by Boyd (1992) or Shen (1984). In addition, a set of notes intended to “fill in some of the blanks” in the discussion given here may be obtained at www.cup.org. Readers who are already familiar with these matters may wish to skip this chapter and proceed to the discussion of particular embodiments in Chapters 3–6. In the remainder of this chapter, we present a brief sketch of some of the fundamental principles of nonlinear frequency conversion that underlie the embodiments described in subsequent chapters.

2.2 BASIC PRINCIPLES OF SHG AND SFG

2.2.1 The nature of the nonlinear polarization

By what mechanism can an infrared beam passing through a crystal generate blue-green light? Consider the following model, which is simplistic but captures the essential idea behind nonlinear frequency conversion. Suppose that the nonlinear crystal is made up of atoms which we can imagine as comprising a positively charged nucleus surrounded by an electron cloud (Figure 2.1). In this equilibrium condition, the centers of positive and negative charge coincide, and there is no net polarization present in the material. Now suppose a light wave with frequency ω_1 is applied to the material. The electric field associated with that light wave exerts a force upon

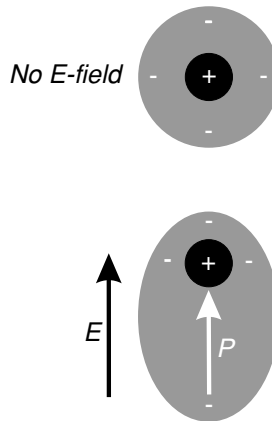


Figure 2.1: Electric polarization in a dense medium can result from the displacement of charge in constituent atoms by an external electric field. The positively charged nucleus is shown in black and the surrounding electron cloud is shown in gray. When no electric field is present, the centers of positive and negative charge coincide and there is no induced polarization. An applied electric field distorts the electron cloud so that the centers of charge are spatially separated and an electric polarization is induced.

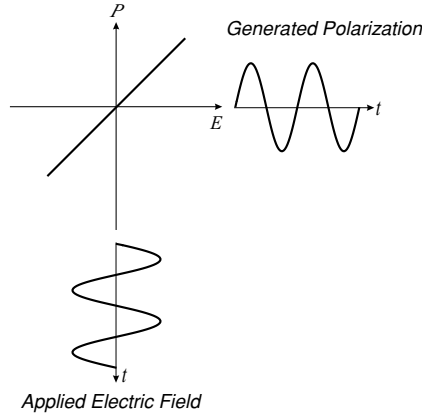


Figure 2.2: For a medium in which the induced polarization is a linear function of the applied electric field, a sinusoidally-varying electric field will induce a polarization that varies sinusoidally at the same frequency.

the electron cloud and distorts it. This distortion causes a spatial separation of the centers of positive and negative charge so that an electric polarization is induced in the material.

As the electron cloud is driven by the time-varying electric field of the light wave – first to one side of the nucleus, then to the other – an oscillating polarization is created. If the relationship between the polarization and applied electric field is linear, the time-varying polarization is also sinusoidal at frequency ω_1 (Figure 2.2). An accelerating charge radiates an electromagnetic wave; thus, this sinusoidally-varying polarization radiates its own electric field at frequency ω_1 , much as the sinusoidally-accelerated charges in an antenna wire do. The electric field radiated by the polarization interferes with the electric field originally present in the material, and this interference leads to the phase shift which we normally describe as being due to the index of refraction of the material (Feynman *et al.*, 1963).

However, if the relationship between the induced polarization P and the electric field E is *not* linear (Figure 2.3), then the generated polarization is not the same for an applied field of magnitude $+E_0$ as it is for an applied field of magnitude $-E_0$. Then, the polarization response to an applied sinusoidal field is not a pure sinusoid, but is distorted. This distortion reflects the presence of components in the polarization response at frequencies other than ω_1 – in the example of Figure 2.3, we can see presence of a strong component at the second-harmonic frequency $2\omega_1$.

We can mathematically describe the functional relationship between the polarization $P(t)$ and the applied electric field $E_a(t)$ depicted in Figure 2.3 by means of a power series expansion:

$$P(t) = f[E_a(t)] = \epsilon_0 \chi^{(1)} E_a(t) + \epsilon_0 \chi^{(2)} [E_a(t)]^2 + \epsilon_0 \chi^{(3)} [E_a(t)]^3 + \cdots \quad (2.1)$$

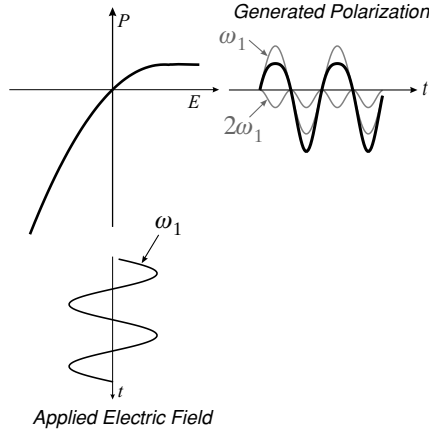


Figure 2.3: For a medium in which the induced polarization is a nonlinear function of the applied electric field, a sinusoidally-varying electric field will induce a polarization that contains frequency components at higher harmonics of the original frequency. In this example, the induced polarization response can be decomposed (gray traces) into a component at the applied frequency and at the second harmonic of the applied frequency.

The first term of this expansion gives rise to first-order (or linear) phenomena, such as the index of refraction. The second-order term, involving the square of the electric field, gives rise to the nonlinear effects in which we are interested: SHG and sum-frequency mixing; it also gives rise to difference-frequency generation, parametric fluorescence, and optical rectification. The third-order term, involving the cube of the electric field, gives rise to effects such as third-harmonic generation, intensity-dependent refractive index, and Brillouin scattering.

2.2.2 Frequencies of the induced polarization

Suppose we apply a monochromatic light wave field $E_a(z, t) = E_1(z, t) = A_1 \cos(\omega_1 t - k_1 z)$ to the nonlinear material. Equation (2.1), becomes

$$\begin{aligned}
 P(z, t) &= f[E_a(z, t)] \\
 &= \epsilon_0 \chi^{(1)} A_1 \cos(\omega_1 t - k_1 z) + \epsilon_0 \chi^{(2)} [A_1 \cos(\omega_1 t - k_1 z)]^2 + \dots \quad (2.2) \\
 &= \epsilon_0 \chi^{(1)} A_1 \cos(\omega_1 t - k_1 z) + \frac{\epsilon_0 \chi^{(2)}}{2} A_1^2 [1 + \cos(2\omega_1 t - 2k_1 z)] + \dots \quad (2.3)
 \end{aligned}$$

In this expansion we can explicitly see the appearance of the dc and second-harmonic terms described above. While this time-domain approach is straightforward with simple monochromatic input fields, it can become cumbersome when more complicated fields are used. Since our concern is with the creation of new

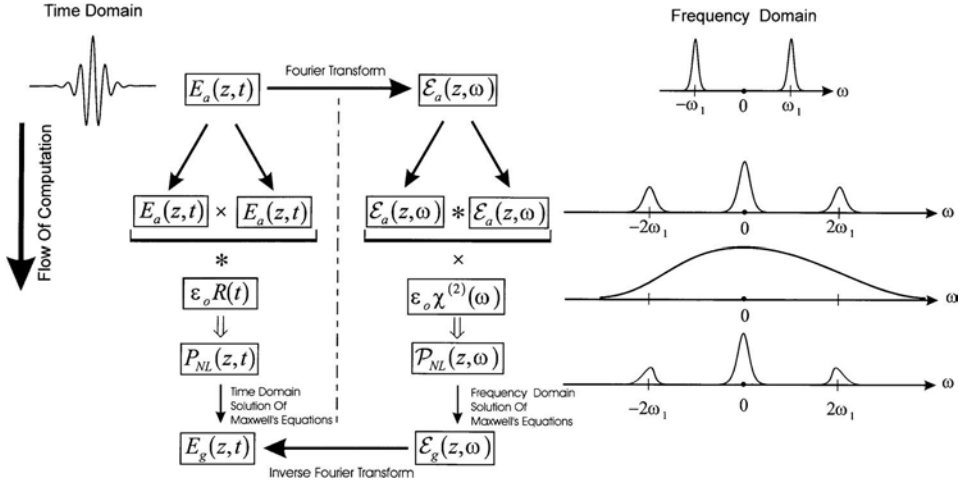


Figure 2.4: The time-domain representation is shown on the left, with the corresponding frequency-domain representation at right. The two domains are linked through the Fourier transform and its inverse. The effect of the nonlinear medium upon the time domain optical pulse shown at the upper right may be considered in either the time domain or the frequency domain.

frequency components, it is perhaps more natural to use a frequency-domain description, which is related to the time-domain description through the Fourier transform (Figure 2.4).¹

If we apply this approach to Equation (2.1), we obtain

$$\mathcal{P}(z, \omega) = \epsilon_0 \chi^{(1)} \mathcal{E}_a(z, \omega) + \frac{\epsilon_0 \chi^{(2)}}{2\pi} [\mathcal{E}_a(z, \omega) * \mathcal{E}_a(z, \omega)] + \dots \quad (2.4)$$

In this expression $\mathcal{P}(z, \omega)$ is the Fourier transform of $P(z, t)$, that is, $\mathcal{P}(z, \omega) = \mathfrak{F}\{P(z, t)\} = \int_{-\infty}^{\infty} P(z, t) e^{-j\omega t} dt$, and similarly $\mathcal{E}_a(z, \omega) = \mathfrak{F}\{E_a(z, t)\}$. In the second term on the right-hand side, $*$ represents convolution.² Clearly, the first term on the right-hand side of Equation (2.4) contains only those frequency components present in the applied optical field. However, we can see that the convolution present in the second term can generate new frequency components. For example, Figure 2.5 shows the result of self-convolving an applied monochromatic field with frequency $\omega = \omega_1$ – clearly, new frequency components are generated at $\omega = 2\omega_1$ and $\omega = 0$, as we learned above.

¹ Reviews of the basic properties of the Fourier transform may be found in Bracewell (1978) and Papoulis (1962).

² We have also used the relationship

$$\mathfrak{F}\{[E_1(z, t)]^2\} = \frac{1}{2\pi} [\mathcal{E}_1(\omega) * \mathcal{E}_1(\omega)] = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathcal{E}_1(\Omega) \mathcal{E}_1(\Omega - \omega) d\Omega$$

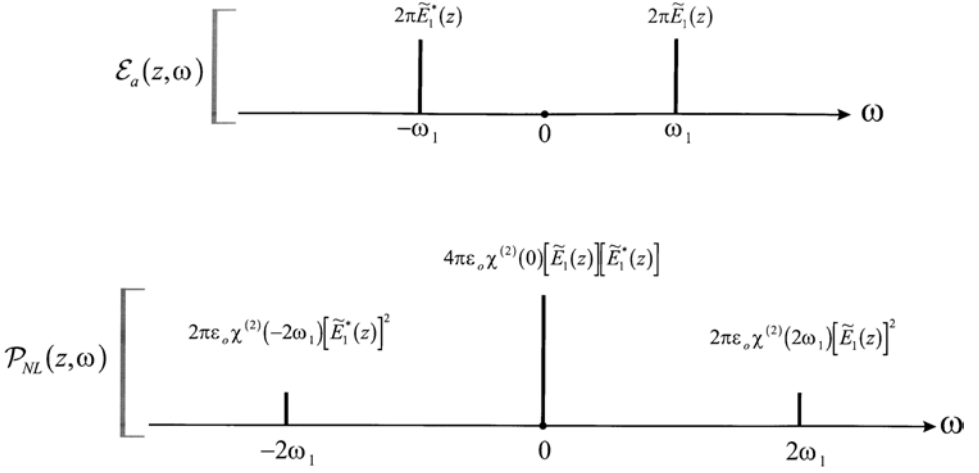


Figure 2.5: Frequency-domain representations for second-harmonic generation of a monochromatic input. The vertical lines represent delta functions.

How large are the polarization components at these new frequencies? The magnitude of the second-order component of the induced polarization, $\mathcal{P}_{NL}$, is related to the self-convolution of the applied field through the *nonlinear susceptibility* $\chi^{(2)}$; however, in general, $\chi^{(2)}$ is not a constant as Equation (2.4) suggests, but is a function of frequency. That is, different frequency components resulting from the self-convolution of $\mathcal{E}_a(z, \omega)$ contribute with different strengths to the generation of nonlinear polarization. Thus, we write $\mathcal{P}_{NL}(z, \omega) = [\epsilon_0 \chi^{(2)}(\omega)/2\pi][\mathcal{E}_a(z, \omega) * \mathcal{E}_a(z, \omega)]$. In a sense, the nonlinear susceptibility behaves in much the same way as a filter in an electrical system, which applies a frequency-dependent amplitude weighting and phase shift to an input signal.

An example will help to crystallize the preceding discussion. We can rewrite the applied field as $E_a(z, t) = E_1(z, t) = A_1 \cos(\omega_1 t - k_1 z) = \frac{1}{2} A_1 e^{-jk_1 z} e^{j\omega_1 t} + \frac{1}{2} A_1 e^{+jk_1 z} e^{-j\omega_1 t} = \tilde{E}_1(z) e^{j\omega_1 t} + \tilde{E}_1^*(z) e^{-j\omega_1 t}$, where $*$ indicates the complex conjugate. The corresponding frequency domain representation is $\mathcal{E}_a(z, \omega) = \mathcal{E}_1(z, \omega) = 2\pi[\tilde{E}_1(z)\delta(\omega - \omega_1) + \tilde{E}_1^*(z)\delta(\omega + \omega_1)]$, where the delta function is the Fourier transform of the exponential.³ By performing the convolution of $\mathcal{E}_a(z, \omega)$ with itself, we obtain $\mathcal{E}_1(z, \omega) * \mathcal{E}_1(z, \omega) = 4\pi^2[\tilde{E}_1^2(z) \cdot \delta(\omega - 2\omega_1) + \tilde{E}_1^{*2}(z) \cdot \delta(\omega + 2\omega_1) + 2\tilde{E}_1(z) \cdot \tilde{E}_1^*(z) \cdot \delta(\omega)]$. Thus, the self-convolution generates polarization components at $\omega = \pm 2\omega_1$, which drive the generation of the second harmonic, and at $\omega = 0$, which drives optical rectification. Multiplication

³ Here we have used

$$\mathfrak{F}\{e^{j\omega_1 t}\} = 2\pi\delta(\omega - \omega_1)$$

by $\epsilon_0 \chi^{(2)}(\omega)/2\pi$ gives

$$\begin{aligned} \mathcal{P}_{NL}(z, \omega) = 2\pi\epsilon_0 & \left[\chi^{(2)}(2\omega_1) \cdot \tilde{E}_1^2(z) \cdot \delta(\omega - 2\omega_1) \right. \\ & + \chi^{(2)}(-2\omega_1) \cdot E_1^{*2}(z) \cdot \delta(\omega + 2\omega_1) \\ & \left. + 2\chi^{(2)}(0) \cdot \tilde{E}_1(z) \cdot \tilde{E}_1^*(z) \cdot \delta(\omega) \right] \end{aligned} \quad (2.5)$$

We see that the magnitude of the polarization term driving SHG generation is proportional to the nonlinear susceptibility $\chi^{(2)}(\pm 2\omega_1)$ and also to the square of the amplitude of the applied electric field. However, we note that the polarization component accounting for optical rectification has a different weighting factor, $\chi^{(2)}(0) \neq \chi^{(2)}(\pm 2\omega_1)$. This difference can be made plausible using the oscillating electron model given earlier. Both SHG and optical rectification result from the application of an electric field with frequency ω_1 , but in the former case, the induced motion of the electron is at an optical frequency ($\omega = 2\omega_1$), while in the latter case, a static offset is created ($\omega = 0$). It is not too hard to believe that the magnitude of these motions could be very different. We can make a similar argument that $\chi^{(2)}$ could depend on the *applied* frequencies, even if the *generated* frequency is the same.

As we have seen, monochromatic fields have delta function frequency representations, and if we know this to be true in advance, it is sometimes convenient to adopt a notation that allows us to keep track of the amplitudes of the relevant delta functions without requiring us to write out the entire delta function representation. Thus we can write Equation (2.5) as:

$$\mathcal{P}_{NL}(z, \omega) = 2\pi \left[\tilde{P}_{NL}^{(2\omega_1)}(z) \delta(\omega - 2\omega_1) + \tilde{P}_{NL}^{(-2\omega_1)}(z) \delta(\omega + 2\omega_1) + \tilde{P}_{NL}^{(0)}(z) \delta(\omega) \right] \quad (2.6)$$

where $\tilde{P}_{NL}^{(2\omega_1)}(z) = \epsilon_0 \cdot \chi^{(2)}(2\omega_1) \cdot \tilde{E}_1^{(\omega_1)^2}$.

As another example, Figure 2.6 shows the frequency components present in the induced polarization for the case of sum-frequency mixing when *two* monochromatic light waves at frequencies ω_1 and ω_2 are applied to the crystal. If we imagine convolving the input spectrum of Figure 2.6 with itself, we obtain the result shown in lower part of that figure. We see that components are present at $\pm 2\omega_1$, $\pm 2\omega_2$, $\pm(\omega_1 + \omega_2)$, $\pm(\omega_2 - \omega_1)$, and 0. The $\pm(\omega_1 + \omega_2)$ frequency component drives the generation of the desired sum-frequency component. The other terms drive difference frequency generation ($\pm(\omega_2 - \omega_1)$), SHG of each of the applied fields ($\pm 2\omega_1$ and $\pm 2\omega_2$), and optical rectification. Using the notation above we can write the amplitude of the sum-frequency component as $\tilde{P}_{NL}^{(\omega_1 + \omega_2)} = 2\epsilon_0 \chi^{(2)}(\omega_1 + \omega_2) \tilde{E}_1^{(\omega_1)} \tilde{E}_2^{(\omega_2)}$.

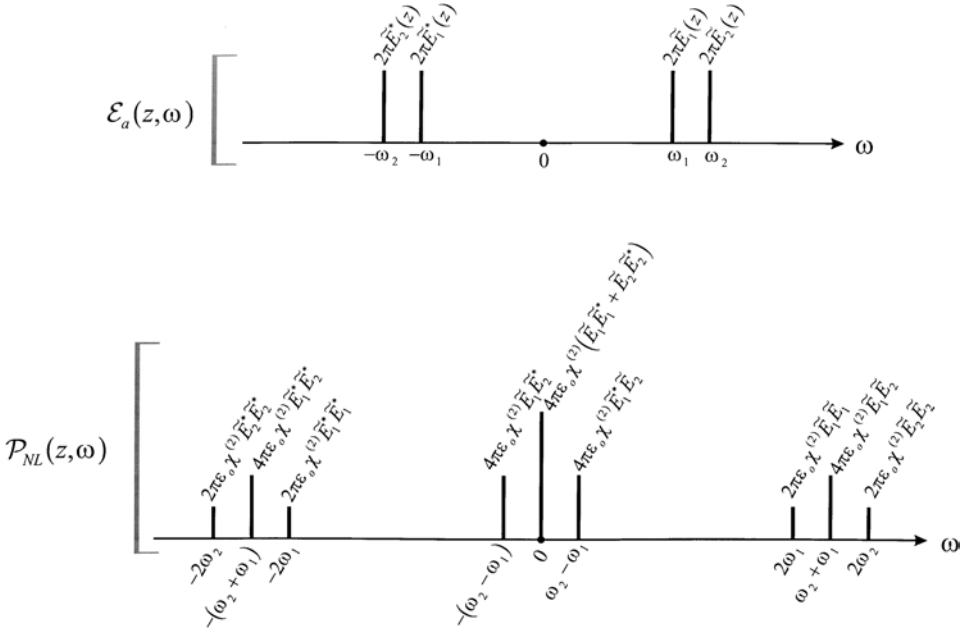


Figure 2.6: Frequency-domain representation for sum-frequency mixing of two monochromatic waves. The vertical lines represent delta functions. The z -dependence has not been explicitly indicated in the lower part of the figure for simplicity. It should be understood that the appropriate value of $\chi^{(2)}$ should be used, as discussed in the text.

In writing this last expression, we have glossed over the fact that both the nonlinear polarization and the applied electric fields are vectors, and are represented by three scalar components in a $x - y - z$ cartesian coordinate system. Thus, each of the three scalar components of the nonlinear polarization can receive contributions from nine possible combinations of the scalar components of applied field. For example, suppose we apply two monochromatic waves to the crystal at frequencies ω_1 and ω_2 (as in Figure 2.6). A particular vector component of the polarization induced at the sum-frequency $\omega_3 = \omega_1 + \omega_2$ – the x -component, for example – can receive contributions from nine separate terms: $\tilde{E}_j^{(\omega_1)} \tilde{E}_k^{(\omega_2)}$, where the index j corresponds to the signal at ω_1 and k corresponds to the signal at ω_2 , and both run over the spatial coordinates x , y , and z . Each of these product terms contributes to $\tilde{P}_{NL,x}^{(\omega_3)}$ through a component of the $\chi^{(2)}$ tensor. Hence, to account for the polarization dependence in the nonlinear interaction, we ought to write something like:

$$\tilde{P}_{NL,i}^{(\omega_2+\omega_1)} = 2\epsilon_0 \sum_{j,k} \chi_{ijk}^{(2)}(\omega_2 + \omega_1) \tilde{E}_j^{(\omega_1)} \tilde{E}_k^{(\omega_2)} \quad (2.7)$$

where i indexes the spatial coordinate of the nonlinear polarization. Thus, in principle, $27 \chi^{(2)}$ components are required to determine $\tilde{P}^{(\omega_2 + \omega_1)}$ from $\tilde{E}^{(\omega_1)}$ and $\tilde{E}^{(\omega_2)}$.

Fortunately, a number of factors reduce the number of independent components of the $\chi^{(2)}$ tensor. A brief review of these factors can be found in the online supplement; a more extended discussion may be found in books by Boyd (1992) or Shen (1984).

2.2.3 The d coefficient

In the technical literature of the blue-green laser field, it is more common to see the “nonlinear coefficient” d specified rather than $\chi^{(2)}$. The two representations are closely related. Consider SHG, in which $\omega_1 = \omega_2$. In this case, the j, k indices (which we previously assigned to the ω_1 and ω_2 signals respectively) indexing $\chi_{ijk}^{(2)}$ are interchangeable, and specifying them both is redundant. Hence, a contracted notation is commonly used, in which the pair of indices j, k is replaced by a single index l according to the following scheme:

$$\begin{array}{cccccc} jk: & 11 & 22 & 33 & 23,32 & 13,31 & 12,21 \\ l: & 1 & 2 & 3 & 4 & 5 & 6 \end{array}$$

We now define $d_{il} = \frac{1}{2} \chi_{ijk}^{(2)}$ using this contracted notation. Thus, for the case of SHG, the cartesian components of the frequency-domain polarization $\tilde{P}_i^{(2\omega_1)}$ are related to the frequency-domain components of the applied electric fields $\tilde{E}^{(\omega_1)}$ by:

$$\begin{bmatrix} \tilde{P}_x^{(2\omega_1)} \\ \tilde{P}_y^{(2\omega_1)} \\ \tilde{P}_z^{(2\omega_1)} \end{bmatrix} = 2\epsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} (\tilde{E}_x^{(\omega_1)})^2 \\ (\tilde{E}_y^{(\omega_1)})^2 \\ (\tilde{E}_z^{(\omega_1)})^2 \\ 2\tilde{E}_y^{(\omega_1)}\tilde{E}_z^{(\omega_1)} \\ 2\tilde{E}_x^{(\omega_1)}\tilde{E}_z^{(\omega_1)} \\ 2\tilde{E}_x^{(\omega_1)}\tilde{E}_y^{(\omega_1)} \end{bmatrix} \quad (2.8)$$

In most cases, this contracted notation can be used for SFG, since frequency dispersion of $\chi^{(2)}$ is small enough that we can still interchange j and k . Thus, for the case of SFG, the cartesian components of the frequency-domain polarization $\tilde{P}_i^{(\omega_2+\omega_1)}$ are related to the frequency-domain components of the applied electric fields $\tilde{E}^{(\omega_1)}$ and $\tilde{E}^{(\omega_2)}$ by:

$$\begin{bmatrix} \tilde{P}_x^{(\omega_2+\omega_1)} \\ \tilde{P}_y^{(\omega_2+\omega_1)} \\ \tilde{P}_z^{(\omega_2+\omega_1)} \end{bmatrix} = 4\epsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} \tilde{E}_x^{(\omega_1)}\tilde{E}_x^{(\omega_2)} \\ \tilde{E}_x^{(\omega_1)}\tilde{E}_y^{(\omega_2)} \\ \tilde{E}_x^{(\omega_1)}\tilde{E}_z^{(\omega_2)} \\ \tilde{E}_y^{(\omega_1)}\tilde{E}_z^{(\omega_2)} + \tilde{E}_z^{(\omega_1)}\tilde{E}_y^{(\omega_2)} \\ \tilde{E}_x^{(\omega_1)}\tilde{E}_z^{(\omega_2)} + \tilde{E}_z^{(\omega_1)}\tilde{E}_x^{(\omega_2)} \\ \tilde{E}_x^{(\omega_1)}\tilde{E}_y^{(\omega_2)} + \tilde{E}_y^{(\omega_1)}\tilde{E}_x^{(\omega_2)} \end{bmatrix} \quad (2.9)$$

The d matrix given here is for the least symmetric crystal class, Class 1. One of the symmetries that reduces the number of independent components is Kleinman symmetry, which says that if all the frequencies involved in the interaction lie sufficiently far away from any resonance frequencies of the nonlinear material, the $\chi_{ijk}^{(2)}$ are not strongly dependent on frequency. Hence, as far as the medium is concerned, the frequencies ω_1 , ω_2 , and $\omega_2 + \omega_1$ are virtually indistinguishable. If this condition is true, it “decouples” all the indices i, j, k associated with the cartesian coordinate system from identification with a specific frequency. Thus, if Kleinman symmetry applies, as is usually the case, the number of independent values in the d matrix reduces from 18 to 10, and the following relationships hold: $d_{14} = d_{25} = d_{36}$; $d_{12} = d_{26}$; $d_{13} = d_{35}$; $d_{15} = d_{31}$; $d_{16} = d_{21}$; $d_{23} = d_{34}$; $d_{24} = d_{32}$.

For crystals that are not in the least symmetric Class 1, some regularity in the arrangement of the crystal causes certain orientations of applied electric fields to be equivalent to others. This spatial symmetry further reduces the number of independent components in the d matrix. The forms and values of the d matrix for the most important materials for blue-green light generation are given in Section 2.5.

Example: Sum-frequency mixing in potassium niobate (KN) Equations (2.8) and (2.9) are crucial for the evaluation of blue-green light generation schemes, and an example may help clarify their use. One approach to blue-green light generation that has been extensively pursued, and which will be discussed in later chapters, is sum-frequency generation in nonlinear materials like potassium titanyl phosphate (KTP) and KN. For example, blue light at 433 nm can be generated by sum-frequency mixing of 982 nm light from an InGaAs laser diode and 775 nm light from an AlGaAs laser diode. In this approach, two electric fields propagating along the x -axis are applied to the KN crystal, one at frequency ω_1 and another at frequency ω_2 , both polarized along the y -axis. KN belongs to crystal class $mm2$, so the corresponding version of Equation (2.9) is:

$$\begin{aligned} \begin{bmatrix} \tilde{P}_x^{(\omega_3)} \\ \tilde{P}_y^{(\omega_3)} \\ \tilde{P}_z^{(\omega_3)} \end{bmatrix} &= 4\epsilon_0 \begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} 0 \\ \tilde{E}_y^{(\omega_1)} \tilde{E}_y^{(\omega_2)} \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix} \\ &= \begin{bmatrix} 0 \\ 0 \\ 4\epsilon_0 d_{32} \tilde{E}_y^{(\omega_1)} \tilde{E}_y^{(\omega_2)} \end{bmatrix} \end{aligned}$$

Hence, the generated polarization is polarized along the z -axis, and the relevant component of the d tensor is d_{32} .

Example: SHG in KN As another example, consider SHG in KN, which has been used to produce 430-nm light from a single 860-nm AlGaAs laser diode. Here, again, propagation is along the x -axis, and the input field at frequency ω_1 is polarized along the y -axis:

$$\begin{aligned} \begin{bmatrix} \tilde{P}_x^{(\omega_3)} \\ \tilde{P}_y^{(\omega_3)} \\ \tilde{P}_z^{(\omega_3)} \end{bmatrix} &= 2\epsilon_0 \begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} 0 \\ [\tilde{E}_y^{(\omega_1)}]^2 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix} \\ &= \begin{bmatrix} 0 \\ 0 \\ 2\epsilon_0 d_{32} [\tilde{E}_y^{(\omega_1)}]^2 \end{bmatrix} \end{aligned}$$

2.2.4 The generated wave

Once we know the nonlinear polarization $\mathcal{P}_{NL}(z, \omega)$, we can determine the sum-frequency or second-harmonic wave radiated by that source using Maxwell's equations. We can show (see the online supplement) that for lossless propagation in the x -direction of a plane wave,⁴ the frequency-domain expression describing the growth of the generated second-harmonic or sum-frequency field $\tilde{\mathcal{E}}_g(x, \omega)$ is given by:⁵

$$\frac{\partial \tilde{\mathcal{E}}_g(x, \omega)}{\partial x} = \frac{\mu_0 \omega^2}{2jk_g(\omega)} \mathcal{P}_{NL}(x, \omega) e^{jk_g(\omega)x} = \frac{-jk_g(\omega)}{2\epsilon_0 n_g^2(\omega)} \mathcal{P}_{NL}(x, \omega) e^{jk_g(\omega)x} \quad (2.10)$$

where μ_0 is the permeability of free space, ϵ_0 is the permittivity of free space, k_g is the propagation constant of the generated wave, n_g is the refractive index for the generated wave. We can integrate over the length l of the crystal to determine the electric field of the generated output wave, $\tilde{\mathcal{E}}_g(l, \omega)$. From that expression, we can determine the power, which is usually the quantity in which we are interested.

⁴ Although we used z as the direction of propagation earlier, here we use x in order to carry on the specific example of SHG and SFG in KN, where propagation is along the crystallographic x -axis.

⁵ Note that $\tilde{\mathcal{E}}_g(z, \omega)$ differs from $\mathcal{E}_g(z, \omega)$ used previously in the specific exclusion of the propagation phase term $e^{-jk_g z}$; that is, $E_g(z, t) = (1/2\pi) \int_{-\infty}^{\infty} \tilde{\mathcal{E}}_g(z, \omega) e^{j[\omega t - k_g(\omega)z]} d\omega$.

Let us apply Equation (2.10) to a specific case of interest here, namely sum-frequency generation with continuous-wave monochromatic waves. (Although SHG is perhaps more common in practice than SFG, the sum-frequency case better illustrates some of the principles involved.) We assume that the input waves for SFG are monochromatic plane waves, $E_1(x, t) = A_1 \cos(\omega_1 t - k_1 x + \phi_1)$ and $E_2(x, t) = A_2 \cos(\omega_2 t - k_2 x + \phi_2)$. We know from our previous discussion that, for monochromatic input waves at frequencies ω_1 and ω_2 , the nonlinear polarization will contain terms at $\pm 2\omega_1$, $\pm 2\omega_2$, $\pm(\omega_1 + \omega_2)$, $\pm(\omega_2 - \omega_1)$, and 0. Thus, the right-hand side of Equation (2.10) will contain a superposition of these terms, and, as in Equation (2.6), each of them can be written in the general form $\tilde{P}^{(\omega_n)}(x)\delta(\omega - \omega_n) + \tilde{P}^{*(\omega_n)}(x)\delta(\omega + \omega_n)$, where for notational convenience we define $\omega_3 = \omega_1 + \omega_2$, $\omega_4 = 2\omega_1$, $\omega_5 = 2\omega_2$, $\omega_6 = \omega_2 - \omega_1$, and $\omega_7 = 0$. These terms were shown in Figure 2.6. We can write Equation (2.10) as:

$$\frac{\partial \tilde{\mathcal{E}}_g(x, \omega)}{\partial x} = \frac{-jk_g(\omega)}{2\epsilon_0 n_g^2(\omega)} \mathcal{P}_{NL}(x, \omega) e^{jk_g(\omega)x} \quad (2.11)$$

$$= \frac{-jk_g(\omega)}{2\epsilon_0 n_g^2(\omega)} \left\{ \sum_{n=3}^7 2\pi [\tilde{P}^{(\omega_n)}(x)\delta(\omega - \omega_n) + \tilde{P}^{*(\omega_n)}(x)\delta(\omega + \omega_n)] \right\} e^{jk_g(\omega)x} \quad (2.12)$$

$$= \frac{\pi}{\epsilon_0} \left\{ \sum_{n=3}^7 \frac{1}{n_{gn}^2} [-jk_n e^{jk_n x} \tilde{P}^{(\omega_n)}(x)\delta(\omega - \omega_n) + jk_n e^{-jk_n x} \tilde{P}^{*(\omega_n)}(x)\delta(\omega + \omega_n)] \right\} \quad (2.13)$$

In writing the last line of this expression, we have anticipated that when we integrate over frequency ω in order to find the power, the delta functions will “pick out” particular values of the continuous functions $k_g(\omega)$ and $n_g^2(\omega)$ corresponding to the discrete frequencies ω_n . To avoid carrying around the unnecessary generality of the functional notation, we simply recognize this fact now and replace $k_g(\omega)$ and $n_g^2(\omega)$ by the appropriate corresponding values $k_n = k_g(\omega_n)$ and $n_{gn}^2 = n_g^2(\omega_n)$.

Each of the complex amplitudes $\tilde{P}^{(\omega_n)}(x)$ appearing in the right-hand side of the equation above has associated with it a phase factor that depends on x . We know how to calculate the values of $\tilde{P}^{(\omega_n)}(x)$ for each of these discrete components from Equation (2.9). Suppose that we are trying to implement sum-frequency mixing in KN, as described in an earlier example. The sum-frequency component of the nonlinear polarization at $\omega_3 = \omega_2 + \omega_1$ will have the form $\tilde{P}_z^{(\omega_3)}(x) = 4\epsilon_0 d_{32} \tilde{E}_y^{(\omega_1)} \tilde{E}_y^{(\omega_2)} = 4\epsilon_0 d_{32} (A_1 A_2 / 4) e^{-j(k_2 + k_1)x} e^{j(\phi_2 + \phi_1)}$. The

second-harmonic component at $\omega_4 = 2\omega_1$ will be $\tilde{P}_z^{(2\omega_1)}(x) = 2\epsilon_0 d_{32} [\tilde{E}_y^{(\omega_1)}]^2 = 2\epsilon_0 d_{32} (A_1^2/4) e^{-j2k_1 x} e^{j2\phi_1}$ and the second-harmonic component at $\omega_5 = 2\omega_2$ will be $\tilde{P}_z^{(2\omega_2)}(x) = 2\epsilon_0 d_{32} [\tilde{E}_y^{(\omega_2)}]^2 = 2\epsilon_0 d_{32} (A_2^2/4) e^{-j2k_2 x} e^{j2\phi_2}$. There are similar terms corresponding to the difference-frequency generation term at ω_6 and the optical rectification term at ω_7 . We can see that each term in the summation in Equation (2.13) will have associated with it a phase term of the form $e^{j\Delta k_n x}$. For example, the contribution at $\omega_3 = \omega_2 + \omega_1$ will contain a term $e^{j(k_3 - k_2 - k_1)x}$, whereas the contribution at $\omega_4 = 2\omega_1$ will contain a term $e^{j(k_4 - 2k_1)x}$. When we perform the integration over x , these contributions will produce terms like

$$\begin{aligned} \int_{x=0}^l e^{(k_3 - k_2 - k_1)x} dx &= l e^{j\frac{1}{2}(k_3 - k_2 - k_1)l} \frac{\sin\left[\frac{1}{2}(k_3 - k_2 - k_1)l\right]}{\frac{1}{2}(k_3 - k_2 - k_1)l} \\ &= l e^{j\frac{1}{2}(k_3 - k_2 - k_1)l} \text{sinc}\left[\frac{1}{2}(k_3 - k_2 - k_1)l\right] \end{aligned} \quad (2.14)$$

for the sum-frequency term at $\omega_3 = \omega_2 + \omega_1$ and

$$\begin{aligned} \int_{x=0}^l e^{(k_4 - 2k_1)x} dx &= l e^{j\frac{(k_4 - 2k_1)l}{2}} \frac{\sin\left[\frac{1}{2}(k_4 - 2k_1)l\right]}{\frac{1}{2}(k_4 - 2k_1)l} \\ &= l e^{j\frac{1}{2}(k_4 - 2k_1)l} \text{sinc}\left[\frac{1}{2}(k_4 - 2k_1)l\right] \end{aligned} \quad (2.15)$$

for the second-harmonic term at $\omega_4 = 2\omega_1$.

The sinc function is plotted in Figure 2.7. We can see that unless $\Delta k \approx 0$, the magnitude of this function will be small. In practice, we arrange by design to make the value of $\Delta k_n = 0$ for the interaction of interest. For example, for SFG, we choose the wavelengths, crystal orientation, and temperature to make $\Delta k_3 = k_3 - k_2 - k_1 = 0$. Usually, all other interactions (corresponding to $\omega_4, \omega_5, \omega_6$, and ω_7) will then have rather large values of $\Delta k_n l$ once we perform the integration over x , and their contribution to the generated field will be negligible. The condition that $\Delta k = 0$ for the interaction of interest is called “phasematching”. Physically, it corresponds to the requirement that, in each section of the crystal, the wave locally generated by the nonlinear polarization must add up in phase with the waves generated by all other sections of the crystal. Achieving phasematching is one of the central problems of practical nonlinear optics and will be discussed further in the following section.

Applying this prescient insight to Equation (2.11), allows us to drop all but the term we intend to phasematch *before* performing the integration, since we know that

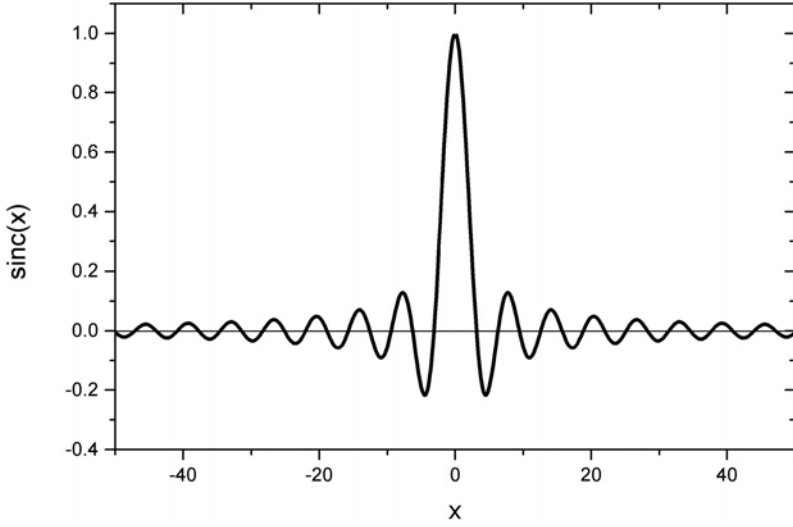


Figure 2.7: The sinc function.

the contributions of the other terms will be small. Thus, Equation (2.11) becomes:

$$\begin{aligned}
 \frac{\partial \tilde{\mathcal{E}}_3(x, \omega)}{\partial x} &= \frac{\pi}{\epsilon_0 n_3^2} \left[-jk_3 e^{jk_3 x} \tilde{P}^{(\omega_3)} \delta(\omega - \omega_3) + jk_3 e^{-jk_3 x} \tilde{P}^{*(\omega_3)} \delta(\omega + \omega_3) \right] \\
 &= \frac{-j\pi k_3 d_{32} A_1 A_2}{n_3^2} \left[e^{j(k_3 - k_2 - k_1)x} e^{j(\phi_2 + \phi_1)} \delta(\omega - \omega_3) \right. \\
 &\quad \left. - e^{-j(k_3 - k_2 - k_1)x} e^{-j(\phi_2 + \phi_1)} \delta(\omega + \omega_3) \right]
 \end{aligned} \tag{2.16}$$

Performing the integration, we obtain:

$$\begin{aligned}
 \tilde{\mathcal{E}}_3(l, \omega) &= \frac{-j\pi k_3 d_{32} A_1 A_2 l}{n_3^2} \left[e^{j\frac{\Delta kl}{2}} \text{sinc}\left(\frac{\Delta kl}{2}\right) e^{j(\phi_2 + \phi_1)} \delta(\omega - \omega_3) \right. \\
 &\quad \left. - e^{-j\frac{\Delta kl}{2}} \text{sinc}\left(\frac{\Delta kl}{2}\right) e^{-j(\phi_2 + \phi_1)} \delta(\omega + \omega_3) \right]
 \end{aligned} \tag{2.17}$$

We now state (without proof, which is provided in the online supplement) that for a wave whose spectral representation is $\tilde{\mathcal{E}}_3(l, \omega) = \tilde{E} \delta(\omega - \omega_0) + \tilde{E}^* \delta(\omega + \omega_0)$, the corresponding power (or intensity) is given by $I_3(l) = |\tilde{E}|^2 / 2\pi^2 \eta$, where $\eta = \sqrt{\mu/\epsilon}$ is the impedance.

Applying this statement to Equation (2.17), we obtain:

$$I_3 = \frac{\left[\frac{k_3 d_{32} A_1 A_2 l}{n_3^2} \text{sinc}\left(\frac{\Delta kl}{2}\right) \right]^2}{2\eta_3} = \frac{8\pi^2 d_{32}^2}{\lambda_3^2 n_1 n_2 n_3 \epsilon_0 c_0} \cdot l^2 \cdot I_1 \cdot I_2 \cdot \text{sinc}^2\left(\frac{\Delta kl}{2}\right) \tag{2.18}$$

2.2.5 SHG with monochromatic waves

The analysis of SHG follows exactly along the lines of that just given for SFG. In this case, we have a single applied field, $E_1(x, t) = A_1 \cos(\omega_1 t - k_1 x + \phi_1)$. The desired component of the induced polarization is $2\pi[\tilde{P}^{(2\omega_1)}\delta(\omega - 2\omega_1) + \tilde{P}^{*(2\omega_1)}\delta(\omega + 2\omega_1)]$, where $\tilde{P}_z^{(2\omega_1)} = 2\epsilon_0 d_{32}[\tilde{E}_y^{(\omega_1)}]^2 = 2\epsilon_0 d_{32}(A_1^2/4)e^{-j2k_1 x}e^{j2\phi_1}$. When we insert this expression into Equation (2.11), we obtain:

$$\begin{aligned} \frac{\partial \tilde{\mathcal{E}}_3(x, \omega)}{\partial x} &= \frac{\pi}{\epsilon_0 n_3^2} \left[-jk_3 e^{jk_3 x} \tilde{P}^{(\omega_3)} \delta(\omega - \omega_3) + jk_3 e^{-jk_3 x} \tilde{P}^{*(\omega_3)} \delta(\omega + \omega_3) \right] \\ &= \frac{-j\pi k_3 d_{32} A_1^2}{2n_3^2} \left[e^{j\Delta k x} e^{2j\phi_1} \delta(\omega - \omega_3) - e^{-j\Delta k x} e^{-2j\phi_1} \delta(\omega + \omega_3) \right] \end{aligned} \quad (2.19)$$

and upon integrating, we have:

$$\begin{aligned} \tilde{\mathcal{E}}_3(l, \omega) &= \frac{-j\pi k_3 d_{32} A_1^2 l}{2n_3^2} \left[e^{j\frac{\Delta k l}{2}} \text{sinc}\left(\frac{\Delta k l}{2}\right) e^{2j\phi_1} \delta(\omega - \omega_3) \right. \\ &\quad \left. - e^{-j\frac{\Delta k l}{2}} \text{sinc}\left(\frac{\Delta k l}{2}\right) e^{-2j\phi_1} \delta(\omega + \omega_3) \right] \end{aligned} \quad (2.20)$$

where, in this case, $\omega_3 = 2\omega_1$, and $\Delta k = k_3 - 2k_1$. Finding the power as we did before, we obtain:

$$I_3(l) = \frac{1}{2\eta_3} \left[\frac{k_3 d_{32} A_1^2 l}{2n_3^2} \right]^2 \text{sinc}^2\left(\frac{\Delta k l}{2}\right) = \frac{2\pi^2 d_{32}^2}{\lambda_3^2 n_3 n_1^2 \epsilon_0 c_0} \cdot l^2 \cdot I_1^2 \cdot \text{sinc}^2\left(\frac{\Delta k l}{2}\right) \quad (2.21)$$

2.2.6 Multi-longitudinal mode sources

So far, we have assumed that the infrared lasers used as input sources for SHG and SFG emit monochromatic waves – that is, waves described by a single sinusoidally-varying component of the form $E_1(x, t) = A_1 \cos(\omega_1 t - k_1 x + \phi_1)$. In practice, this is never truly the case. All lasers emit light over some bandwidth, although in many cases this bandwidth is sufficiently narrow to justify representing the signal as a monochromatic sinusoid. One situation in which this representation is *not* justified is the case of a laser operating in several longitudinal (or *axial*) modes.⁶ This circumstance is often the case when titanium:sapphire (Ti:S), dye lasers, or diode-pumped solid-state lasers are used as the source of infrared light. The question we seek to answer in this section is: “What is the impact on the SHG efficiency

⁶ Another is when the laser is pulsed. SHG with pulsed fields is not covered here, but a discussion may be found in the online supplement. The reader is also referred to Ahkmanov *et al.* (1969) and Glenn (1969).

if the input source emits in several longitudinal modes rather than only one?" We will answer this question by solving Eq. (2.10) for an input source having several longitudinal modes, and comparing the result with the solution we have already obtained for the single-mode (monochromatic) case.

We will begin by considering a specific case, in which the infrared input source has three closely spaced longitudinal modes: $E_0(x, t) = A_0 \cos(\omega_0 t - k_0 x + \phi_0)$, $E_+(x, t) = A_+ \cos[(\omega_0 + \Omega)t - k_+ x + \phi_+]$, and $E_-(x, t) = A_- \cos[(\omega_0 - \Omega)t - k_- x + \phi_-]$. We will assume that the modes are very closely spaced ($\Omega \ll \omega_0$), so that $k_+ \approx k_- \approx k_0$. However, the phases ϕ_+ , ϕ_- , and ϕ_0 of the modes are assumed to be completely independent of one another.

The total intensity of this input infrared source is

$$I_1(0) = \frac{1}{2\eta_1}(A_0^2 + A_+^2 + A_-^2)$$

If all three amplitudes are the same, and we have the same power as in the single-frequency case, we find that we must have $A_0 = A_+ = A_- = A_1/\sqrt{3}$, where A_1 is the amplitude of an "equivalent" single-frequency laser.

We can now calculate the second-harmonic power arising from this input wave. Following the procedure outlined previously, we see that the nonlinear polarization contains five spectral components that can drive the generation of a second-harmonic wave (Figure 2.8).

We assume that these components are sufficiently close together in frequency that the phasematching condition $\Delta k = 0$ is satisfied for all of them simultaneously (which is usually true), and that we can take the frequency and propagation constant to be ω_3 and k_3 , respectively, for all components. We can insert these polarization components into Equation (2.19) in order to determine the generated electric field. From the generated electric field, we find the generated intensity to be:

$$I_3(l) = \left(\frac{k_3 d_{\text{eff}} l}{n_3^2} \right)^2 \times \left\{ \frac{A_1^4}{9} + \frac{4A_1^4}{9} + \frac{A_1^4}{9} [5 + 4 \cos(2\phi_0 - \phi_+ - \phi_-)] + \frac{4A_1^4}{9} + \frac{A_1^4}{9} \right\} \quad (2.22)$$

We now assume that the phases of the longitudinal modes are uncorrelated and may assume any value between 0 and 2π with equal probability. The expectation value of the cosine term involving the phases is then zero, so that we obtain

$$\langle I_3(l) \rangle = \left(\frac{k_3 d_{\text{eff}} l}{n_3^2} \right)^2 \left\{ \frac{5A_1^4}{3} \right\} = \frac{5}{3} \left\{ \frac{2\pi^2 d_{\text{eff}}^2 l^2 I_1^2}{\lambda_3^2 n_3 n_1^2 \epsilon_0 c_0} \right\} \quad (2.23)$$

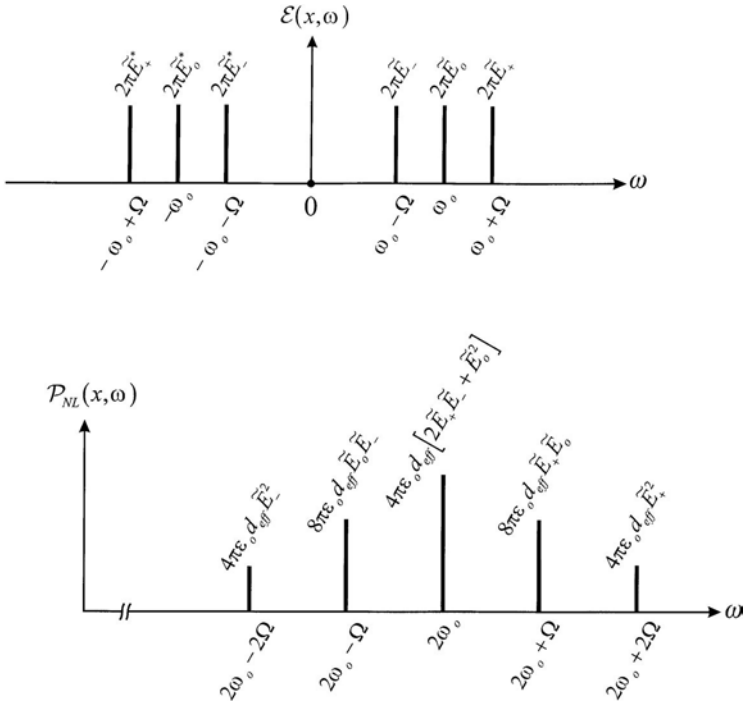


Figure 2.8: Frequency-domain representations of the applied electric field (above) for a source with three closely-spaced longitudinal modes, and the frequency-domain representation of the induced polarization (below). In the lower portion of the figure, only the positive-frequency half of the spectrum is shown. The corresponding negative-frequency components are related through the complex conjugate.

In contrast, if we were frequency-doubling a single-frequency laser, we would have

$$I_3(l) = \left(\frac{k_3 d_{eff} l}{n_3^2} \right)^2 \{A_1^4\} = \left\{ \frac{2\pi^2 d_{eff}^2 l^2 I_1^2}{\lambda_3^2 n_3 n_1^2 \epsilon_0 c_0} \right\} \quad (2.24)$$

Thus, the second-harmonic intensity is a factor of $\frac{5}{3}$ higher using a three-frequency laser than would be obtained using a single-frequency laser with the same power! This increase in power comes about through sum-frequency mixing between the longitudinal modes.

For an input spectrum consisting of N closely-spaced modes, the induced nonlinear polarization will contain $2N - 1$ components, which generate $2N - 1$ spectral components in the radiated second-harmonic wave. It can be shown that the enhancement in the second-harmonic power is given by $2 - 1/N$. Hence, as the number of modes N within the phasematching bandwidth becomes large, the output second-harmonic power can become a factor of 2 larger than what would be generated using a single-frequency laser having the same power.

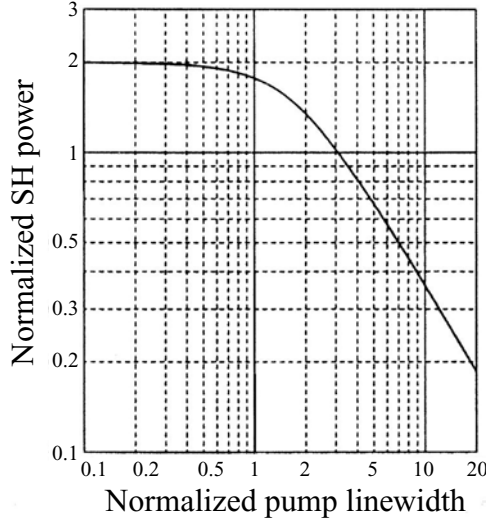


Figure 2.9: Enhancement factor for second-harmonic power generated with a multi-mode pump as a function of pump linewidth. The output power is normalized to be 1 for a corresponding case with a single-mode pump, and the linewidth is normalized by the phasematching bandwidth of the process. (Reprinted with permission from Helmfrid and Arvidsson (1991).)

This effect was observed as early as 1963 in SHG experiments using a $1.15 \mu\text{m}$ He–Ne laser as the infrared source (Ashkin *et al.*, 1963) and was analyzed in detail (Ducuing and Bloembergen, 1964). The intense interest in compact blue-green lasers beginning in the late 1980s led to a renewed concern with this phenomenon. Kwok and Chiu (1985) observed this behavior when generating ultraviolet light by frequency-doubling of a dye laser. Helmfrid and Arvidsson (1991) analyzed the effect for the particular case of quasi-phasematching, and considered more general spectra for the input source. In particular, they analyzed the case of a parabolic power spectral density and considered the dependence of the second-harmonic power on the ratio of the laser linewidth to the phasematching bandwidth for the nonlinear process. Their result is shown in Figure 2.9.

Qu and Singh (1993) investigated this phenomenon experimentally. In their work, they were able to systematically vary the number of longitudinal modes in the spectrum of their fundamental beam from 1 to 7 and observe the effect on the second-harmonic output. Their result closely follows the $2 - 1/N$ behavior for power equally distributed among N modes (Figure 2.10).

Risk *et al.* (1993a) reported an experiment that emphasized the importance of this effect in compact blue-green lasers. They used quasi-phasematched nonlinear waveguides for blue light generation of a single-frequency diode laser using an extended-cavity configuration (described further in Chapter 6). Initially, they

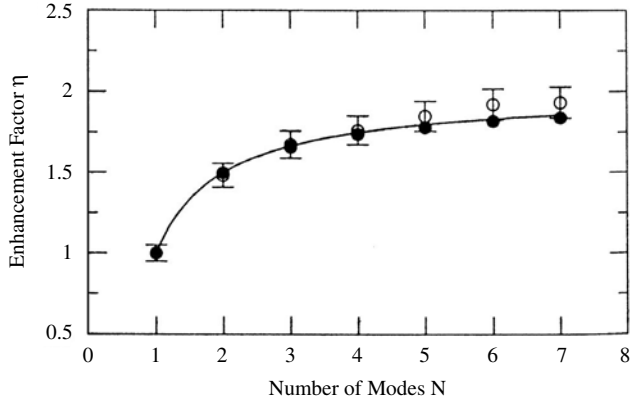


Figure 2.10: Enhancement of the second-harmonic output power for a source having N longitudinal modes compared to the single-mode case ($N = 1$). The solid curve represents the $2 - 1/N$ behavior expected when the power is equally distributed between the N modes, and is meaningful only for integer values of N . Open circles denote experimental points, and solid circles denote the theoretical prediction of a model that takes into account the unequal distribution of power between modes found in a real laser. (Reprinted with permission from Qu and Singh (1993). Copyright: American Physical Society.)

characterized these waveguides using Ti:S lasers, and found that the measured SHG conversion efficiency was 2.75 times greater when a multi-longitudinal-mode Ti:S laser was used than when a single-frequency Ti:S laser was used. (That the difference is larger than a factor of 2 was probably a result of spatial mode differences between the two lasers.) When a single-frequency diode laser was used as the source of infrared light, the efficiency was very close to that measured with the single-frequency Ti:S laser. The moral of this story is that characterization of nonlinear materials and waveguides with multi-frequency Ti:S lasers, which are less expensive and less complex than single-frequency versions, may provide an unduly optimistic estimate of what will be obtained with single-frequency diode lasers unless this effect is taken into account.

2.2.7 Pump depletion

2.2.7.1 SHG

We can rewrite Equation (2.21) for the phasematched case as:

$$\eta_{SHG} = \frac{I_3}{I_1} = \frac{2\pi^2 d_{32}^2}{\lambda_3^2 n_1^2 n_3 \epsilon_0 c_0} \cdot I^2 \cdot I_1 \quad (2.25)$$

where we have introduced the conversion efficiency η_{SHG} . Note that if we make the input power large enough and the length of the nonlinear crystal long enough,

mathematically there is nothing to prevent us from making $\eta_{SHG} > 1$, which means that more energy emerges from the crystal in the second harmonic than we supplied in the fundamental! Of course, this result should offend our *physical* sensibilities, since we believe in the law of conservation of energy, and we must suspect that something in this equation is wrong. The flaw is in our previous assumption that the amplitudes of the input fields A_1 (and A_2 in the SFG case) were constants and did not change as a function of propagation length. But since we are simply *converting* power from ω_1 to $2\omega_1$ – not *creating* it – the build up of power at the second harmonic at $2\omega_1$ *must* eventually begin to deplete the strength of the signal at ω_1 , and A_1 and A_2 *must* be functions of x .

We saw in Section 2.2.2 how to determine the spectral components of the induced polarization by self-convolution of the frequency-domain representation of the electric field present in the crystal. Initially, the only electric field present in the crystal is the applied field at ω_1 , which leads to a polarization component at $2\omega_1$. However, as the amplitude of the second-harmonic wave builds up, we begin to have significant electric fields at both ω_1 and $2\omega_1$ present in the crystal – the situation is now as shown in Figure 2.11. As the amplitude of the second harmonic

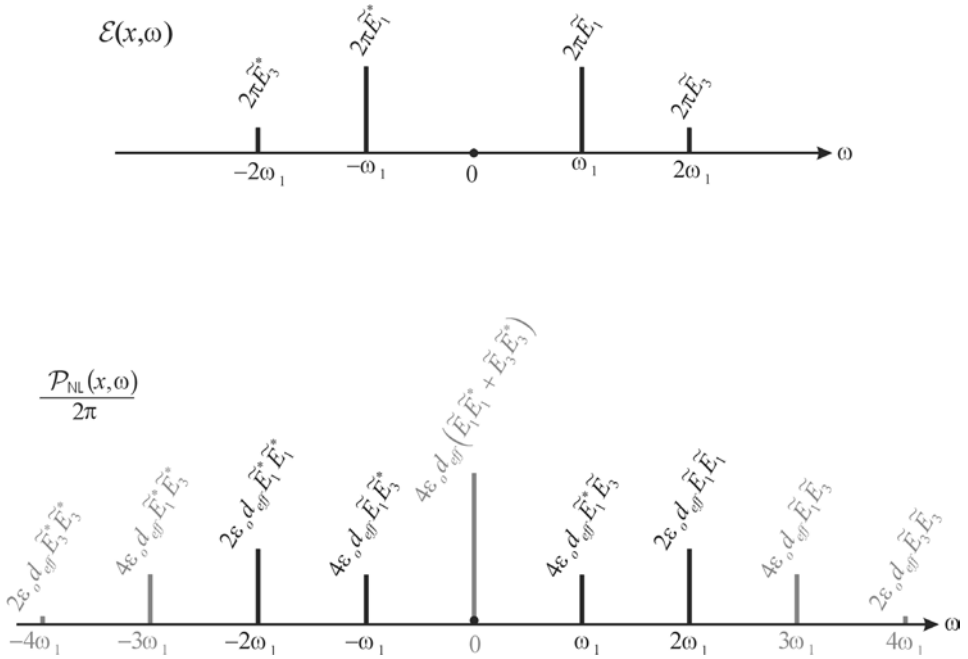


Figure 2.11: Frequency-domain representation of the total electric field once the second harmonic begins to build up (above), and the corresponding frequency-domain representation of the induced polarization. Spectral components irrelevant to the present discussion are shown in gray.

becomes significant, the difference-frequency interaction between the input wave at ω_1 and the generated second-harmonic wave at $2\omega_1$ produces a component in the nonlinear polarization at ω_1 . This polarization component at ω_1 serves to deplete the fundamental.

Thus, we find that not only do we have an equation describing the growth of the second harmonic that depends on the amplitude of the fundamental, as we did previously, but we have a similar equation describing the depletion of the fundamental that depends on the amplitude of the second harmonic. We therefore must solve a set of coupled equations to arrive at our answer. The solution of this case is beyond our scope here (but may be found in the online supplement). In the end, we find that the intensity of the second harmonic grows according to:

$$I_3(l) = I_1(0) \tanh^2(\Gamma l) \quad (2.26)$$

where $\Gamma = A_1(0)\sqrt{\eta_1/\eta_3}$ and $\kappa = k_1 d_{\text{eff}}/n_1^2$. This function is plotted in Figure 2.12. We can see that the conversion efficiency $\eta_{\text{SHG}} = I_3(l)/I_1(0)$ cannot exceed 1, as our physical intuition led us to expect. When the conversion efficiency is low ($\Gamma l \ll 1$), $\tanh^2(\Gamma l) \simeq \Gamma^2 l^2$, and Equation (2.26) becomes identical to Equation (2.25). We can see from Figure 2.12 that the low efficiency approximation is adequate to $\sim 10\%$ conversion efficiency.

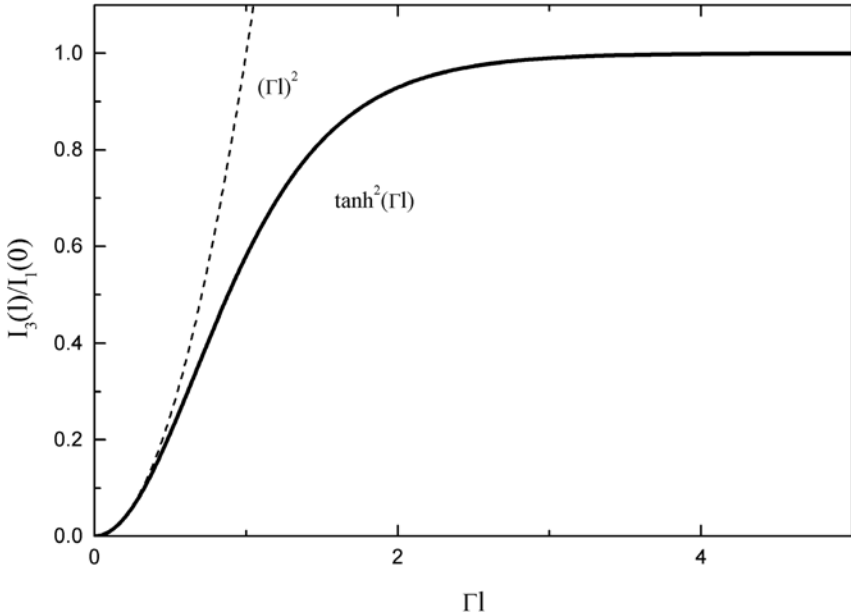


Figure 2.12: Second-harmonic conversion efficiency versus fundamental intensity including the effects of pump depletion. The dashed curve shows the quadratic “no depletion” approximation.

2.2.7.2 SFG

Practical implementations of SFG also provide an example of depletion of one of the interacting waves. Schemes to produce blue light by SFG often mix a very strong beam from a diode-pumped solid-state laser with a weaker beam from a semiconductor laser diode. In the ideal case, all of the power present in the weaker diode laser beam would be converted to blue light. Clearly, this is a situation in which depletion of at least one of the beams involved in the mixing must be taken into account. Figure 2.13 shows the convolution of the frequency spectrum when frequencies ω_1 , ω_2 , and ω_3 are all present, as would be the case in sum-frequency mixing once the sum-frequency builds up to a significant level. We can see that there will be a component in the nonlinear polarization at frequency ω_1 , which arises from difference-frequency mixing of the strong “pump” beam at ω_2 (which we assume remains undepleted) and the sum-frequency beam at ω_3 . Again,

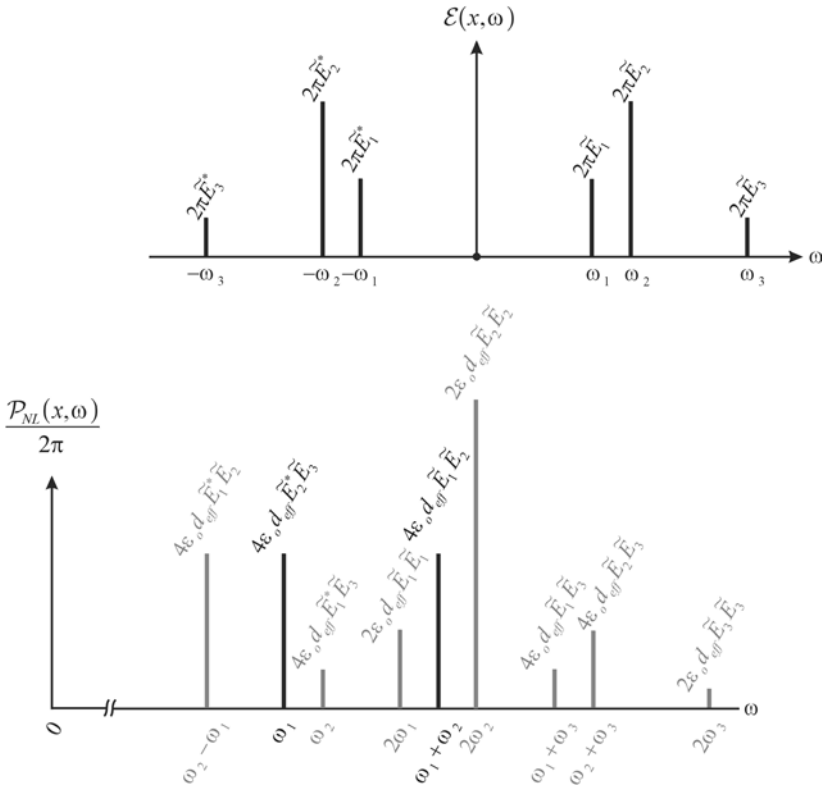


Figure 2.13: Frequency-domain representation of the total electric field present in the nonlinear crystal during SFG (above) and frequency-domain representation of the induced polarization (below). In the lower plot, spectral components irrelevant to the present discussion are shown in gray, and only the positive-frequency components are shown.

we refer the reader to the online supplement for the details, but claim that the intensity of the sum-frequency wave at the end of the nonlinear crystal can be written as:

$$I_3(l) = \frac{\lambda_1}{\lambda_3} I_1(0) \sin^2 \left(\frac{\pi}{2} \frac{l}{l_{max}} \right) \quad (2.27)$$

where we have introduced $l_{max} = \pi/2\sqrt{\kappa_1\kappa_3}$, where $\kappa_1 = k_1 d_{eff} \sqrt{2\eta_2 I_2(0)}/n_1^2$ and $\kappa_3 = k_3 d_{eff} \sqrt{2\eta_2 I_2(0)}/n_3^2$, as the length at which the maximum sum-frequency intensity is produced. Similarly, we can find that

$$I_1(l) = I_1(0) \cos^2 \left(\frac{\pi}{2} \frac{l}{l_{max}} \right) \quad (2.28)$$

The variation of $I_1(x)$ and $I_3(x)$ is shown in Figure 2.14.

A more general solution for SFG involves the doubly periodic Jacobi elliptic function, and is beyond the scope of this text. Discussions of this solution have appeared in the literature (Band *et al.*, 1989).

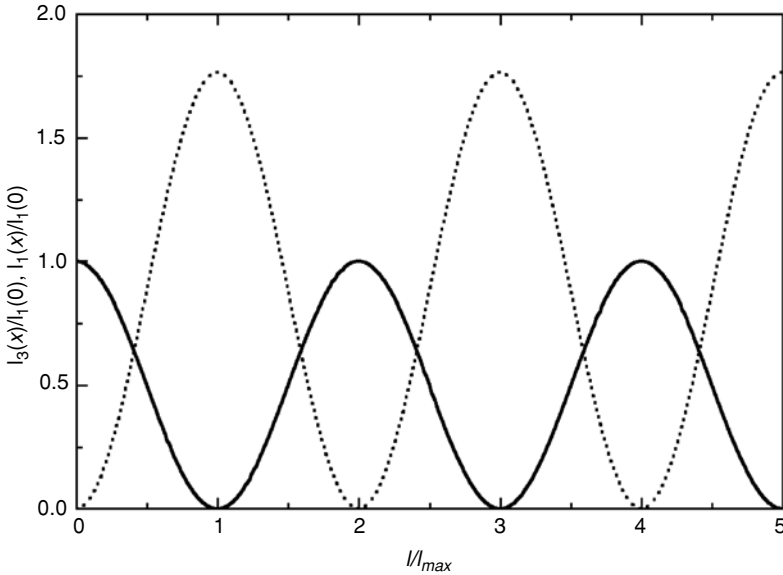


Figure 2.14: Variation of the relative power in the applied wave at ω_1 (solid curve) and generated sum-frequency wave at ω_3 (dashed curve) versus interaction length. For this figure, $\lambda_1 = 809$ nm and $\lambda_3 = 459$ nm were assumed, corresponding to a common mixing process used with KTP. Although it may look like energy conservation is being violated, $I_3(x) > I_1(0)$ because the generated wave also extracts power from the strong pump at ω_2 .

2.3 SPATIAL CONFINEMENT

Equation (2.21) states that for plane waves, the efficiency with which we can convert fundamental to second-harmonic light depends on the square of the intensity of the fundamental beam. In practice, we do not use unbounded plane waves, but rather a beam emitted by a laser that has some finite spatial extent in the plane transverse to the direction of propagation. Intensity is power per unit area; thus, if we have a fixed power to work with, the more tightly confined we can make the beam, the greater we would expect the efficiency to be.

When dealing with beams propagating in free space, there is a catch to this argument. The more tightly we confine the beam, the more rapidly it will diffract. Thus, although we may manage to confine the beam to a small area, we will not be able to maintain that confinement over a very long length. Equation (2.21) also says that the interaction length plays a crucial role in determining conversion efficiency; thus, we are forced to accept a trade-off between tight confinement and long interaction length. We might therefore expect that for a crystal of a given length, there is an optimum focusing condition that represents a compromise between tight confinement and long interaction length. This is indeed the case, as we shall see in Section 2.3.1.

One way to circumvent the compromise inherent in the use of free-space beams is to use a waveguide. The guiding properties of such a structure overcome diffraction; thus, we can confine the beams to a small region over lengths constrained only by loss, substrate size, or the limitations imposed by waveguide fabrication. However, the use of waveguide devices introduces another concern: mode overlap. The modes in a waveguide have well-defined transverse spatial variations that depend on wavelength; thus, the distribution of energy in the fundamental and second-harmonic waves may differ, thereby reducing the conversion efficiency. In Section 2.3.2, we will consider how to calculate the efficiency of a nonlinear upconversion process when guided waves are used.

2.3.1 Boyd–Kleinman analysis for SHG with circular gaussian beams

Boyd and Kleinman (1968) examined the case of Type-I SHG using a focused, circular gaussian beam in a material exhibiting walk-off. The online supplement recites their derivation using SI units, rather than the cgs units used in the original paper. Here we summarize the results so that we may begin using them immediately.

We assume that the fundamental beam is a circular gaussian beam, having an electric field with a radial distribution e^{-r^2/w_0^2} , focused to a waist w_0 at the center of the nonlinear medium. Such a beam can be characterized by its confocal parameter

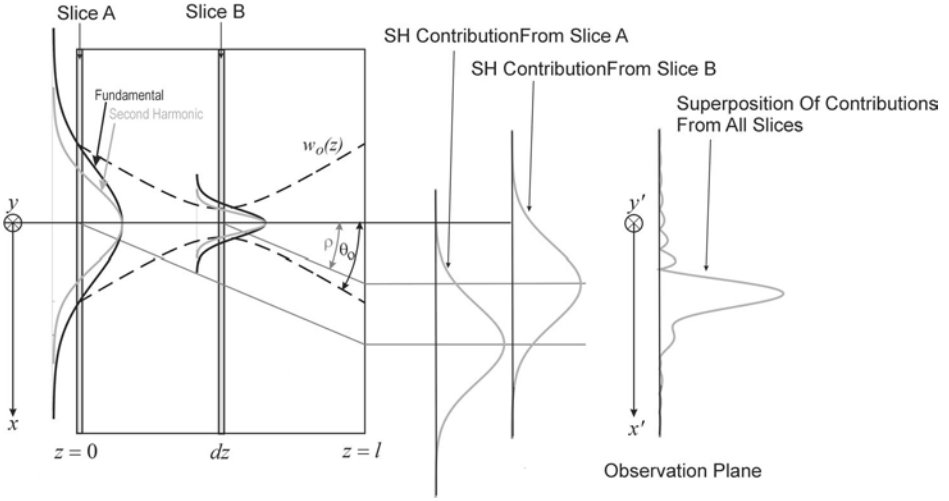


Figure 2.15: Geometry for the Boyd–Kleinman analysis of SHG by a focused gaussian beam.

$b = 2\pi n w_0^2 / \lambda$, where n is the refractive index of the crystal at the fundamental wavelength λ . The angle at which the beam diverges in the far-field is $\theta_0 = \lambda / \pi n w_0$.

The approach underlying the Boyd–Kleinman treatment is illustrated in Figure 2.15. We conceptually divide up the crystal into slices of infinitesimal width. In each slice, the interaction of the fundamental beam with the nonlinear crystal generates second-harmonic light. This second-harmonic light propagates through the crystal to arrive at the “observation plane”. In order to determine the total second-harmonic field produced by the crystal, we must add all the contributions from these infinitesimal slices taking their relative phases into account. Note that because the fundamental beam is focused, the effective area over which the infrared energy is distributed will be different in each slice; thus, the intensity of the blue light generated by each slice will be different.

This seems fairly straightforward; however, a complication arises for anisotropic media. Here we will treat the case of so-called “Type-I” phasematching, in which the fundamental and second-harmonic waves have orthogonal linear polarizations. As we shall see in Section 2.4.2, many advantages accrue if the direction of propagation lies along a crystallographic axis. However, it is very often the case that phasematching cannot be achieved for the wavelengths of interest unless propagation is at some angle to the crystallographic axes. When propagation is *not* along a crystallographic axis, the effect of birefringent walk-off must be taken into account. Thus, as shown in Figure 2.15, the generated second-harmonic wave propagates through the crystal of length l at an angle ρ relative to the fundamental wave. The result of this walk-off is that the second-harmonic contribution from

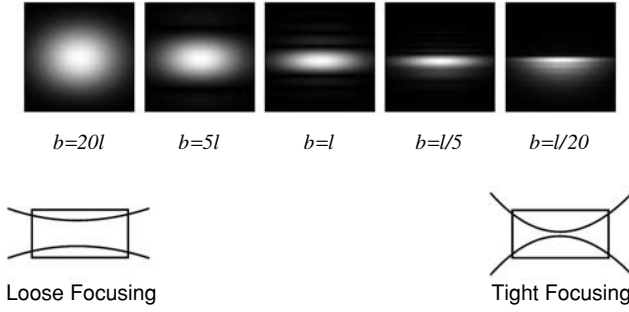


Figure 2.16: Second-harmonic beam profiles calculated using Boyd–Kleinman theory for a 1-cm long crystal with $\rho = 1^\circ$, $\lambda_1 = 1\mu\text{m}$ and $n = 2$.

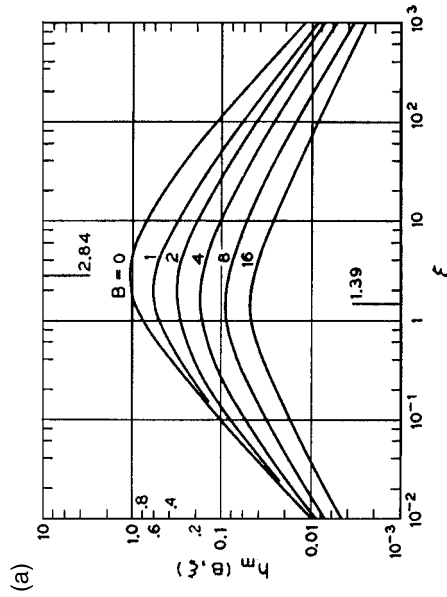
slices near the input end of the crystal are spatially displaced to a greater degree than the second-harmonic contributions arising from near the output end of the crystal. This lateral spatial displacement results in an output second-harmonic beam which no longer has a gaussian distribution.

This distortion of the spatial distribution of the second-harmonic output beam is further illustrated in Figure 2.16. Here, we imagine a nonlinear crystal of fixed length and fixed walk-off angle. Each picture shows the calculated spatial distribution of the output second-harmonic beam, for various degrees of focusing. At the extreme left, very loose focusing is used, while at the extreme right, very tight focusing is used. The distortion of the beam profile is obvious. This effect can be extremely important in applications where the mode quality of the blue beam is a critical factor.

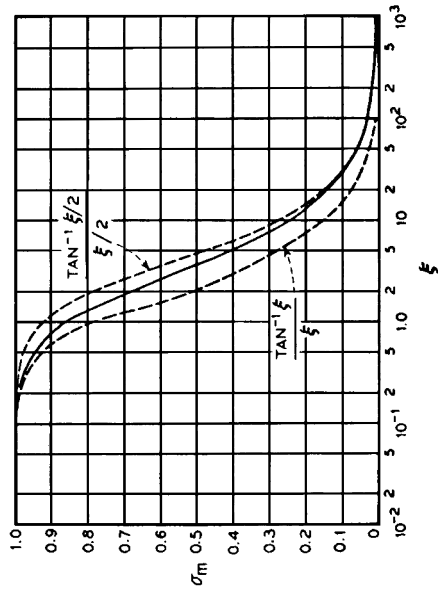
The detailed derivation of SHG with focused beams according to the Boyd–Kleinman approach is given in the online supplement. Here we summarize the key result. The second-harmonic power is given by the expression:

$$P_3 = \frac{16\pi^2 d_{\text{eff}}^2}{\epsilon_0 c \lambda_1^3 n_3 n_1} P_1^2 e^{-\alpha' l} h(\sigma, \beta, \kappa, \xi, \mu) \quad (2.29)$$

In order to determine the second-harmonic power, we need to evaluate $h(\sigma, \beta, \kappa, \xi, \mu)$, the “Boyd–Kleinman focusing factor”. The parameters in the argument of the function are $\kappa = \alpha b/2$, $\sigma = b\Delta k/2$, $\beta = \rho/\theta_0$ and $\xi = l/b$, and $\mu = (l - 2f)/l$, where α is a factor accounting for loss at both wavelengths, and f describes the position of the focus. If we neglect loss and set the focus of the gaussian beam at the center of the crystal, we have $\kappa = \mu = 0$, so that what we must evaluate is $h(\sigma, \beta, 0, \xi, 0)$. In order to determine how h changes as we alter the focusing of the gaussian beam, we would like to hold everything else constant and vary only the confocal parameter b . We define a parameter $B = \rho\sqrt{k_1}/2 = \beta\sqrt{\xi}$, and define the function $h(\sigma, B, \xi) = h(\sigma, B\xi^{-\frac{1}{2}}, 0, \xi, 0)$. B depends only on λ_1 , n_1 , ρ , and l ,



(a)



(b)

Figure 2.17: Plots for various values of B of: (a) h_m versus ξ ; (b) σ_m versus ξ . [Reprinted by permission from G. D. Boyd and D. A. Kleinman, Parametric interaction of focused Gaussian light beams, *J. Appl. Phys.*, **39**, 3597–3639 (1968). Copyright 1968 American Institute of Physics.]

all of which we regard as fixed in a particular experiment. Our task is to determine the maximum value of h , which we can do by picking a value of $\xi = l/b$ (that is, selecting the degree of focusing) and adjusting $\sigma = b\Delta k/2$ to maximize h (that is, adjusting Δk to maximize the second-harmonic output). The value of σ that maximizes h for our fixed values of B and ξ we call $\sigma_m(B, \xi)$ and the maximum value of h we describe as $h_m(B, \xi) = h(\sigma_m, B, \xi) = h[\sigma_m(B, \xi), \beta\sqrt{\xi}, 0, \xi, 0]$.

The calculated values of $h_m(B, \xi)$ are shown in Figure 2.17(a). For each value of B , there is a single optimum value of $\xi = \xi_m$. The quantity we really want to know is the value of h_m at this value of ξ , that is, $h_{mm}(B) = h_m[B, \xi_m(B)]$. As the value of B increases, the optimum value of ξ decreases, meaning that optimum SHG occurs for looser focusing than in the case of no walk-off. Further, the maximum value of h , and hence, the output second-harmonic power decreases rapidly.

For the important case when $B = 0$ (no walk-off), we see that $h_{mm} = 1.068$ and $\xi_m = 2.84$. This translates to the focusing condition $b = l/2.84$; that is, the confocal parameter should be a factor of 2.84 shorter than the crystal length. Thus, for optimally-focused SHG, we have:

$$P_3 = 1.068 \frac{16\pi^2 d_{\text{eff}}^2}{\epsilon_0 c \lambda_1^3 n_3 n_1} P_1^2 l \quad (2.30)$$

Figure 2.17(b) shows how the maximum value of σ varies with ξ in the case $B = 0$. We see that for all values of ξ , $\sigma_m > 0$, which means that we must have $\Delta k > 0$. This result runs counter to our instinct that SHG is most efficient when $\Delta k = 0$; however, this intuition was based on plane waves, and is not accurate for focused beams, which can be thought of as containing plane waves having a range of propagation angles. Thus, when $\Delta k > 0$, phasematching of sum-frequency mixing processes involving these off-axis rays is possible, while this is not the case if $\Delta k < 0$. Since we usually calculate expected phasematching wavelengths based on $\Delta k = 0$, we may find that the experimentally-observed phasematching wavelength is somewhat longer when using a focused beam.

A number of approximations are possible that describe the behavior of Figure 2.17(a) in certain regions. For example, in the case of loose focusing, $\xi \ll 1$, $h_m(B, \xi) \rightarrow \xi$. In this case, Equation (2.29) reduces to

$$P_3 = \frac{8\pi d_{\text{eff}}^2}{\epsilon_0 c \lambda_1^2 n_3 n_1^2 w_0^2} P_1^2 l^2 \quad (2.31)$$

This expression is equivalent to Equation (2.21) when rewritten in terms of intensities.

Example: Blue light at 430 nm can be generated by noncritically phasematched SHG of 860 nm light in KN at room temperature. A consequence (and highly

desirable feature) of noncritical phasematching is that there is no walk-off, i.e., $B = 0$. Suppose we wish to work with a 10-mm long crystal. We should set the focusing of our 860-nm beam such that $b = l/2.84$, which translates to

$$\begin{aligned} w_0 &= \sqrt{\frac{\lambda_1 l}{2\pi n_1(2.84)}} = \sqrt{\frac{(860 \times 10^{-9} \text{ m})(10 \times 10^{-3} \text{ m})}{2\pi(2.2791)(2.84)}} \\ &= 1.45 \times 10^{-5} \text{ m} = 14.5 \text{ } \mu\text{m} \end{aligned}$$

Suppose our 860-nm source is capable of providing 100 mW in a single longitudinal mode. Then we would expect an output power, neglecting loss, of:

$$P_3 = 1.068 \frac{16\pi^2 d_{\text{eff}}^2}{\epsilon_0 c \lambda_1^3 n_3 n_1} P_1^2 l \quad (2.32)$$

$$\begin{aligned} &= 1.068 \frac{16\pi^2 (21 \times 10^{-12} \text{ m/V})^2}{(8.85 \times 10^{-12} \text{ F/m})(3 \times 10^8 \text{ m/s})(860 \times 10^{-9} \text{ m})^3 (2.2791)^2} \\ &\quad \times (10^{-1} \text{ W})^2 (10 \times 10^{-3} \text{ m}) \end{aligned} \quad (2.33)$$

$$= 0.85 \text{ mW} \quad (2.34)$$

2.3.1.1 Experimental verification of Boyd–Kleinman theory

Does Boyd–Kleinman theory really work? Does it accurately describe the effect of focusing on SHG efficiency? An early investigation of this question used lithium niobate for noncritically phasematched SHG of the 1084-nm radiation from a He–Ne laser (McGeoch and Smith, 1970). The experimenters took care to ensure that the spatial distribution of the light emitted from the laser was very close to the gaussian ideal and they used a crystal of high optical quality. The lithium niobate crystal was held in a temperature-controlled oven in order to achieve phase-matching, which was fine tuned by applying an electric field to the crystal (Soffer and Winer, 1967). McGeoch and Smith used a series of different lenses in order to change the focusing of the He–Ne laser beam in the lithium niobate crystal, and obtained the result shown in Figure 2.18(a). The experimentally-observed dependence of conversion efficiency upon focusing closely follows the prediction of Boyd–Kleinman theory.

McGeoch and Smith also investigated the phasematching condition for focused SHG and experimentally verified that optimum SHG occurs for $\Delta k > 0$ (Figure 2.18(b)), as was also verified by Kleinman and Miller (1966), who also investigated the dependence of SHG efficiency on the location of the focus.

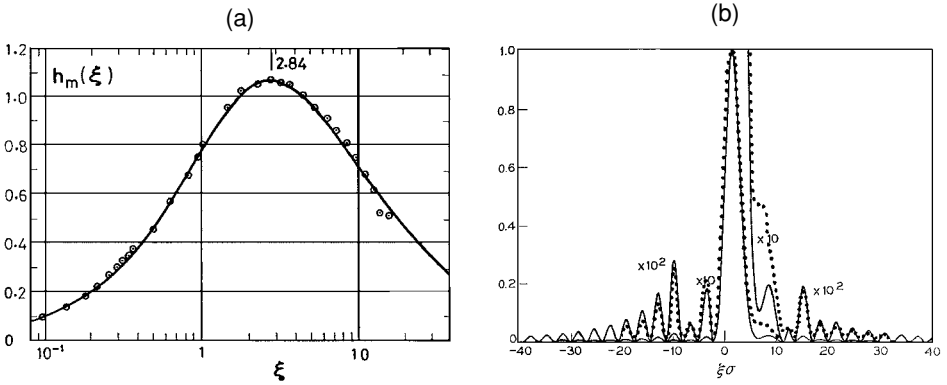


Figure 2.18: Experimental verification of Boyd–Kleinman focusing theory: (a) variation of conversion efficiency with focusing; (b) variation of second-harmonic power with phase mismatch for $\xi = 2.8$. (Reprinted by permission from McGeoch and Smith (1970).)

2.3.1.2 Extensions of Boyd–Kleinman theory

The analysis given by Boyd and Kleinman applies to so-called “Type-I” SHG using circular gaussian beams in either positive or negative uniaxial materials. In Type-I phasematching, both fundamental photons have the same polarization, which is orthogonal to the polarization of the second-harmonic photon. In many important materials, particularly KTP, a “Type-II” process is often used, in which the two fundamental photons have orthogonal polarizations. In this case, both the second-harmonic and one component of the fundamental may experience walk-off. This case has been analyzed by Zondy (1991) and Asaumi (1992).

Although many lasers produce circular gaussian beams, some produce beams with an elliptical gaussian shape. This is especially true of diode laser beams, which are often also astigmatic (that is, the waist occurs at different locations in each of the two planes containing the direction of propagation and an axis of the ellipse). In addition, it has been found that for SHG schemes involving crystals having large walk-off angles, such as BBO (beta barium borate), an increase in second-harmonic power can be achieved by deliberately using an elliptical beam. For example, when BBO is used for frequency-doubling of the 514.5-nm line of an argon-ion laser, the second-harmonic output power can be increased by three orders of magnitude if an elliptical beam is used instead of a circular one (Figure 2.19) (Taira, 1992). For further information on this subject, the interested reader is referred to the following papers: Librecht and Simons (1975), Steinbach *et al.* (1996), and Freearge *et al.* (1997).

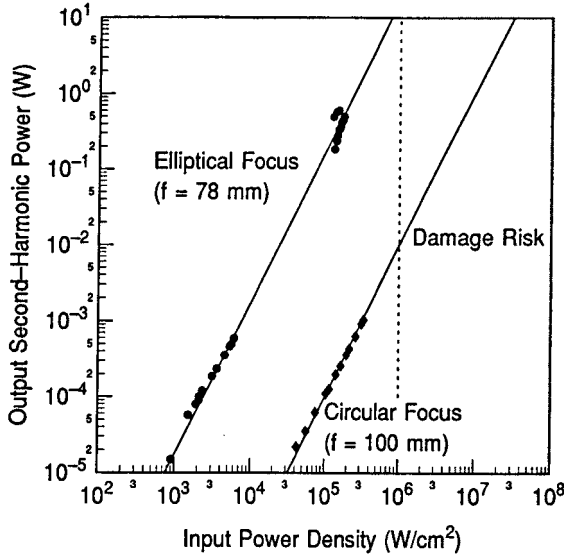


Figure 2.19: The advantages of elliptical focusing in certain cases are illustrated by this example showing intracavity SHG of an argon-ion laser with a BBO crystal. (Reprinted by permission from Taira (1992).)

2.3.1.3 SFG

The analysis of sum-frequency generation with gaussian beams is complicated by the fact that two distinct input beams are involved, which could have different confocal parameters, different foci, different positions in the crystal, and different directions of propagation. However, Boyd and Kleinman's analysis can be readily generalized to a special case of SFG in which the beams have the same confocal parameter b , have waists in the same location, and lie along the same axis. The result is a formula that can be used to obtain at least an initial estimate of the blue-green power produced by SFG:

$$P_3 = \frac{32\pi^2 d_{\text{eff}}^2}{\epsilon_0 n_1 n_2 n_3 \lambda_3^2} P_1 P_2 \frac{lh(\sigma, \beta, \kappa, \xi, \mu)}{\frac{\lambda_1}{n_1} + \frac{\lambda_2}{n_2}} = \frac{32\pi^2 d_{\text{eff}}^2}{\epsilon_0 c n_3^2 \lambda_1 \lambda_2 \lambda_3} P_1 P_2 lh(\sigma, \beta, \kappa, \xi, \mu) \quad (2.35)$$

In the special case $B = 0$ (no walk-off), it can be shown that the SFG efficiency is maximized when the confocal parameter for each beam equals the crystal length divided by 2.84, as in the case of SHG. For $B \neq 0$, a numerical analysis shows that, in general, the efficiency is maximized when the confocal parameters have different values (Guha and Falk (1980)).

2.3.2 Guided-wave SHG

In the preceding section, we have seen that when we use focused gaussian beams for SHG or SFG, we are forced to accept a trade-off between achieving a small spot size and maintaining a long interaction length, both of which are desirable for an efficient nonlinear interaction. This trade-off arises because the smaller we make the focused spot of the beam, the more rapidly it diffracts, thus limiting the length over which that tight confinement can be preserved.

A guided-wave geometry eliminates this trade-off by decoupling the confinement from the interaction length. The tendency of a tightly confined beam to diffract is overcome by the guiding properties of the structure. Thus, by using a waveguide interaction it is possible to maintain a tight confinement over a long length, limited only by the loss or nonuniformity of the waveguide.

There are a number of important conceptual differences between waveguide nonlinear interactions and the gaussian beam interactions described previously. In the gaussian beam case, the spatial distribution of the fundamental beam is continuously variable in the sense that the beam can be focused by an appropriate lens to essentially any spot size desired. In contrast, for a waveguide of a given geometry, the spatial distribution of the electric field has a very specific form, corresponding to a particular “mode” of the waveguide, which is a solution of Maxwell’s equations for the guided-wave structure. At the fundamental wavelength, the guiding structure will support certain modes with specific spatial distributions; similarly, at the second harmonic, the waveguide will support certain modes with specific spatial distributions. If the fundamental wave is confined to one of the modes of the waveguide whose spatial distribution is given by $\mathcal{E}_1(x, y)$, the induced polarization will have a form like $[\mathcal{E}_1(x, y)]^2$, but it can only generate light in a second-harmonic mode whose spatial distribution is given by $\mathcal{E}_3(x, y)$ (Figure 2.20). The efficiency with which a polarization having a spatial

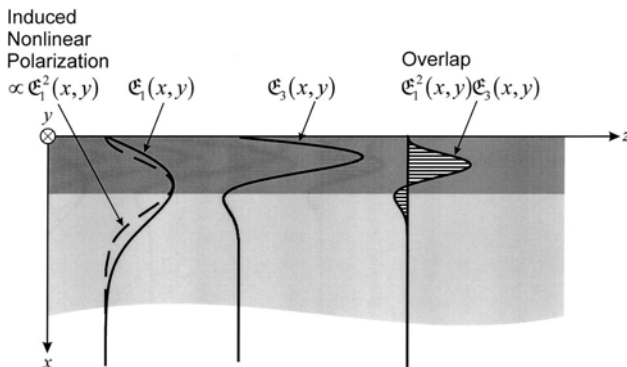


Figure 2.20: Illustration of the mode overlap problem in waveguide SHG.

distribution like $[\mathfrak{E}_1(x, y)]^2$ can excite a second-harmonic wave with a distribution like $\mathfrak{E}_3(x, y)$ is given by an overlap integral between these two functions, as we shall see shortly.

In a bulk medium, we describe the propagation of a wave using the wave vector $k = 2\pi n/\lambda$, which depends only on the wavelength and the refractive index n of the medium. In a waveguide, we must use the corresponding propagation constant β , which is obtained by solving Maxwell's equations for the guided-wave structure and depends on the details of the waveguide geometry and composition. However, in analogy with the case of bulk material, we can write $\beta = 2\pi N_{\text{eff}}/\lambda$, where we call N_{eff} the “effective index” of the guided mode.

How do we calculate the second-harmonic power generated by a guided-wave interaction? We adopt an approach that closely follows that of Regener and Sohler (1988). We can write the electric field for the fundamental waveguide mode as:

$$E_1(x, y, z, t) = A_1(z)\mathfrak{E}_1(x, y)\cos(\omega_1 t - \beta_1 z + \phi_1) \quad (2.36)$$

and the second harmonic as

$$E_3(x, y, z, t) = A_3(z)\mathfrak{E}_3(x, y)\cos(\omega_3 t - \beta_3 z + \phi_3) \quad (2.37)$$

where we take z as the direction of propagation and x and y as the transverse coordinates. Here, $\mathfrak{E}_1(x, y)$ and $\mathfrak{E}_3(x, y)$ are the transverse field distributions of the guided modes and β_1 and β_3 are the propagation constants of those modes.

In order to evaluate the power carried by each mode, we must integrate over the transverse x, y coordinates:

$$P_1 = \frac{A_1^2}{2\eta_1} \iint_{-\infty}^{\infty} \mathfrak{E}_1^2(x, y) dx dy \quad (2.38)$$

$$P_3 = \frac{A_3^2}{2\eta_3} \iint_{-\infty}^{\infty} \mathfrak{E}_3^2(x, y) dx dy \quad (2.39)$$

where now $\eta_1 = 1/\epsilon_0 c N_{\text{eff},1}$ and similarly for η_3 . Assuming that phasematching has been achieved (i.e., $\beta_3 = 2\beta_1$), the generated second-harmonic power can be expressed as:

$$P_3 = P_1 \tanh^2(\Gamma l) \quad (2.40)$$

The quantity Γ is given by:

$$\Gamma = \left(\frac{8\pi^2 d_{\text{eff}}^2 \mathfrak{J}_{ov}^2}{\lambda_1^2 \epsilon_0 c N_{\text{eff}}^3} P_1 \right)^{\frac{1}{2}} \quad (2.41)$$

where the “overlap integral,” $\mathcal{I}_{ov}$, is given by:

$$\mathcal{I}_{ov} = \frac{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathfrak{E}_3(x, y) \mathfrak{E}_1^2(x, y) dx dy}{\left[\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathfrak{E}_3^2(x, y) dx dy \right]^{\frac{1}{2}} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathfrak{E}_1^2(x, y) dx dy} \quad (2.42)$$

In the low-conversion efficiency approximation, we have:

$$P_3 \approx P_1 \Gamma^2 l^2 = \frac{8\pi^2 d_{eff}^2 \mathcal{I}_{ov}^2}{\lambda_1^2 \epsilon_0 c N_{eff}^3} P_1^2 l^2 \quad (2.43)$$

(As an exercise, the interested reader may wish to show that if the field distributions $\mathfrak{E}_1(x, y)$ and $\mathfrak{E}_3(x, y)$ are taken to be gaussians, Equation (2.43) reduces to Equation (2.31), the result for a loosely-focused gaussian in bulk material.) It is also sometimes useful to express the overlap between modes as an effective area, $A_{eff} = 1/\mathcal{I}_{ov}^2$. If nonnegligible losses are present in the system, Equation (2.43) becomes:

$$P_3 = \frac{8\pi^2 d_{eff}^2 \mathcal{I}_{ov}^2}{\lambda_1^2 \epsilon_0 c N_{eff}^3} P_1^2(z=0) \left(\frac{e^{-\alpha_1 l} - e^{-\frac{\alpha_3 l}{2}}}{\alpha_1 - \frac{\alpha_3}{2}} \right)^2 \quad (2.44)$$

where α_1, α_3 are the attenuation coefficients at the fundamental and second harmonic, respectively.

Equation (2.44) makes clear the importance of achieving low loss in waveguide SHG. The potential benefit of waveguide SHG lies in the possibility of achieving tight confinement over a longer length than would be possible using focused gaussian beams. However, if loss limits the effective interaction length by substantially reducing the intensities of the interacting waves, this benefit may not be realized. We can define a loss-limited effective interaction length as:

$$l_{eff} = \frac{e^{-\alpha_1 l} - e^{-\frac{\alpha_3 l}{2}}}{\alpha_1 - \frac{\alpha_3}{2}} \quad (2.45)$$

Figure 2.21 illustrates the deleterious effect of loss on conversion efficiency for waveguide SHG, and the potential increase in efficiency over a bulk interaction. If losses are negligibly small, $l_{eff} \approx l$, and Equation (2.44) reduces to Equation (2.31).

In the preceding discussion, we have implicitly assumed that the effective nonlinear coefficient d_{eff} does not vary over the x - y cross-section of the waveguide. However, this may not always be true, and we may be forced to rewrite

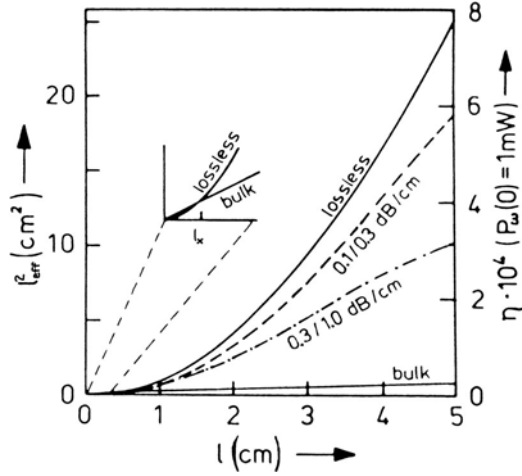


Figure 2.21: The effect of waveguide loss on SHG efficiency. On the left, the square of the loss-limited effective length is shown, and on the right conversion efficiency is given. The inset shows that there is a “cross-over length” l_x at which the efficiency for a lossless guide begins to exceed that from the bulk interaction. This graph assumes a fundamental power of 1 mW, an effective interaction area of 260 mm² and parameters appropriate to Ti:LiNbO₃ waveguides. When losses are given, the first number is α_1 , the loss for the fundamental, and the second is α_3 , the loss for the second harmonic. (Reprinted with permission from Regener and Sohler (1988).)

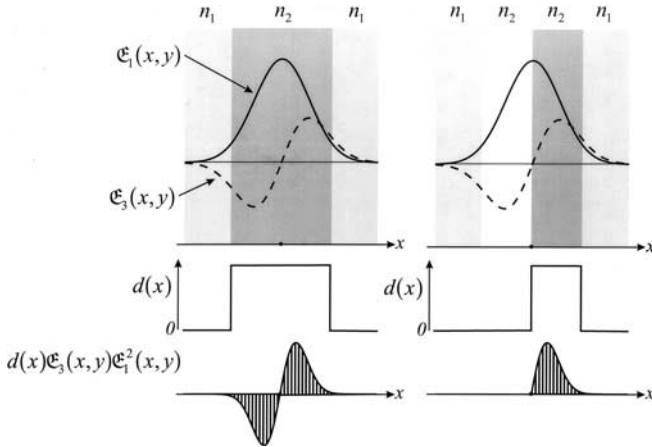


Figure 2.22: The spatial variation of the nonlinear coefficient is particularly important in multi-layer waveguides, where it can be used to advantage to suppress SHG in regions where the polarization reverses phase. In this example, the lowest-order mode at ω_1 is coupled to the first-order mode at ω_3 . On the left is a three-layer waveguide consisting of a central nonlinear core surrounded by linear cladding material of lower index ($n_1 < n_2$). Because of the sign reversal in $\mathcal{E}_3$, the overlap is zero. On the right is a four-layer waveguide in which the left half of the nonlinear material has been replaced by a linear material with the same refractive index. This substitution eliminates the contribution of the out-of-phase lobe of $\mathcal{E}_3$ and produces a nonzero overlap.

Equation (2.42) as:

$$P_3 = \frac{8\pi^2}{\lambda_1^2 \epsilon_0 c N_{eff}^3} P_1^2 l^2 \left\{ \frac{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d_{eff}(x, y) \mathfrak{E}_3(x, y) \mathfrak{E}_1^2(x, y) dx dy}{\left[\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathfrak{E}_3^2(x, y) dx dy \right]^{\frac{1}{2}} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathfrak{E}_1^2(x, y) dx dy} \right\}^2 \quad (2.46)$$

There are at least two kinds of practical situations in which the variation of d_{eff} over the waveguide cross-section must be taken into account. The first is when the waveguide is composed of layers of various materials which intrinsically have different nonlinearities. An example is shown in Figure 2.22. As we will discuss further in Section 2.4.4, waveguide phasematching can sometimes result in the excitation of a higher-order mode at the second harmonic. The overlap between this higher-order mode and the fundamental in the lowest-order mode is often rather small, because the higher-order mode has both positive and negative lobes, which produce contributions that tend to cancel in the overlap integral. However, by

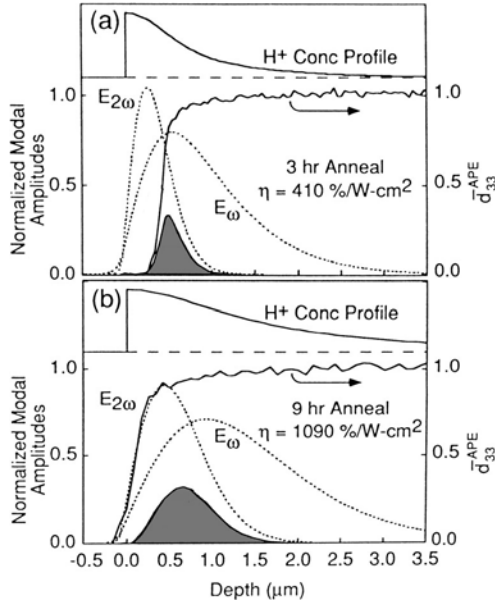


Figure 2.23: Overlap of fundamental and second-harmonic modes (dashed lines) with the spatially-varying nonlinear coefficient (solid line) in LN waveguides fabricated by annealed proton exchange. The shaded region shows the product $d_{eff} \mathfrak{E}_3 \mathfrak{E}_1^2$. The difference between (a) and (b) is the annealing time, showing how adjustment of processing conditions can improve the conversion efficiency by increasing the overlap. (Reprinted with permission from Bortz *et al.* (1993). Copyright: American Institute of Physics.)

adjusting the spatial distribution of the nonlinear coefficient, it is sometimes possible to eliminate one of these portions and increase the overlap (Ito and Inabe, 1978).

Another situation in which a spatially-varying nonlinearity often arises is in waveguides fabricated in nonlinear materials using processes like ion exchange and in-diffusion in which some substance is introduced into the material to locally raise the refractive index. The introduction of this material may also change the nonlinear coefficient. The problem has been a particular concern in the fabrication of waveguides in lithium niobate (LN) by proton exchange. Replacement of lithium ions by hydrogen ions creates the centrosymmetric compound HNbO_3 , which has no intrinsic nonlinearity. Some of the nonlinearity can be regained by annealing the waveguide. Figure 2.23 shows the measurements of Bortz and coworkers of the spatially-varying d_{eff} in such waveguides and the influence of annealing on the overlap.

2.4 PHASEMATCHING

2.4.1 Introduction

In Section 2.2.4, we saw that the condition $\Delta k \approx 0$ must be satisfied in order for SHG or SFG to become efficient.

For collinear SHG, the phasematching condition $\Delta k = 0$ becomes $n_3 - n_1 = 0$. Since the index of refraction is a measure of the speed (phase velocity) with which the waves travel through the medium, this condition says that the fundamental wave at frequency ω_1 and the second-harmonic wave at frequency $\omega_3 = 2\omega_1$ must travel through the nonlinear material at the same rate.

With a simple argument, we can see why this condition is important. Consider the second-harmonic light that is generated in a narrow slice of material near $z = 0$ (Figure 2.24). By the time this second-harmonic light has propagated to $z = z_0$, it has acquired a phase, due to propagation, of $\phi_a = (2\pi n_3/\lambda_3)z_0$. In contrast, second-harmonic light that is generated near the end of the crystal is produced by a fundamental wave that has already propagated a distance z_0 from the input of the crystal and has thus acquired a phase $\phi_b = (2\pi n_1/\lambda_1)z_0$; the second-harmonic light generated here has a phase $2\phi_b$. Therefore, in order for the second-harmonic wave generated near $z = 0$ to add constructively with the second-harmonic generated near $z = z_0$ for an arbitrary value of z_0 , the phase difference $2\phi_b - \phi_a$ must be zero – which again reduces to the condition $n_3 - n_1 = 0$. Making $\Delta k = 0$ thus ensures that contributions to the second-harmonic wave generated at each point along the crystal add up in phase with the contributions generated at every other point along the crystal, thereby maximizing the total generated second-harmonic power.

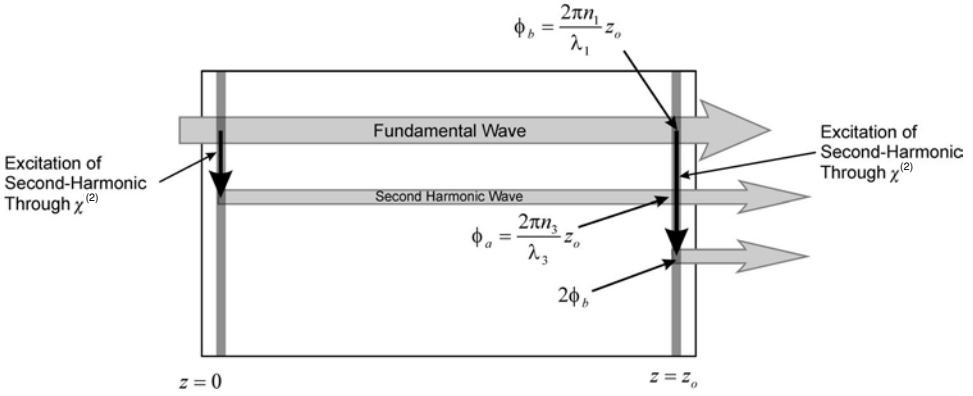


Figure 2.24: Illustration of the physical origin of the phasematching condition for SHG.

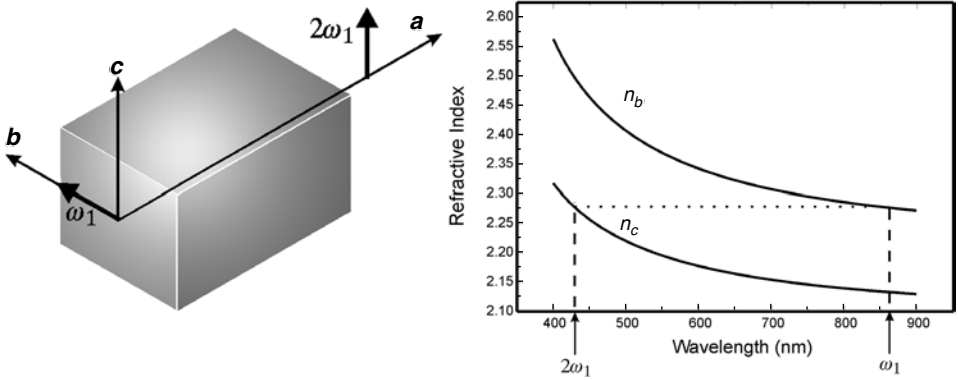


Figure 2.25: An illustration of how crystal birefringence can be used to achieve phasematching in KN.

Satisfying the condition $n_3 - n_1 = 0$ presents a problem. Common nonlinear optical materials exhibit “normal” dispersion, that is, $n_3 > n_1$ (Figure 2.25). Several approaches have been proposed to overcome this difficulty, but in practice, there are two that have been the most heavily exploited: birefringent phasematching and quasi-phasematching. Since birefringent phasematching has been the primary “workhorse” of nonlinear optics until recently, we will discuss it first.

2.4.2 Birefringent phasematching

2.4.2.1 How birefringent phasematching works

In certain optical materials, the refractive index depends on the polarization of the wave. This property can be used to compensate the refractive index difference

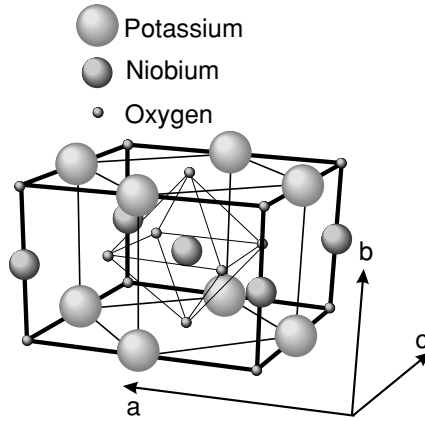


Figure 2.26: Unit cell structure of orthorhombic KN. (Adapted with permission from Zysset *et al.* (1992).)

between fundamental and second-harmonic resulting from normal dispersion. For example, consider SHG using KN. In this material, the crystal structure – that is, the particular arrangement of constituent atoms in a three-dimensional lattice – defines three orthogonal axes, denoted a , b , and c (Figure 2.26). It turns out that light propagating along the a -axis of the crystal travels more slowly if it is linearly polarized along the b -axis than if it is linearly polarized along the c -axis (that is, $n_b > n_c$). Thus, if we constrain the polarization of the fundamental wave to lie along the b -axis (the “slow” polarization) and constrain the polarization of the second harmonic to lie along the c -axis (the “fast” polarization), the phase velocity difference due to dispersion can be compensated for by the phase velocity difference due to birefringence (Figure 2.25), so that $n_c(2\omega_1) = n_b(\omega_1)$ and $\Delta k = 0$.

However, this trick only works for a particular fundamental wavelength; in the example shown in Figure 2.25, $\lambda_1 = 858$ nm. For $\lambda_1 < 858$ nm, the dispersion of the material exceeds the birefringence; for $\lambda_1 > 858$ nm, the birefringence exceeds the dispersion. In general, phasematching will be sensitive not only to wavelength, but to the direction of propagation and to the temperature of the crystal. Calculating the tolerance of the phasematching condition to these (and sometimes other) parameters is important in practice and will be discussed in greater detail later in this chapter.

Although this arrangement provides phasematching, it is not immediately obvious that it will efficiently generate the second harmonic. The nonlinear d tensor must contain an appropriate component that couples a b -polarized fundamental to a c -polarized second harmonic. An examination of the d tensor for KN given in the example of Section 2.2.3 shows that the required component is d_{32} , which *does* exist in this material.

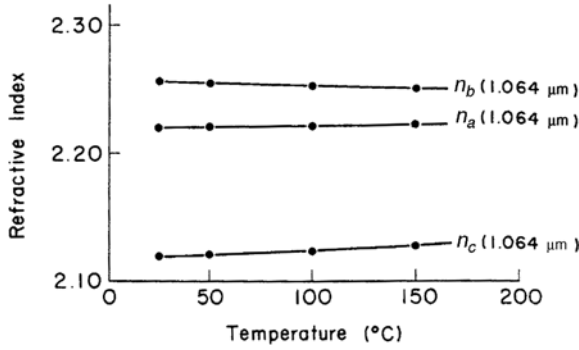


Figure 2.27: Variation of the refractive indices of KN at 1064 nm with temperature. (Adapted with permission from Uematsu (1974).)

In a similar way, if we were to choose the crystallographic b -axis as the direction of propagation, we would find that $\Delta k = 0$ for a wavelength of 983 nm, with the fundamental polarized along the a -axis and the second-harmonic polarized along the c -axis.

2.4.2.2 Temperature tuning

What if we wish to achieve phasematching for some wavelength other than 858 nm or 983 nm? The refractive indices involved change at slightly different rates with temperature (Figure 2.27), so that the wavelength at which $\Delta k = 0$ changes with temperature. We can use this behavior to try to produce noncritical phasematching for the wavelength of interest.

We show an example in Figure 2.28 for both a - and b -axis propagation in KN. At room temperature, the phasematching wavelength for a -axis propagation is 858 nm, as we have already seen. As the temperature is increased, phasematching is pushed to longer wavelengths. By adjusting the temperature to $\sim 180^\circ\text{C}$, we can increase the noncritical phasematching temperature to ~ 946 nm. For b -axis propagation, at room temperature, noncritical phasematching occurs at 983 nm, but by heating the crystal to $\sim 185^\circ\text{C}$, we can obtain noncritical phasematching for SHG of the 1064-nm emission of a Nd:YAG laser.

2.4.2.3 Angle tuning

In the preceding example, rather high temperatures were required to achieve phase-matching for SHG of 946 nm and 1064 nm. In some applications, these high temperatures may be a liability. An alternative tuning technique changes the direction of propagation rather than the temperature in order to achieved phasematching. We

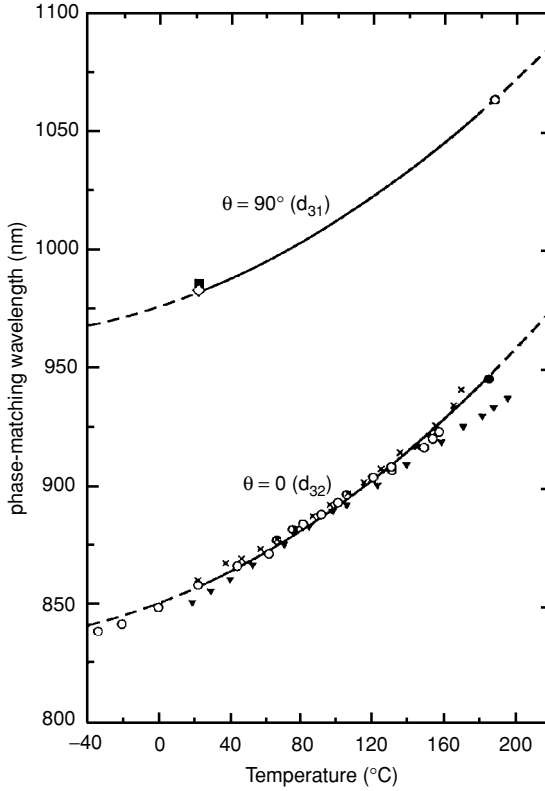


Figure 2.28: Temperature dependence of the wavelengths for noncritical Type-I phasematching in KN. $\theta = 0^\circ$ corresponds to propagation along the a -axis, and $\theta = 90^\circ$ corresponds to propagation along the b -axis. (Reprinted with permission from Biaggio *et al.* (1992).)

just saw that for propagation along the a -axis, we could achieve phasematching at 858 nm, while for propagation along the b -axis, the corresponding wavelength was 983 nm. Hence, we might well expect phasematching for SHG of, say, 946 nm could occur for some intermediate direction of propagation in the a - b plane. In this arrangement (Figure 2.29), the second harmonic is still polarized along the c -axis, but the polarization of the fundamental lies in the a - b plane. Since the second harmonic is polarized parallel to the c -axis, the refractive index experienced by the second-harmonic wave will not change as we rotate the crystal about that axis; however, the refractive index experienced by the fundamental will, ranging between n_a and n_b . It can be shown (see the online supplement) that the variation of refractive index with angle in the a - b plane is given by

$$n_\theta = \left[\frac{\sin^2 \theta}{n_a^2} + \frac{\cos^2 \theta}{n_b^2} \right]^{-\frac{1}{2}} \quad (2.47)$$

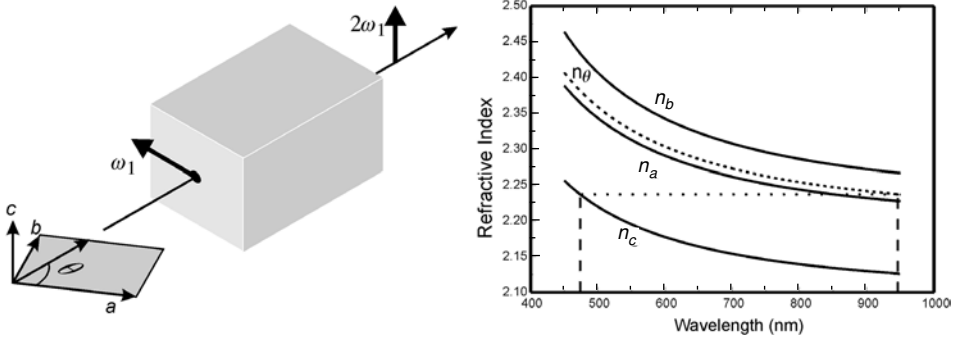


Figure 2.29: Angle-tuned birefringent phasematching for SHG of a 946-nm fundamental in KN.

When $\theta = 0$, $n_\theta = n_b$. When $\theta = 90^\circ$, $n_\theta = n_a$. For what angle θ does $n_\theta(946 \text{ nm}) = n_c(473 \text{ nm})$? From the material properties of KN given in Table 2.5, we calculate that $n_c(473 \text{ nm}) = 2.2378$, $n_a(946 \text{ nm}) = 2.2280$, and $n_b(946 \text{ nm}) = 2.2670$. A little manipulation of Equation (2.47) gives:

$$\cos^2 \theta = \frac{\frac{1}{n_\theta^2} - \frac{1}{n_a^2}}{\frac{1}{n_b^2} - \frac{1}{n_a^2}} \quad (2.48)$$

from which we find that $\theta = 59.9^\circ$ gives the desired result. This phasematching solution is pictured in Figure 2.29, in analogy to the depiction in Figure 2.25 for SHG of 858 nm.

2.4.2.4 Walk-off in angle-tuned phasematching

Angle-tuning often makes it possible to achieve phasematching for SHG of a given wavelength at room temperature. However, it has some disadvantages that we must now discuss. A useful graphical representation (which can be rigorously derived mathematically – see Yariv and Yeh (1984)) helps to illustrate why. We just saw that the index of refraction for a wave polarized in the a – b plane varies with the angle of propagation relative to the a -axis, while the index of refraction for a wave polarized parallel to the c -axis was independent of angle. Suppose we represent this information in a different way. For each direction of propagation, we will draw a line from the origin in that direction and plot two points, one point corresponding to the refractive index for the c -polarized wave and one point corresponding to the refractive index for the a – b polarized wave. As we do this for all possible angles, we find that the curve generated for the c -polarized wave is simply a circle of

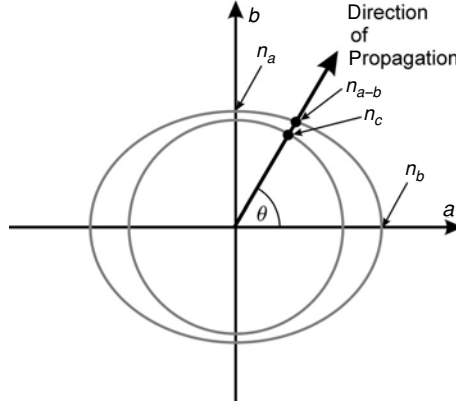


Figure 2.30: Polar plot of the refractive indices of the two eigenpolarizations versus propagation direction for propagation in the a - b plane of KN.

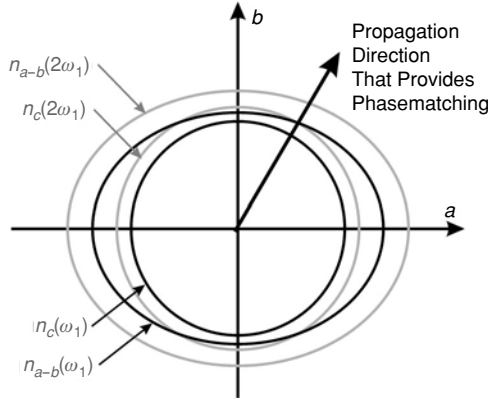


Figure 2.31: Using the index normal curves to find a phasematching solution for SHG using propagation in the a - b plane in KN.

radius n_c . For the other polarization, the plot will be an ellipse that has value n_b for propagation along the a -axis and n_a for propagation along the b -axis (Figure 2.30). This representation directly gives us the refractive indices for any direction of propagation in the a - b plane.

We can see how this representation might be useful for phasematching problems. Consider again the example of SHG in KN. Suppose that we plot out these curves at both the fundamental and the second harmonic (Figure 2.31). We can see that there is an intersection between the curve corresponding to a fundamental wave polarized in the a - b plane and the curve corresponding to a second-harmonic wave polarized parallel to the c -direction. This intersection represents a direction where the refractive index for the fundamental matches that for the second harmonic and phasematching is obtained for that interaction.

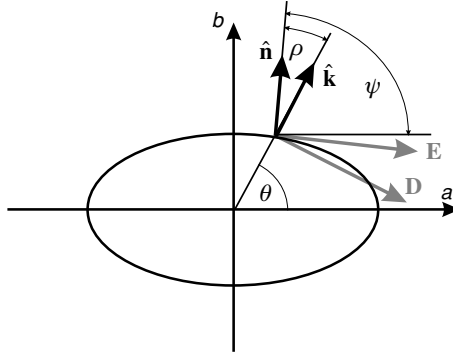


Figure 2.32: Illustration of useful properties of the index normal surface.

These so-called “normal curves” have other useful properties, shown in Figure 2.32. Suppose we draw a line outward from the origin in the direction of propagation until it intersects the normal curve, and then construct a line perpendicular to the curve at this point. This direction, represented by the unit vector $\hat{\mathbf{n}}$, is the direction of the Poynting vector, giving the energy flow in the wave. It may well be that the direction of energy flow is not the same as the direction of propagation (represented by the propagation vector $\hat{\mathbf{k}}$); this phenomenon is the well-known Poynting vector walk-off. We previously saw that for interactions involving focused beams, the walk-off angle ρ was an important parameter; now we can see how to calculate it.

Example: Calculate the walk-off angle for SHG of 946 nm in KN.

For propagation in the a - b plane at an angle θ to the a -axis, the refractive index is given by

$$\frac{1}{n_\theta^2} = \frac{\cos^2 \theta}{n_b^2} + \frac{\sin^2 \theta}{n_a^2} \quad (2.49)$$

Since the refractive index n_θ is equal to the radial distance from the origin to the normal curve, we can write the description of the normal curve as the following equation in polar coordinates:

$$\Phi = \frac{1}{r^2} - \frac{\cos^2 \theta}{n_b^2} + \frac{\sin^2 \theta}{n_a^2} = 0 \quad (2.50)$$

The normal to the curve is given by the gradient, which in polar coordinates is:

$$\nabla \Phi = \frac{\partial \Phi}{\partial r} \hat{\mathbf{a}}_r + \frac{\partial \Phi}{\partial \theta} \hat{\mathbf{a}}_\theta \quad (2.51)$$

where $\hat{\mathbf{a}}_r$ and $\hat{\mathbf{a}}_\theta$ are unit vectors perpendicular to and tangential to the curve,

respectively. The walk-off angle ρ is thus given by

$$\tan \rho = \frac{\sin \rho}{\cos \rho} = \frac{\hat{\mathbf{a}}_\theta \cdot \nabla \Phi}{\hat{\mathbf{a}}_r \cdot \nabla \Phi} = \frac{\tan \theta \left(\frac{n_b^2}{n_a^2} - 1 \right)}{1 + \frac{n_b^2}{n_a^2} \tan^2 \theta} \quad (2.52)$$

If we calculate this value for the case of our phasematching solution for 946-nm doubling, we obtain $\rho = 0.86^\circ$.

2.4.2.5 Effective nonlinearity

When propagation in KN is along the a -axis, the relevant nonlinear coefficient is d_{32} ; when along the b -axis, d_{31} – what is the appropriate nonlinearity when propagation is along some other direction in the a – b plane? We can answer this question using the “normal curve” representation. Figure 2.32 shows that $\mathbf{D}$ is always perpendicular to the direction of propagation $\hat{\mathbf{k}}$, and $\mathbf{E}$ is always perpendicular to the surface normal $\hat{\mathbf{n}}$. When there is walk-off, $\mathbf{D}$ and $\mathbf{E}$ will not be parallel. The induced polarization is related to the electric field $\mathbf{E}$; thus, we need to know its orientation (not that of $\mathbf{D}$) in order to calculate the induced polarization components. By examining Figure 2.32, we can see that the components of $\mathbf{E}$ along the principal axes are:

$$E_a = E_0 \sin \psi \quad (2.53)$$

$$E_b = -E_0 \cos \psi \quad (2.54)$$

Thus, the induced nonlinear polarization is:

$$\begin{bmatrix} \tilde{P}_a^{(\omega_3)} \\ \tilde{P}_b^{(\omega_3)} \\ \tilde{P}_c^{(\omega_3)} \end{bmatrix} = 2\epsilon_0 \begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_0^2 \sin^2 \psi \\ E_0^2 \cos^2 \psi \\ 0 \\ 0 \\ 0 \\ -2E_0^2 \cos \psi \sin \psi \end{bmatrix} \quad (2.55)$$

The c -polarized term is

$$\tilde{P}_c^{(\omega_3)} = 2\epsilon_0 E_0^2 (d_{31} \sin^2 \psi + d_{32} \cos^2 \psi) \quad (2.56)$$

We can define an *effective* nonlinear coefficient $d_{\text{eff}} = d_{31} \sin^2 \psi + d_{32} \cos^2 \psi$. From the material properties given in Table 2.5, we find that $d_{31} = 16.7$ pm/V and $d_{32} = 19.4$ pm/V. With $\psi = \theta + \rho = 60.8^\circ$, we obtain $d_{\text{eff}} = 17.3$ pm/V.

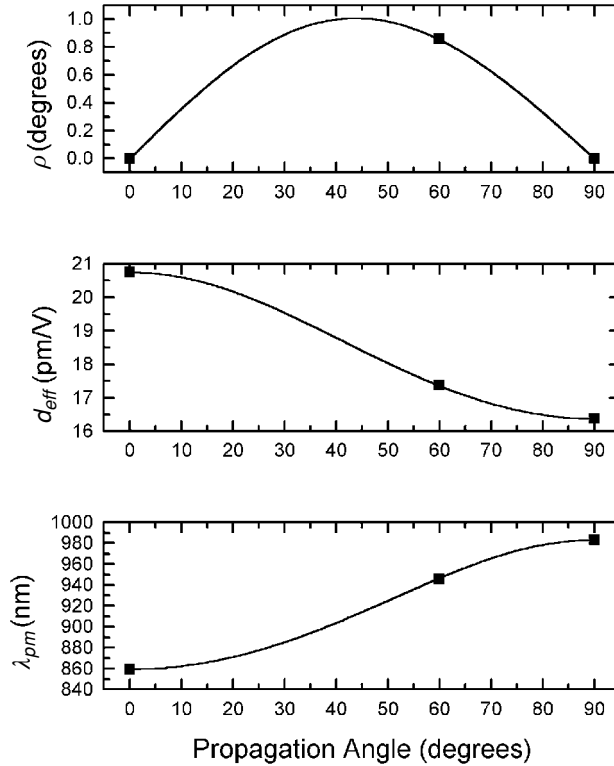


Figure 2.33: Walk-off angle ρ , effective nonlinear coefficient d_{eff} , and phasematching wavelength λ_{pm} for Type-I SHG versus angle of propagation (measured from the a -axis) in KN. Marked data points indicate the extremes of operation as well as the solution for SHG of 946 nm.

Figure 2.33 shows the walk-off angle ρ , the effective nonlinearity d_{eff} , and the angle of propagation θ in the a - b plane as a function of phasematching wavelength λ_{pm} for KN.

2.4.2.6 Noncritical phasematching

Let us return again to the case where propagation is along a crystallographic axis. Figure 2.34 compares the relevant normal curves for phasematching along a principal axis and at an angle. We can see that when the propagation direction lies along a principal axis, the walk-off angle $\rho = 0$. This condition is advantageous for several reasons. As we saw in Section 2.3.1, the spatial distribution of the output second-harmonic beam can be distorted if $\rho \neq 0$. Furthermore, with beams of finite transverse size, walk-off causes a physical separation of the beams in a distance referred to as the aperture length l_a . A simple geometric construction

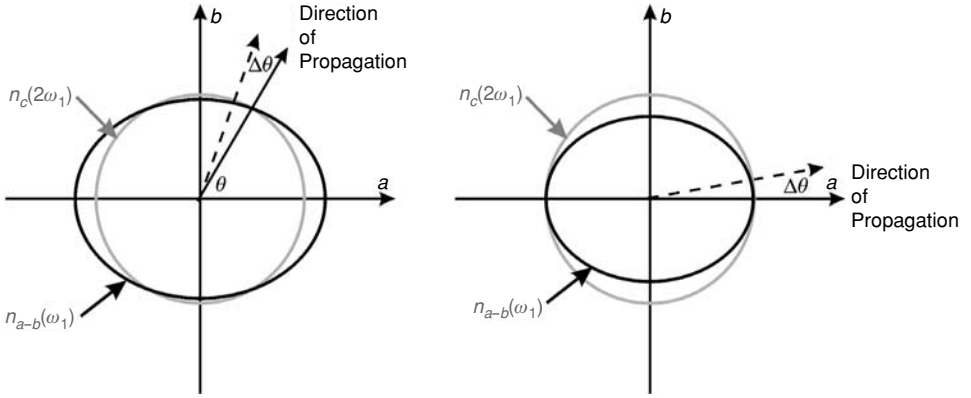


Figure 2.34: Comparison of index normal curves for critical (left) and noncritical (right) phasematching in KN.

gives $l_a \approx 2w_0/\rho$, where w_0 is the radius of the fundamental beam; a more sophisticated calculation for gaussian beams gives $l_a = \sqrt{\pi} w_0/\rho$. We can see that if $\rho \rightarrow 0$, $l_a \rightarrow \infty$, so that the interaction will not be limited by the spatial separation of the beams due to walk-off.

Consider also what happens when we detune the propagation direction from perfect phasematching by a small angle $\Delta\theta$. We can see that in the case of critical phasematching, Δn changes sign as we go through the phasematching angle θ_{pm} . In the case of noncritical phasematching, Δn has the same sign on either side of the propagation direction. Thus, Δn is an odd function of $\Delta\theta$ for critical phase-matching and an even function of $\Delta\theta$ for noncritical phasematching, and therefore we might expect that $\partial \Delta n / \partial \Delta\theta|_{\Delta\theta=0}$ is much smaller when propagation is along a principal axis. This increased tolerance to angular deviation of the phasematching condition when propagation is along a principal axis is, in fact, the origin of the terms *noncritical* and *critical* phasematching.

2.4.2.7 Phasematching tolerances

In preceding sections, we have seen that phasematching is sensitive to a variety of parameters, such as the input wavelength, direction of propagation in the crystal, and temperature. An important practical question is: “Just how sensitive is it?” How precisely must this delicate balance between the propagation speeds of the fundamental and second harmonic be maintained? Once we have found a phasematching solution for the wavelength of interest, how tightly must we control the temperature of the crystal, or the wavelength of the laser, or the direction of propagation in order to maintain the blue-green output power?

The basic methodology for answering this question is the same regardless of the parameter we are interested in investigating, which we will call ξ , understanding that it could be temperature, wavelength, or angle. Generally, what we are interested in knowing is how much this parameter can be varied before the second-harmonic or sum-frequency output is degraded by a certain amount. A common measure is the change in ξ that causes the output power to drop to 50% of its maximum value. Thus, we can recast Equation (2.21) in the form

$$P_3 = P_{max} \text{sinc}^2 \left[\frac{\Delta k(\xi)l}{2} \right] \quad (2.57)$$

where we have acknowledged that Δk is a function of ξ . The sinc^2 function has its maximum value of 1 when $\xi = \xi_{pm}$; that is, $\Delta k(\xi_{pm}) = 0$. It will have the value 0.5 when the argument is equal to 1.39. Therefore, the value of ξ which makes the output value one-half of its maximum must satisfy $\Delta k(\xi_{1/2})l/2 = 1.39$. We can expand Δk as a function of ξ in a Taylor series:

$$\Delta k(\xi) = \left. \frac{\partial \Delta k}{\partial \xi} \right|_{\xi=\xi_{pm}} (\xi - \xi_{pm}) + \frac{1}{2} \left. \frac{\partial^2 \Delta k}{\partial \xi^2} \right|_{\xi=\xi_{pm}} (\xi - \xi_{pm})^2 + \dots \quad (2.58)$$

Often, we will find that the term involving the second derivative can be neglected in comparison with the term involving the first derivative. If this is the case, we find that:

$$\left. \frac{1}{2} \frac{\partial \Delta k}{\partial \xi} \right|_{\xi=\xi_{pm}} (\xi_{1/2} - \xi_{pm})l = 1.39 \quad (2.59)$$

or

$$\Delta \xi_{1/2} = (\xi_{1/2} - \xi_{pm}) = \frac{2.78}{\left. \frac{\partial \Delta k}{\partial \xi} \right|_{\xi=\xi_{pm}}} l \quad (2.60)$$

Example: Angular tolerance for frequency-doubling of 946-nm light in KN
Critical phasematching for frequency-doubling of 946-nm light can be obtained in KN when the fundamental propagates in the a - b plane at an angle $\theta = 59.9^\circ$ to the a -axis and $T = 23^\circ\text{C}$; the generated second harmonic is polarized along the c -axis. Alternatively, noncritical phasematching is obtained for $\theta = 0^\circ$ and $T = 183.7^\circ\text{C}$. Let us calculate the angular tolerances for these phasematching conditions.

In both cases,

$$\Delta k = \frac{4\pi}{\lambda_1} [n_c(2\omega_1) - n_\theta(\omega_1)]$$

Thus,

$$\frac{\partial \Delta k}{\partial \theta} = -\frac{4\pi}{\lambda_1} \frac{\partial n_\theta}{\partial \theta}$$

By differentiating Equation (2.47), we find that

$$\frac{\partial n_\theta}{\partial \theta} = -\frac{1}{2} n_\theta^3 \sin 2\theta \left(\frac{1}{n_a^2} - \frac{1}{n_b^2} \right) \quad (2.61)$$

Earlier we found that for $\theta = 59.9^\circ$, $n_\theta = 2.2378$, and that $n_a = 2.2280$ and $n_b = 2.2670$; hence we find that

$$\Delta\theta_{1/2} = \frac{6.3 \times 10^{-6}}{l} \text{ rad} \quad (2.62)$$

Thus, for a crystal of length 1 cm, the angle must deviate by less than ± 0.63 mrad or $\pm 0.036^\circ$ in order to keep the output power within 50% of its maximum value.

Now let us consider the noncritical phasematching case, in which $\lambda_1 = 946$ nm and $\theta = 0$ at $T = 183.7^\circ\text{C}$. Here we find that $\partial n_\theta / \partial \theta|_{\theta=0} = 0$, and we must consider the second term in the Taylor series expansion. We can determine that

$$\frac{\partial^2 n_\theta}{\partial \theta^2} = -\frac{1}{2} \left(\frac{1}{n_a^2} - \frac{1}{n_b^2} \right) \left(2n_\theta^2 \frac{\partial n_\theta}{\partial \theta} \sin 2\theta + 2n_\theta^3 \cos 2\theta \right) \quad (2.63)$$

so that

$$\left. \frac{\partial^2 n_\theta}{\partial \theta^2} \right|_{\theta=0} = -n_\theta^3 \left(\frac{1}{n_a^2} - \frac{1}{n_b^2} \right) \quad (2.64)$$

From Equation (2.60), we find that

$$\Delta\theta_{1/2} = \sqrt{\frac{1.39\lambda_1}{\pi \left| \frac{\partial^2 n_\theta}{\partial \theta^2} \right|_{\theta=0}}} l = \frac{2.3 \times 10^{-3}}{\sqrt{l}} \text{ rad} \quad (2.65)$$

For a 1-cm long crystal, $\Delta\theta_{1/2} = 2.3 \times 10^{-2}$ rad = 23 mrad = 1.3° . We can see why this form of phasematching is called noncritical! Here we may deviate by $\pm 1.3^\circ$ from the proper angle before losing 50% of the output power, while for critical phasematching with $\theta = 59.9^\circ$ at room temperature, the tolerance was only $\pm 0.036^\circ$. Although heating the crystal and maintaining a uniform temperature over its entire length may add to the complexity of the experiment or final device, this additional complexity may be justified in order to obtain the advantages of noncritical phasematching.

Experiment. Baumert (1985) investigated noncritically-phasematched SHG of 860 nm at room temperature with a -axis propagation in a 8.97-mm long crystal and

measured the angular tolerance to be 2.1° . If we calculate the angular bandwidth using Equation (2.65), we obtain 1.6° , in good agreement with what Baumert measured.

Example: Wavelength tolerance for 860-nm doubling in KN Here we need to find

$$\frac{\partial \Delta k}{\partial \lambda} = \frac{4\pi}{\lambda} \left(\frac{\partial \Delta n}{\partial \lambda} - \frac{\Delta n}{\lambda} \right)$$

If we have phasematching, $\Delta n(\lambda_{pm}) = 0$. When we write out $\partial \Delta n / \partial \lambda$ we find that it is $\partial n_c(\lambda/2) / \partial \lambda - \partial n_b(\lambda) / \partial \lambda$, which can also be written as

$$\frac{1}{2} \frac{\partial n_c}{\partial \lambda} \Big|_{\lambda=\lambda_{pm}} - \frac{\partial n_b}{\partial \lambda} \Big|_{\lambda=\lambda_{pm}}$$

when evaluated at the wavelength of interest. We then find:

$$\Delta \lambda_{FWHM} = \left| \frac{1.39 \lambda_{pm}}{\pi l \left[\frac{1}{2} \frac{\partial n_c}{\partial \lambda} \Big|_{\lambda=\lambda_{pm}/2} - \frac{\partial n_b}{\partial \lambda} \Big|_{\lambda=\lambda_{pm}} \right]} \right| \quad (2.66)$$

The derivatives can be found either by differentiating the Sellmeier equations or by numerical evaluation.

Experiment. Baumert (1985) measured the wavelength bandwidth for noncritically phasematched frequency-doubling of 848.9-nm light with propagation along the a -axis of KN. He used a 8.97-mm long crystal cooled to $T \approx 0^\circ\text{C}$ to obtain phase-matching. We can calculate the expected bandwidth from Equation (2.66). Using the Sellmeier equation for KN given in Chapter 4, we find that $\partial n_c / \partial \lambda|_{\lambda=\lambda_{pm}/2} = -1.181 \mu\text{m}^{-1}$ and $\partial n_b / \partial \lambda|_{\lambda=\lambda_{pm}} = -0.1426 \mu\text{m}^{-1}$. Substituting these values into Equation (2.66), we obtain $\Delta \lambda_{FWHM} = 0.9 \text{ \AA}$, in good agreement with Baumert's measured value of $0.88 \text{ \AA}$.

Example: Temperature tolerance for 860-nm doubling in KN In this case, not only may Δk vary with temperature, but so may the crystal length l , as result of thermal expansion. Hence, we need to evaluate $\partial (\Delta k l) / \partial T$ rather than simply $\partial \Delta k / \partial T$. When we do this, we find that

$$\frac{\partial (\Delta k l)}{\partial T} = l \frac{\partial \Delta k}{\partial T} + \Delta k \frac{\partial l}{\partial T} \quad (2.67)$$

$$= l \frac{\partial \Delta k}{\partial T} + \Delta k \alpha l \quad (2.68)$$

where we have introduced the coefficient of thermal expansion $\alpha = (1/l)\partial l/\partial T$. However, we note that since $\Delta k \approx 0$, the term involving thermal expansion of the crystal should be negligible. Hence, we find for the temperature tuning bandwidth:

$$\Delta T_{FWHM} = \left| \frac{1.39\lambda_{pm}}{\pi l \left[\left. \frac{\partial n_c(\lambda_{pm}/2)}{\partial T} \right|_{T=T_{pm}} - \left. \frac{\partial n_b(\lambda_{pm})}{\partial T} \right|_{T=T_{pm}} \right]} \right| \quad (2.69)$$

Experiment. Baumert *et al.* (1983) measured the temperature bandwidth for SHG of 838.5-nm light. The 8.97-mm long crystal was cooled to -34.15°C in order to achieve noncritical phasematching for this wavelength. Using the temperature-dependent Sellmeier equations given in Table 2.5 for KN, we find $\partial n_c(\lambda_{pm}/2)/\partial T = 8.64 \times 10^{-5} \text{ }^\circ\text{C}^{-1}$ and $\partial n_b(\lambda_{pm})/\partial T = -3.80 \times 10^{-5} \text{ }^\circ\text{C}^{-1}$ and therefore, $\Delta T_{FWHM} = 0.33^\circ\text{C}$. Baumert and colleagues measured $\Delta T_{FWHM} = 0.3^\circ\text{C}$.

2.4.2.8 The effect of nonuniformity

In our preceding discussion, we expanded Δk in a Taylor's series, usually keeping only the first-order term unless it happened to be zero. Thus, we obtained a

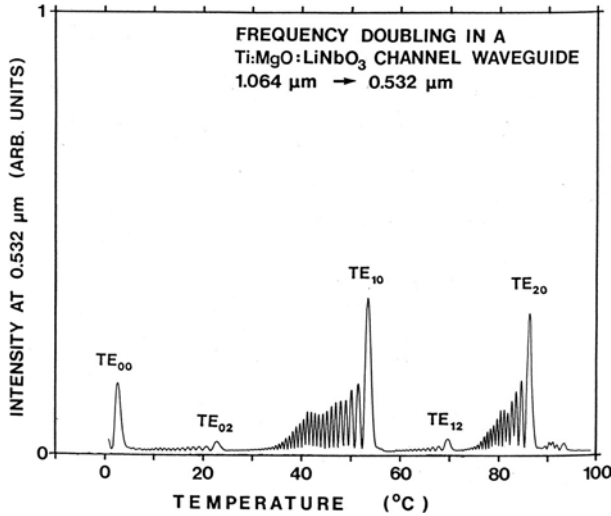


Figure 2.35: Measured green power produced by guided-wave SHG in a LN waveguide. The ripples appearing on the low-temperature side of each peak are due to nonuniformities. (Reprinted with permission from Laurell and Arvidsson (1988).)

phasematching response of the form

$$\text{sinc}^2\left(\frac{l}{2} \frac{\partial \Delta k}{\partial \xi} \bigg|_{\xi=\xi_{pm}} \Delta \xi\right)$$

where ξ is a phasematching parameter we wish to vary, such as temperature, wavelength, or angle. We see that this expression is symmetric in $\Delta \xi$, so that if we vary ξ slightly above ξ_{pm} , the effect on the blue-green output power should be the same as if we vary ξ the same amount below ξ_{pm} . In practice, however, one sometimes observes asymmetric phasematching behavior (Nash *et al.*, 1970, Tsuya *et al.*, 1970). Often, such behavior is more pronounced in waveguides than in bulk crystals, since both the inherent uniformity of the crystal and the uniformity of the fabricated waveguide become important. Figure 2.35 shows an example where frequency-doubling is achieved in a LN waveguide, and the temperature tuning curve is highly asymmetric about the main peak. This behavior has been attributed to a parabolic variation of $\Delta \beta$, based upon the results of numerical simulations, as shown in Figure 2.36 (Laurell and Arvidsson, 1988).

2.4.3 Quasi-phasematching (QPM)

2.4.3.1 How QPM works

We have just seen that the birefringence of certain nonlinear optical materials can be used to compensate for dispersion and thereby achieve phasematching; however, we have also seen that this approach is subject to several limitations. It may not be possible to find a material that has sufficient birefringence to provide phasematching at the wavelength of interest; we may be forced to use critical phasematching, which can result in uncomfortably tight tolerances or a distorted output beam; we may find that the nonlinearity associated with birefringent phasematching is too small to be useful.

QPM achieves phasematching through artificial structuring of the nonlinear material rather than through its inherent birefringent properties. In fact, QPM may be used to produce phasematching in optically isotropic materials that have no birefringence at all. This is an exciting prospect, since cubic semiconductors such as GaN, ZnSe, ZnS, ZnTe, and GaAs have extremely large second-order nonlinearities compared to materials like LN, LT (lithium tantalate), KTP, and KN, but are optically isotropic. Thus, QPM potentially makes a wider range of materials available for nonlinear optical applications than does birefringent phasematching.

Historically, QPM was suggested prior to birefringent phasematching (Armstrong *et al.*, 1962). Although some attempts to implement QPM were made

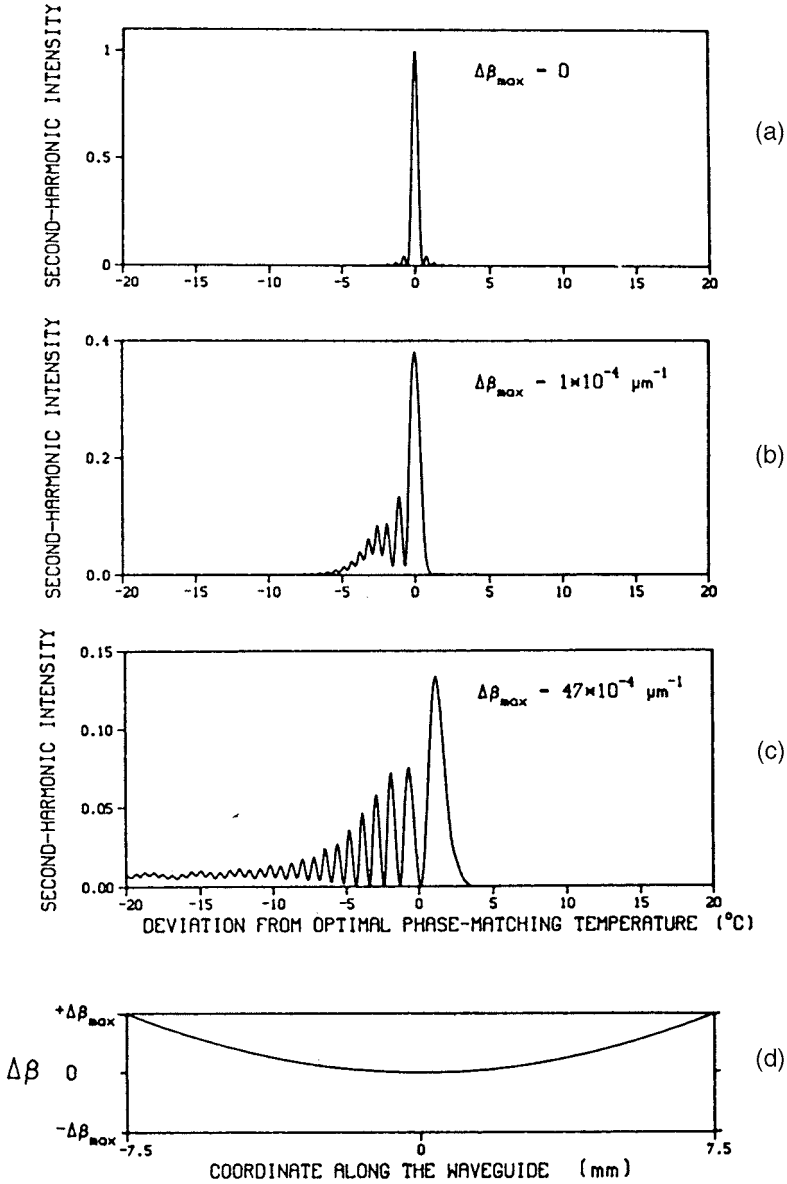


Figure 2.36: Results of a modeling calculation showing the effects of nonuniformity in $\Delta\beta$, the waveguide equivalent of Δk . (a) shows the second-harmonic power obtained as the temperature is varied for a homogeneous waveguide having no variation in $\Delta\beta$ over the interaction length. (b) and (c) show the same quantity for guides having a parabolic variation in $\Delta\beta$ along the guide, as illustrated in (d). (Reprinted with permission from Laurell and Arvidsson (1988).)

in the 1970s (for example, Bloembergen and Sievers (1970), Dewey and Hocker (1975), Okada *et al.*, (1976), Thompson *et al.*, (1976)), the difficulties inherent in structuring nonlinear materials on the micrometer length scale required for most interactions of interest limited the usefulness of QPM until very recently.

We can understand the principle of QPM by considering SHG in a material which, in its initial state, does not provide phasematching. We saw in Section 2.2.5 that for a monochromatic input wave, the generated second-harmonic field in the frequency-domain representation would be:

$$\frac{\partial \tilde{\mathcal{E}}_g(x, \omega)}{\partial x} = \frac{-j\pi k_3 d_{\text{eff}} A_1^2}{2n_3^2} [e^{j\Delta k x} \delta(\omega - \omega_3) - e^{-j\Delta k x} \delta(\omega + \omega_3)] \quad (2.70)$$

When we perform the integration over x , we obtain a term like $\int e^{j\Delta k x} dx$ and its complex conjugate. This term expresses mathematically what we described verbally in Section 2.4.1: the superposition of second-harmonic waves generated at different points in the crystal. The complex number $e^{j\Delta k x}$ has magnitude 1 and phase angle $\Delta k x$. We can represent this complex number as a phasor of length 1 at an angle $\phi = \Delta k x$. For purposes of visualization, we can approximate the continuous integral $\int_0^l e^{j\Delta k x} dx$ with a discrete sum: $(1 + e^{j\Delta k \delta x} + e^{j\Delta k 2\delta x} + \dots + e^{j\Delta k (N-1)\delta x})\delta x = \delta x \sum_{i=1}^N e^{j\Delta k (i-1)\delta x}$, where $l = N\delta x$. We can graphically represent this sum of complex numbers using phasors and employing phasor addition as depicted in Figure 2.37. If $\Delta k = 0$, then $e^{j\Delta k \delta x} = 1$ and this integral has its maximum

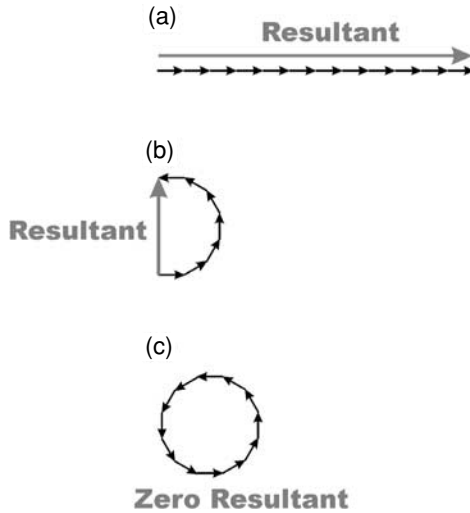


Figure 2.37: Phasor addition interpretation of phasematching: (a) the accumulation of second-harmonic amplitude for the case $\Delta k = 0$, (b) the resultant at $x = l_c$ for $\Delta k \neq 0$, and (c) the resultant at $x = 2l_c$ for $\Delta k \neq 0$.

value – that is, all the phasors align in the same direction and add to give the maximum sum (Figure 2.37(a)), corresponding to perfect constructive interference between second-harmonic contributions from different sections of the crystal. However, if $\Delta k \neq 0$, there is a slight phase error between second-harmonic contributions from different portions of the crystal, so that perfect constructive interference does not occur and the second-harmonic output is reduced. In phasor terms, there is now an angle between each phasor and its neighbor (Figure 2.37(b)), and the resultant is smaller than for $\Delta k = 0$. In fact, if $\Delta kl = 2\pi$ (or an integer multiple thereof), the second-harmonic output will be zero (Figure 2.37(c)).

We note that in the situation depicted in Figure 2.37(c), the maximum second-harmonic field occurs at $x = l/2$. At that location, a phase mismatch of π has accumulated. Suppose that we could somehow intervene in the crystal at this point and introduce an additional π phase shift in the generated light – then we would have the situation in Figure 2.38. The introduction of this additional phase shift restores constructive interference so that the second-harmonic field continues to grow.

We call the distance over which a phase shift of π is accumulated the “coherence length”, $l_c = \pi/\Delta k$. In the materials most commonly used for blue-green light generation, the coherence length is on the order of 3–7 μm (Figure 2.39). Although we showed only two coherence lengths in Figure 2.38, a real crystal will be hundreds or thousands of coherence lengths long, and we must introduce an additional π phase shift every l_c throughout the entire length of the material. Thus, we are faced with the rather daunting prospect of somehow altering a nonlinear crystal every few micrometers to produce the required phase correction and doing this consistently over a length of several millimeters! This stringent requirement is one of the main reasons that the practical development of QPM materials has taken some 30 years.

The periodic phase correction inherent in QPM causes the second-harmonic power to build up in a stepwise fashion (Figure 2.40). The build up is less rapid

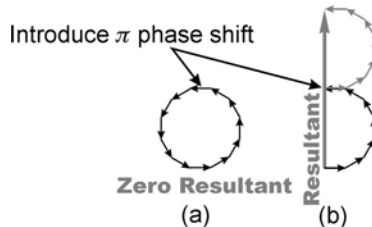


Figure 2.38: Incorporation of an additional π phase shift at a location where a π phase error has already accrued due to nonzero Δk causes the second-harmonic amplitude to continue growing: (a) the growth of the second-harmonic amplitude over a path $2l_c$ long, resulting in a net zero amplitude (same as Figure 2.37(c)); (b) the growth of the second-harmonic amplitude over a path $2l_c$ long, but with a π phase shift introduced after one l_c , resulting in a continued increase in the second-harmonic amplitude.

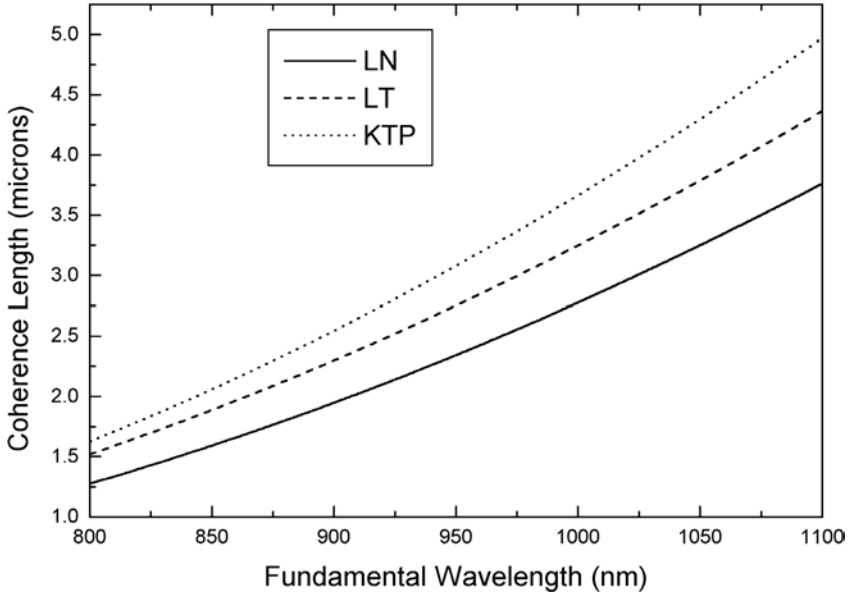


Figure 2.39: Coherence length versus fundamental wavelength for LN, LT, and KTP.

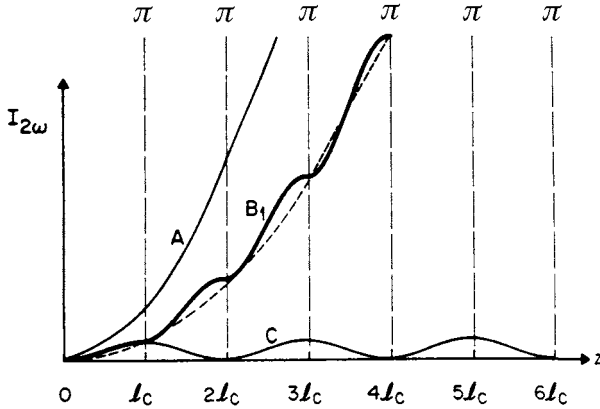


Figure 2.40: Stepwise build up of second-harmonic power due to QPM. Curve A shows the build up with perfect $\Delta k = 0$ phasematching. Curve B shows the build up achieved with $\Delta k \neq 0$ and QPM, where an additional π phase shift is introduced every coherence length l_c . Curve C shows the periodic build up and decrease of second-harmonic power with $\Delta k \neq 0$ and no QPM. (Reprinted with permission from Fejer *et al.* (1992). Copyright: IEEE.)

than would occur with birefringent phasematching. Later in this section, we shall calculate just how much less rapid the build up is. Despite this reduced efficiency, the overriding advantage of QPM is that it can be employed where birefringent phasematching is impossible, and can provide *noncritical* phasematching for

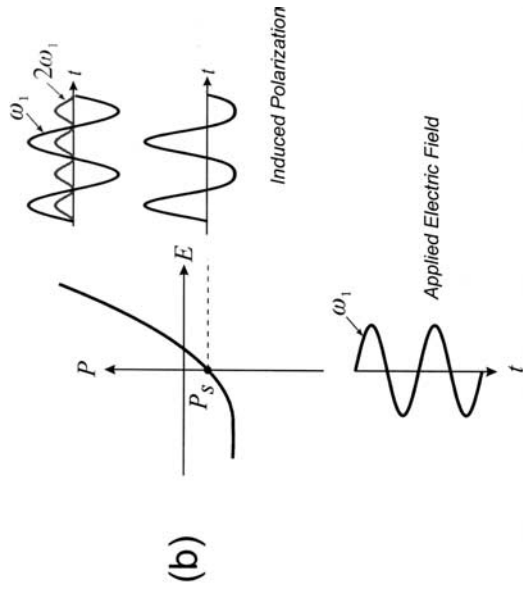
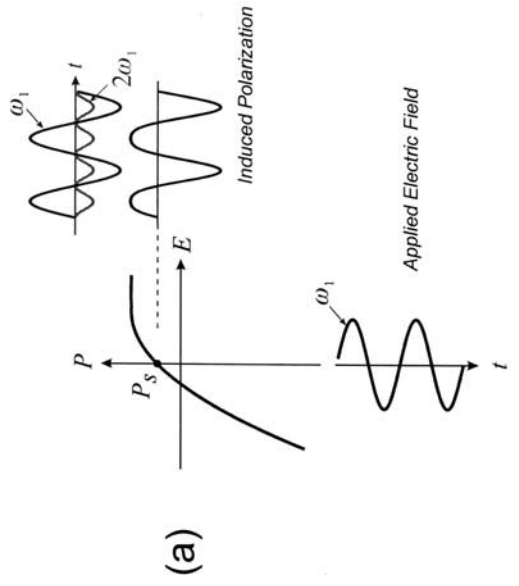


Figure 2.41: Inversion of the spontaneous polarization from (a) to (b) leads to a π phase shift in the generated second harmonic.

any nonlinear interaction permitted by the transparency range of the material. In particular, it can be used to implement interactions involving the d_{33} coefficient, which is typically the largest in ferroelectric nonlinear materials like LN, LT, and KTP. Since an interaction using d_{33} couples a fundamental and second harmonic that have the same polarization, birefringent phasematching can never be used for such an interaction.

2.4.3.2 Fabrication of QPM structures

One way, in principle, to introduce this additional π phase shift is evident from Equation (2.70). If the sign of the nonlinear coefficient d_{eff} could be reversed, that is, switched from $+$ to $-$, this reversal would introduce a π phase shift into the locally generated wave. How can this be done? In principle, we could slice the nonlinear crystal into sections one coherence length long and rotate every other section by 180° about the direction of propagation. This approach has been pursued for frequency-doubling of $10.6\ \mu\text{m}$ (Gordon *et al.*, 1993). A stack of extremely clean and highly polished GaAs wafers was prepared such that every other one was rotated by 180° , then they were fused together under pressure at high temperature. At a wavelength of $10.6\ \mu\text{m}$, the coherence length in GaAs is $106\ \mu\text{m}$ – long enough to make preparation of a rotated stack of this kind feasible. However, in blue-green generation, where the coherence length is only a few micrometers, such a mechanical approach is not practical.

However, there is another approach that is well suited to many of the materials that are traditionally used for blue-green generation, like LN, LT, and KTP. These materials are ferroelectric, which means that below a certain temperature (called the Curie temperature), they exhibit a spontaneous electric polarization P_s , even when no external electric field is applied. This polarization arises from an internal separation of charge due to the spatial arrangement of the atoms in the crystal. This separation of charge defines a direction connecting the negative center-of-charge to the positive center-of-charge; thus, ferroelectric materials have a “polar axis” that acts as a directional reference by which the crystal can “distinguish” the difference between an applied electric field that points in the same direction as the spontaneous polarization and one that points in the opposite direction.

Figure 2.41 depicts this effect of the spontaneous polarization on SHG. Figure 2.41(a) is similar to Figure 2.3, except that in Figure 2.3, $P = 0$ when $E = 0$, whereas in Figure 2.41(a), $P = P_s$ when $E = 0$. The applied electric field at the fundamental induces a separation of charge that adds to and subtracts from the charge separation associated with the spontaneous polarization. In the figure, the nonlinear response of the induced polarization has been decomposed into components at the fundamental and second-harmonic frequencies. Figure 2.41(b) shows the same

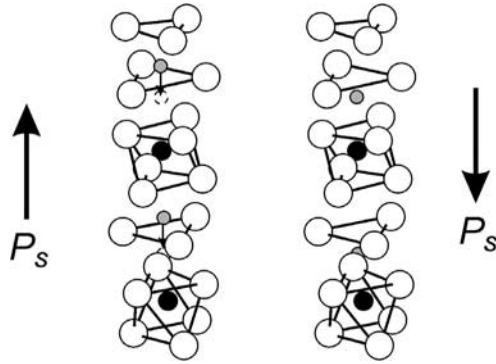


Figure 2.42: Crystal structure of LN, showing the physical basis for inverting the spontaneous polarization. The small gray circles represent lithium ions. (Adapted with permission from Haycock and Townsend (1986). Copyright: American Institute of Physics.)

effect when the direction of the spontaneous polarization is reversed. It is evident that the second-harmonic components of the two responses are 180° out of phase with each other. Thus, periodic inversion of the spontaneous polarization provides a means for producing the 180° phase shift required to implement QPM.

How can this inversion of spontaneous polarization be achieved? Figure 2.42 shows the crystalline structure of LN. We can see that the lithium ions lie just above a triangle formed by three oxygen atoms. If these lithium ions are pushed through the oxygen triangles so that they reside on the other side, we find that this structure is the same as if we had rotated the original structure by 180° (the interested reader can verify this by photocopying Figure 2.42 onto a transparency and overlaying it on the original figure); thus the spontaneous polarization must be opposite to that of the original structure. As the temperature of the crystal is increased, the lithium ions move closer to the oxygen triangles, and the barrier opposing their passage through the oxygen triangle is reduced, becoming easier to surmount. At the Curie temperature, where the spontaneous polarization is zero, the average position of the lithium ions is in the plane of the triangle. Some processes for producing this inversion rely on heating the crystal, which lowers the barrier and produces pyroelectric fields that aid in pushing the lithium ion across the triangle; others rely on a sufficiently large electric field applied at room temperature to accomplish the transition. Essentially the identical situation occurs with domain inversion in LT; in KTP, the movement of potassium ions into alternative sites induces the domain inverted structure (Thomas and Glazer, 1991, Stolzenberger and Scripsick, 1999). These techniques are discussed briefly below and in greater detail in subsequent chapters. The process of aligning the direction of the spontaneous polarization is called “poling,” and a contiguous region of the crystal in which the spontaneous polarization has the same alignment is called a ferroelectric domain. Thus, a crystal

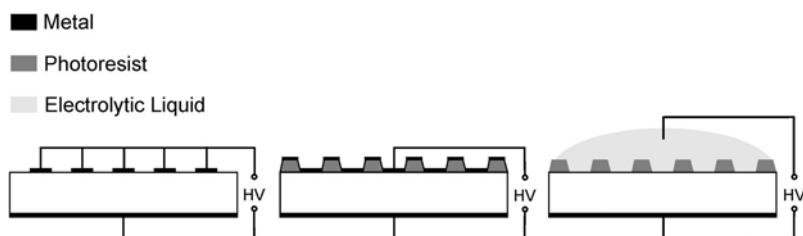


Figure 2.43: Configurations for poling with a periodic electrode.

having periodic reversals of the spontaneous polarization is said to be “periodically poled” or “periodically domain-inverted.”

How can a domain-inverted structure with a period of a few micrometers be produced in a nonlinear material? Several methods have been demonstrated. At present, the most successful and widely used involves the definition of a periodic electrode on one surface of the crystal. This periodic electrode can be a patterned metal film, or a photoresist layer overlaid with a metal film or a liquid electrolyte (Figure 2.43). A uniform electrode is applied to the opposite surface. When a sufficiently large electric field is applied to these electrodes, inverted domains begin to nucleate under the regions where the periodic electrode is in contact with the crystal. Under the influence of the applied field, these domains grow until they fill the area directly under the electrode and extend across the entire thickness of the crystal. Periodic poling has been demonstrated using this approach in LN, LT, KTP, rubidium titanyl arsenate (RTA), and KN. It is also possible to develop the electric field necessary to produce domain inversion by depositing charge using an electron beam. This approach has been used to produce domain inversion in LN, LT, and KTP; however, the process is time consuming and tends to be less reproducible than electric field poling using a periodic electrode.

Although these methods can produce a domain grating that extends through the entire thickness of the crystal, the thickness that can be achieved is limited by dielectric breakdown of the material, and by the greater difficulty of maintaining excellent uniformity as the thickness of the crystal is increased. The poling field required for LN (~ 25 kV/mm) limited initial experiments to thicknesses of about 0.25 mm (Burns *et al.*, 1994). More recently, poling of 0.5-mm thick crystals with periods of ~ 4 μm (Batchko *et al.*, 1999) and of 3-mm thick crystals with a period of ~ 30 μm has been achieved (Grisard, *et al.*, 2000). Increasing the thickness of the poled crystal by stacking a number of thinner pieces and bonding them together has been demonstrated (Misse *et al.*, 1998). One advantage of KTP is that the field required for domain inversion is only about 2 kV/mm; thus, thicker crystals can be poled in the ambient atmosphere without risking breakdown. Poling in air also allows direct monitoring of the poling process by *in situ* SHG (Chen and Risk, 1994).

Another approach for achieving thick periodically-poled crystals for bulk interactions was one of the first that was attempted – inducing a periodic domain structure during the growth of the crystal. For example, periodically-poled LN crystals were grown with periodic domain structures by Feng *et al.* (1980), who used a rotating temperature gradient to produce pyroelectric fields that caused domain reversals during the growth process. Although work on this approach has continued, particularly with single-crystal fibers, it has proven to be extremely difficult to grow crystals with the short periods necessary for blue-green generation with adequate uniformity.

Under certain circumstances, domain inversion can also be produced by diffusing certain ions into nonlinear crystals at elevated temperature, or by allowing constituents of the material to diffuse out. The chemical modification in the first several micrometers of the material caused by these processes can cause the spontaneous polarization to invert in those regions. This approach has been demonstrated, for example, using titanium in-diffusion in LN and LT, LiO out-diffusion in LN, and $\text{Rb} \leftrightarrow \text{K}$ ion exchange in KTP. The cross-sectional shapes of these shallow domains depend upon the material and the method used. Triangular, semicircular, and rectangular domains have been observed (Figure 2.44). Because the domain-inverted regions are shallow, this approach has been best suited for producing periodically-poled guided-wave structures. Rectangular domains are desirable for efficient SHG, but good results have been obtained with other domain shapes in waveguides. Since $\text{Rb} \leftrightarrow \text{K}$ ion-exchange also produces a refractive index increase in KTP, a single step can be used to create both periodic poling and a segmented waveguide.

Finally, a combination of chemical alteration and electric field poling can be used to produce periodic domain inversion. It has been discovered that the susceptibility

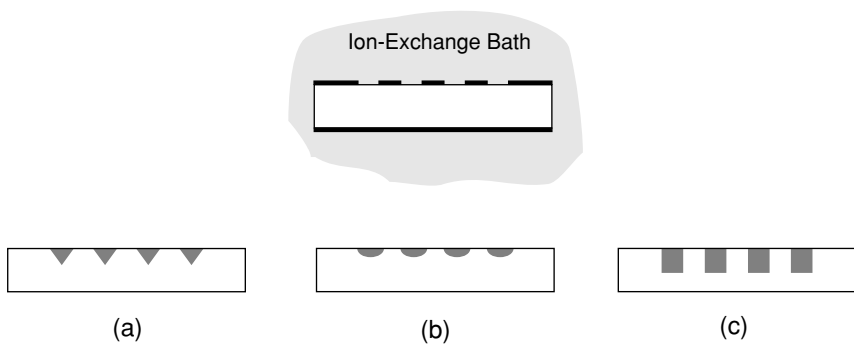


Figure 2.44: Domain shapes produced by the ion-exchange approach. Triangular domains (a) are produced by titanium indiffusion in LN. Semicircular domains (b) are produced by proton exchange followed by a heat treatment in LT. Rectangular domains (c) are produced by $\text{Rb} \leftrightarrow \text{K}$ ion exchange in KTP.

of some materials to electric-field poling can be influenced by chemical treatments applied to the surface of the crystal. For example, it has been observed that replacing the potassium ions at the surface of a hydrothermally-grown KTP crystal by rubidium ions makes that region of the crystal more resistant to domain inversion by an applied electric field (Risk and Lau, 1996). Not only does the exchanged region itself resist poling, but it tends to “shadow” the region of untreated material underneath it. Similar effects have been observed for proton-exchanged regions in LN and LT. Thus, chemical patterning of the surface can be used to define the regions to remain unpoled, then a uniform electric field can be applied to the entire crystal. Since a periodic electrode is not required, this approach ameliorates some of the problems associated with fringing fields in conventional electric-field periodic poling.

2.4.3.3 Theory of QPM

In the development of SHG theory earlier in this chapter, we assumed the nonlinear coefficient d_{eff} to be constant over the length of the crystal. Yet, we have just been speaking about intentionally imposing a periodic modulation on d_{eff} in order to implement QPM. Let us again consider SHG from a monochromatic source, but allow for a nonlinear coefficient that varies along the direction of propagation. If the nonlinear coefficient is not a constant, but varies along the direction of propagation, we can write Equation (2.19) as:

$$\frac{\partial \tilde{\mathcal{E}}_3(z, \omega)}{\partial z} = \frac{-j\pi k_3 d(z) A_1^2}{2n_3^2} [e^{j\Delta k z} \delta(\omega - \omega_3) - e^{-j\Delta k z} \delta(\omega + \omega_3)] \quad (2.71)$$

Integrating over the interaction length l , we obtain:

$$\tilde{\mathcal{E}}_3(l, \omega) = \frac{-j\pi k_3 A_1^2}{2n_3^2} \left\{ \left[\int_0^l d(z) e^{j\Delta k z} dz \right] \delta(\omega - \omega_3) - \left[\int_0^l d(z) e^{-j\Delta k z} dz \right] \delta(\omega + \omega_3) \right\}$$

This expression contains the quantity $\int_0^l d(z) e^{j\Delta k z} dz$ and its complex conjugate. If we define the function $d(z)$ so that it is zero outside the crystal ($z < 0$ and $z > l$), then we can write this expression as:

$$D(\Delta k) = \left[\int_{-\infty}^{\infty} d(z) e^{j\Delta k z} dz \right] \quad (2.72)$$

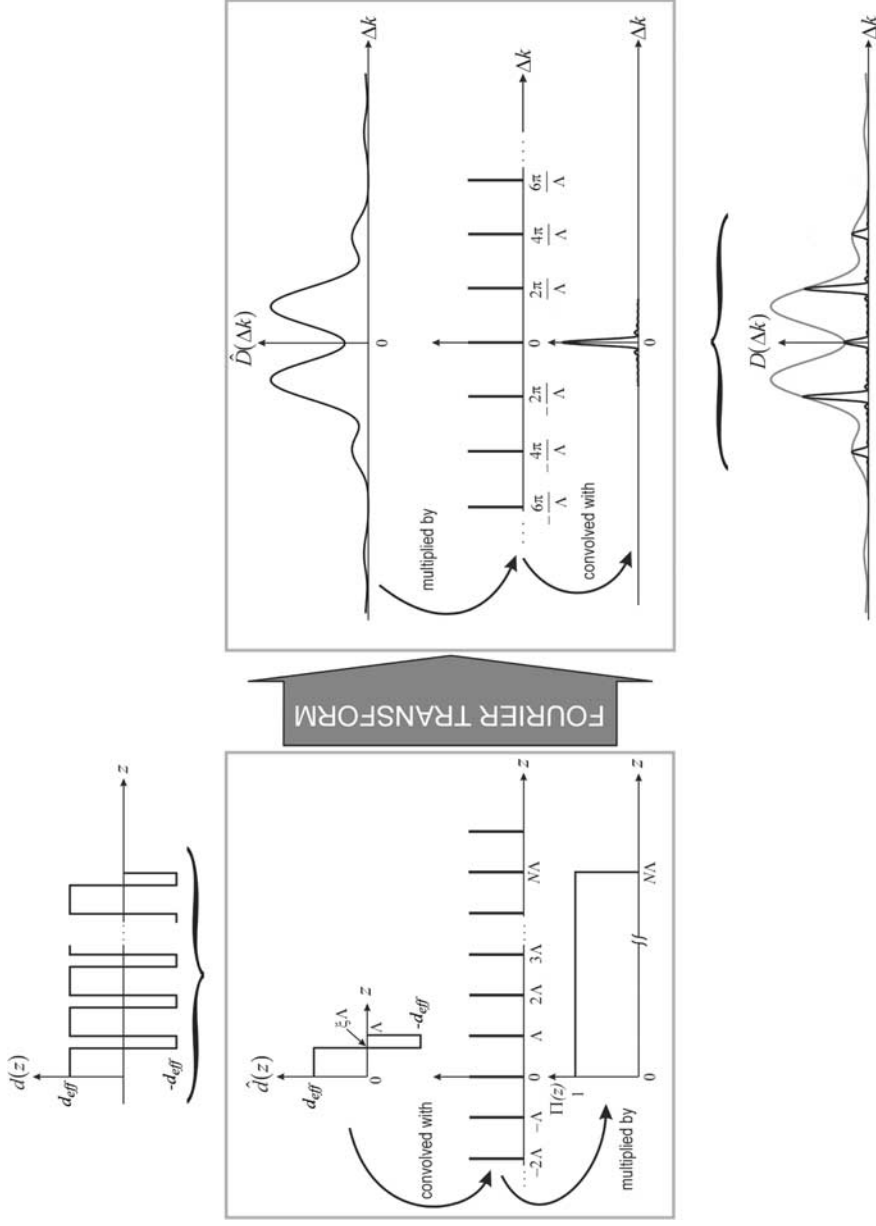


Figure 2.45: Determination of the quantity $D(\Delta k)$ corresponding to the spatial variation of nonlinear coefficient $d(z)$.

The integral has the familiar appearance of a Fourier transform, where the transform variables are z and Δk . This transform relates the propagation constant mismatch Δk to the variation of the nonlinear coupling along the direction of propagation. The maximum conversion efficiency will be obtained where $D(\Delta k)$ is maximum.

If $d(z)$ is constant, that is, $d(z) = d_{\text{eff}}$ over the crystal length, we have the result that:

$$D(\Delta k) = l d_{\text{eff}} e^{j \frac{\Delta k l}{2}} \text{sinc}\left(\frac{\Delta k l}{2}\right) \quad (2.73)$$

which, will give us the same result we obtained earlier for uniform coupling (this is demonstrated in the online supplement). In this case, $D(\Delta k)$ has its maximum value when $\Delta k = 0$. However, if $d(z)$ varies with z , the maximum value of $D(\Delta k)$ can be shifted to a nonzero value of Δk , which is what we desire for QPM. As an example, consider a periodic modulation of $d(z)$, in which the sign of $d(z)$ reverses with spatial period Λ (Figure 2.45). We can “synthesize” this $d(z)$ by convolving the spatial variation over one period, $\hat{d}(z)$, with an infinite series of δ functions spaced Λ apart: $\sum_{n=-\infty}^{\infty} \delta(z - n\Lambda)$. We can then truncate this pattern by multiplying it with the Π function depicted in Figure 2.45, where $l = N\Lambda$. In the Fourier domain, the corresponding operations are multiplication of the Fourier transform of $\hat{d}(z)$, $\hat{D}(\Delta k)$, by an infinite series of delta functions spaced by $2\pi/\Lambda$ in the Δk domain. The function $\hat{D}(\Delta k)$ therefore acts as an envelope to give the weightings of the δ functions. Multiplication by the Π function in the z domain corresponds to convolution in the Δk domain with a sinc function of width $\approx 2\pi/l = 2\pi/N\Lambda$. If, as is usually the case, N is very large (l is typically several *millimeters*, while Λ is typically several *micrometers*, so that $N \sim 1000$), these sinc functions will be very narrow compared to the spacing between them, so that each δ function in the infinite series is simply replaced by a sinc. The end result for $D(\Delta k)$ is a series of sinc functions, centered on $\Delta k = 2\pi m/\Lambda$, where $m = \dots, -3, -2, -1, 0, 1, 2, 3, \dots$.

We can easily calculate $\hat{D}(\Delta k)$ for the particular $\hat{d}(z)$ shown in Figure 2.45. The result is:

$$\hat{D}(\Delta k) = d_{\text{eff}} \Lambda \left[2\xi e^{j \frac{\Delta k \xi \Lambda}{2}} \text{sinc}\left(\frac{\Delta k \xi \Lambda}{2}\right) - e^{j \frac{\Delta k \Lambda}{2}} \text{sinc}\left(\frac{\Delta k \Lambda}{2}\right) \right] \quad (2.74)$$

where, as shown in the figure, ξ is the fraction of the period Λ for which $\hat{D}(\Delta k) > 0$. We are interested in the values of this function at $\Delta k = 2m\pi/\Lambda$:

$$\hat{D}(2m\pi/\Lambda) = \begin{cases} 2d_{\text{eff}} \Lambda \xi e^{jm\pi\xi} \text{sinc}(m\pi\xi) & \text{if } m \neq 0 \\ d_{\text{eff}} \Lambda (2\xi - 1) & \text{if } m = 0 \end{cases} \quad (2.75)$$

Note that, as claimed earlier, the maximum value of $D(\Delta k)$ occurs for $\Delta k \neq 0$. For the example depicted in Figure 2.45, in which $\xi = \frac{2}{3}$, $D(\Delta k)$ is maximum for $\Delta k = 2\pi/\Lambda$.

In so-called “first-order QPM,” we intentionally create a spatial periodicity in $d(z)$ in order to produce a component of $D(\Delta k)$ at $\Delta k = 2\pi/\Lambda$ to assist in satisfying the phasematching condition. In order to determine the value of ξ that maximizes the component at $\Delta k = 2\pi/\Lambda$, we can differentiate the expression for $|\hat{D}(2\pi/\Lambda)|$ given above with respect to ξ and set the result equal to zero. We find that $\xi = \frac{1}{2}$ maximizes the size of the component at $\Delta k = 2\pi/\Lambda$; that is, the duty factor of the periodic modulation should be 50/50 to maximize the efficiency of first-order QPM. Under these conditions, we find that $\hat{D}(2\pi/\Lambda) = j(2/\pi)d_{\text{eff}}\Lambda$, and thus that $|D(2\pi/\Lambda)| = (2/\pi)d_{\text{eff}}l$. If $d(z)$ is uniform, we would have $|D(0)| = d_{\text{eff}}l$. Thus, for first-order QPM, we can define an effective nonlinear coefficient for QPM: $d_{\text{QPM}} = (2/\pi)d_{\text{eff}}$.

Exercise: The interested reader may wish to consider the following questions:

- (1) What is the optimum duty cycle for other orders of QPM?
- (2) Rather than periodically switching the sign of the nonlinear coefficient from $+d_{\text{eff}}$ to $-d_{\text{eff}}$, we can also achieve QPM by periodically destroying the nonlinearity of the material. In other words, after a coherence length of material with nonlinear coefficient d_{eff} , we would have a coherence length of material with zero nonlinearity. What is the effective nonlinear coefficient for this case?

More complex variations of $d(z)$ can tailor the amplitude and phase response of $D(\Delta k)$ to produce interesting and desirable effects. For example, aperiodic gratings can be used to produce a frequency-dependent phase for $D(\Delta k)$ that can be used to compensate for the phase variation of a chirped input pulse (Arbore *et al.*, 1997). The resulting cancellation of phase terms causes a compression of the second-harmonic pulse. The phasematching response with respect to wavelength can be tailored by causing the domain inversions to conform to a repeated 13-bit Barker code having the pattern $+ - + - + + - - + + + + +$, rather than the standard pattern $+ - + - + - + - + - + -$ for a uniform grating (Bortz *et al.*, 1994). The result of this modulation was a phasematching bandwidth 15 times broader than that of a uniform grating of the same length (Figure 2.46). The price paid for this increased bandwidth, however, was a factor of 10 reduction in conversion efficiency. Structures derived from a Fibonacci sequence have been investigated for tailoring certain characteristics of the phasematching spectrum (Zhu *et al.*, 1997), as have devices having multiple QPM periods (Mizuuchi *et al.*, 1994a).

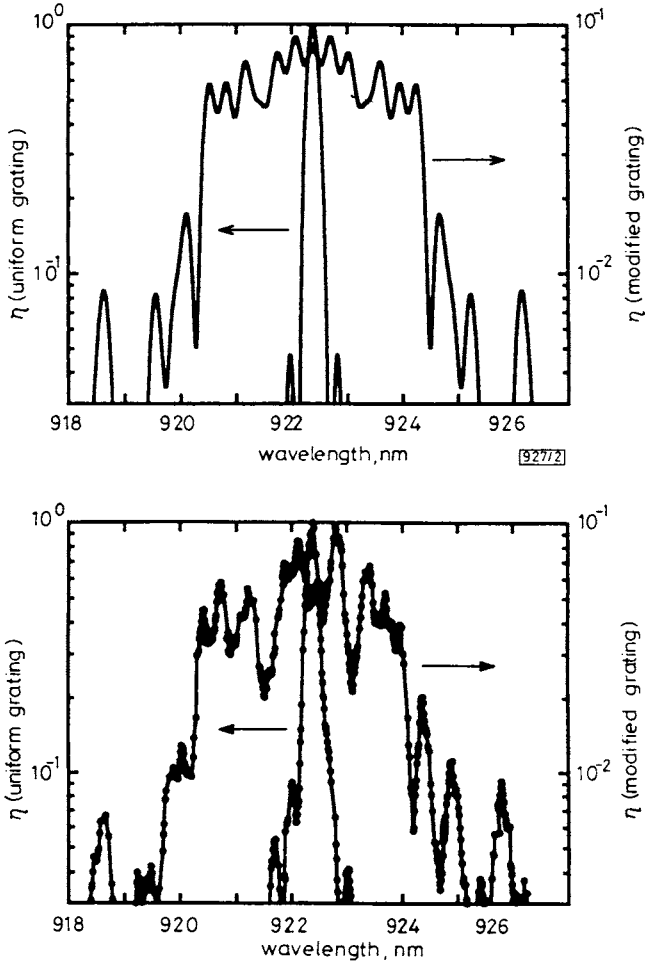


Figure 2.46: Calculated (above) and measured (below) second-harmonic response with respect to wavelength for a uniform QPM grating and one having Barker encoding. (Reprinted with permission from Bortz *et al.*, (1994).)

2.4.3.4 Phasematching tolerances for QPM with an ideal grating

Once we have produced a QPM structure designed to implement a particular SHG or SFG interaction, we are faced with the same question that we considered previously for birefringent phasematching: “How sensitive is the phasematching condition to changes in wavelength, temperature, or direction of propagation?” The analysis for the QPM case follows along exactly the same lines as that for birefringent phase-matching. The main difference is that the phasematching condition now involves a term corresponding to the domain-inverted grating. For example, in the case of

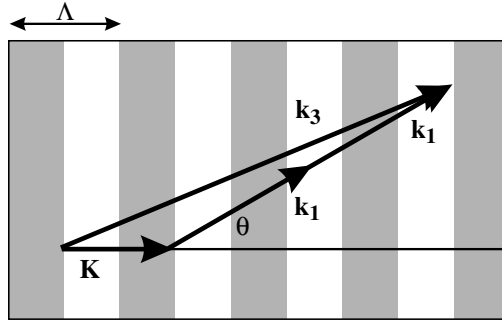


Figure 2.47: Geometry for analyzing SHG phasematching tolerances for a bulk QPM crystal. collinear SHG, we would have:

$$\Delta k = k_3 - 2k_1 - K \quad (2.76)$$

where $K = 2\pi m / \Lambda$, Λ is the period of the periodic poling and m is the order of the phasematching. Here, we briefly present the analogs to the equations derived previously for birefringent phasematching, comment on the differences between these and the equations in that section, and calculate some sample values. In discussing birefringent phasematching, we used KN as a canonical example; here we will use periodically-poled KTP (PPKTP) (Risk 1996). The geometry for this interaction is shown in Figure 2.47.

Example: Wavelength tolerance in PPKTP The equation for wavelength tolerance corresponding to Equation (2.66) is

$$\Delta\lambda_{FWHM} = \left| \frac{1.39\lambda_{pm}}{\pi l \left[\frac{1}{2} \frac{\partial n_3}{\partial \lambda} \Big|_{\lambda=\lambda_{pm}/2} - \frac{\partial n_1}{\partial \lambda} \Big|_{\lambda=\lambda_{pm}} - \frac{n_3 - n_1}{\lambda_{pm}} \right]} \right| \quad (2.77)$$

Here, $n_3 = n_z(\lambda/2)$ and $n_1 = n_z(\lambda)$, where for KTP z is the principal axis corresponding to the crystallographic c -direction. Comparing this expression with Equation (2.66), we note the addition of the last term in the denominator. For birefringent phasematching (Type I) we would have $n_3 - n_1 = 0$, and this term would vanish.

Using the Sellmeier equation for KTP given in Table 2.3, with period $\Lambda = 3.9 \mu\text{m}$ and $\lambda_{pm} = 837 \text{ nm}$, we calculate $\Delta\lambda_{FWHM} = 7.47/l$, where $\Delta\lambda_{FWHM}$ is in ångströms and l is in millimeters. Therefore, for a 5-mm long QPM region, we would expect $\Delta\lambda_{FWHM} = 1.5 \text{ Å}$. The measured bandwidth for a PPKTP crystal having these parameters is 1.1 Å , in good agreement with the calculation.

Example: Temperature tolerance in PPKTP The equation for temperature tolerance corresponding to Equation (2.69) is:

$$\Delta T_{FWHM} = \left| \frac{1.39\lambda_{pm}}{\pi l \left[\left(\frac{\partial n_3}{\partial T} - \frac{\partial n_1}{\partial T} \right) \Big|_{T=T_{pm}} - \alpha(n_3 - n_1) \right]} \right| \quad (2.78)$$

Again, we note that the last term in the denominator would disappear for the case of Type-I birefringent phasematching.

Evaluating this expression for $\lambda_{pm} = 837$ nm and $\Lambda = 3.9$ μm , we obtain $\Delta T_{FWHM} = 12.4/l$, where the units of ΔT_{FWHM} are degrees Celsius and the units of l are millimeters. For a crystal with a periodically-poled region that is nominally 5-mm long, we obtain $\Delta T_{FWHM} = 2.5$ $^{\circ}\text{C}$. The measured temperature bandwidth for a PPKTP crystal having these properties is 2.8 $^{\circ}\text{C}$, which agrees well with the calculated value.

Example: Angle tuning in PPKTP Generally, we choose the spatial period of a QPM structure to provide phasematching for a particular wavelength of interest, and assume propagation perpendicular to the domain-inverted grating ($\theta = 0$, Figure 2.47). If $\theta \neq 0$, phasematching will occur for some longer wavelength. How strongly does the phasematching wavelength λ_{pm} depend on the angle θ for a fixed period Λ ? We find that:

$$\frac{\lambda_{pm}}{\sqrt{[2(n_3 - n_1)]^2 + 8n_3n_1 \left[1 - \sqrt{1 + \left(\frac{\lambda_{pm} \sin \theta}{2\Lambda n_3} \right)^2} \right]}} - \Lambda = 0 \quad (2.79)$$

The phasematching wavelength corresponding to a particular angle θ and a particular QPM period Λ can be found by solving this equation, taking into account the variation of n_1 and n_3 with λ . When $\theta = 0$, we obtain $\lambda_{pm} = \lambda_0 = 2(n_3 - n_1)\Lambda$, as before. We can simplify this expression for small angles, using $\sin \theta \approx \theta$, to obtain:

$$\Delta\lambda = \lambda_{pm} - \lambda_0 \approx \frac{1}{2} \frac{\lambda_0^2}{\Lambda} \frac{n_1}{n_3} \theta^2 \quad (2.80)$$

Figure 2.48 shows a comparison between the exact solution, the approximate solution and experimental data for a PPKTP sample with $\Lambda = 3.9$ μm .

Example: Angle tolerance in PPKTP Suppose now that the wavelength is fixed, and the angle θ is changed – how rapidly does the second-harmonic power decrease? This problem has been analyzed under very general conditions (Fejer *et al.*, 1992). In most cases, we design our QPM structure to provide phasematching at $\theta = 0$;

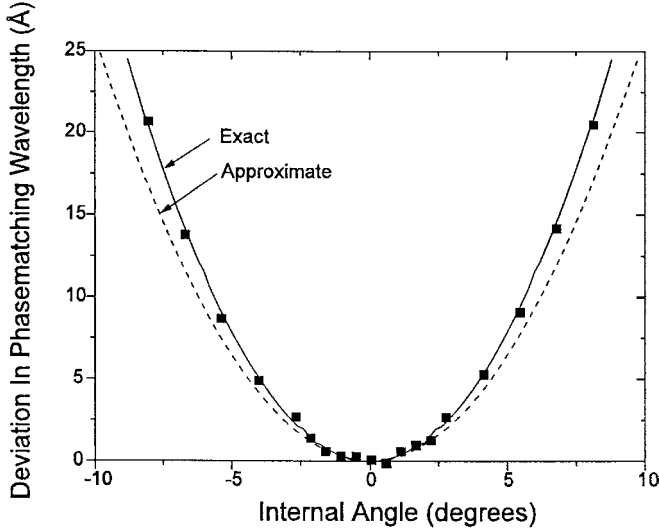


Figure 2.48: Variation of phasematching wavelength with internal angle of propagation in PPKTP. (Reprinted with permission from Risk (1996).)

for this particular case, we have:

$$\Delta\theta_{FWHM} = 1.883 \sqrt{\frac{n_3}{n_1} \frac{\Lambda}{l}} \quad (2.81)$$

Thus, for a 5-mm long PPKTP crystal with $\Lambda = 3.9 \mu\text{m}$, we would obtain $\Delta\theta_{FWHM} = 0.054 \text{ rad} = 3.1^\circ$.

2.4.3.5 Imperfect QPM structures

In the analysis of phasematching tolerances in the previous section, we assumed a perfect first-order QPM structure with a 50/50 duty factor. In practice, it is difficult to produce such a perfect structure. We may have some missing domains – regions that were supposed to invert but did not. We may not achieve a perfect 50/50 duty factor, because of process variability in the fabrication of the periodic electrode or ion-exchange mask, or because of the difficulty in controlling the width of the inverted domain during electric field poling. How do such imperfections affect the performance of a QPM structure? Or, put differently, to what degree must we suppress such imperfections for our device to work adequately well?

Studies of the influence of such imperfections on QPM device performance have appeared in the literature (Helmfrid and Arvidsson, 1991, Fejer *et al.*, 1992). The analysis of such imperfections can be complicated, due to their random nature.

The interested reader is referred to the cited works for a more complete discussion of these effects; here, we simply summarize the results that are most important for practical blue-green light generation.

Missing reversals Occasionally, we find that the perfection of the domain-inverted structure is marred by regions in which we intended to produce an inverted domain, but did not. Thus, instead of producing three domains of length l_c with the pattern $+-+$, we instead produced the pattern $+++$, which is equivalent to a single domain $3l_c$ long. We would like to know how the blue-green output power from such a device compares to what we would have obtained with a perfect device that is otherwise identical to our flawed device. We will define this ratio of output powers as a relative efficiency $\hat{\eta} \equiv I_3/I_{3,ideal}$. If there are M such incorrectly-poled domains in a device with a total of N domains, it can be shown that $\hat{\eta} = (1 - 2f)^2$, where $f = M/N$ is the fraction of improperly-oriented domains. Thus, in order to have $\hat{\eta} > 50\%$, we must have $f < 14.6\%$.

Boundary position errors In our perfect QPM structure, the domain boundaries come precisely at locations $z_i = i\Lambda/2 = iml_c$, for m^{th} -order QPM, where $i = 0, 1, 2, 3, \dots$. If the positions of these boundaries deviate from these ideal locations, such that $z_i = i\Lambda/2 + \Delta_i$, the efficiency of device can be degraded. In order to analyze such a situation, we must make some assumptions about how large these errors, Δ_i , are. We assume them to be random errors, following a normal distribution with zero mean – that is, smaller errors are more common than large ones – the deviated boundary is just as likely to be to the left of its ideal location as to the right, and, when averaged over the entire crystal, the boundaries are about in the right place. We will designate the variance of the normal distribution as σ_Δ . It can be shown that the expectation value of $\hat{\eta}$, $\langle \hat{\eta} \rangle$, for large N is given by:

$$\langle \hat{\eta} \rangle \approx e^{-\left(\frac{2\pi m \sigma_\Delta}{\Lambda}\right)^2} \quad (2.82)$$

Thus, in order to have $\langle \hat{\eta} \rangle > 50\%$, we must have $\sigma_\Delta/l_c < \sqrt{\ln 2}/\pi = 0.22$. In other words, the variance of the boundary position error can be as great as 22% of the coherence length before we would expect a 50% degradation in the performance of the device.

Quantization errors Suppose that we wish to design a QPM device for frequency-doubling of some specific wavelength. We would probably begin by calculating the required period using $\Lambda = 2l_c = 2\pi/\Delta k$. When we design the photomask required to define this period, we may find that the equipment used to produce the mask requires the pattern to be composed of “blocks” with a certain

minimum size laid out on a fixed grid. For example, it may be that the interaction we wish to implement requires a period of $4.5 \mu\text{m}$, but the mask must be laid out such that features must be integral multiples of $1 \mu\text{m}$. This quantization places a limitation on how we can design our mask. If we wanted a $4\text{-}\mu\text{m}$ period, we could make $2\text{-}\mu\text{m}$ lines and $2\text{-}\mu\text{m}$ spaces to achieve it. If we wanted a $5\text{-}\mu\text{m}$ period, we could make $2\text{-}\mu\text{m}$ lines and $3\text{-}\mu\text{m}$ spaces, but the equipment will not let us make $2.5\text{-}\mu\text{m}$ lines and $2.5\text{-}\mu\text{m}$ spaces. And if we wanted a $4.5\text{-}\mu\text{m}$ period, it is not immediately clear how we could achieve it at all under such constraints.

However, there is a way to overcome this problem. Normally, we seek a grating that has N repetitions of the same period. However, if we cannot directly obtain the period we desire because of quantization, we can try the following trick. Suppose we can create a pattern that is M periods long and is composed of blocks that are integer multiples of the quantization size of the mask, such that the average period of this pattern is equal to our desired period – for example, if we have the pattern of line and space widths 3222, the average period of this pattern is $4.5 \mu\text{m}$. Then we repeat this pattern K times, where $N = MK$.

How does the efficiency of such a QPM structure compare with an ideal grating consisting of equal lines and spaces $2.25 \mu\text{m}$ long? It can be shown that:

$$\hat{\eta} = \frac{1}{4M^2} \left[\sum_{i=1}^{2M} (-1)^i \exp \left(j \frac{2\pi m}{\Lambda} z_m \right) \right]^2 \quad (2.83)$$

Thus, if we take $\Lambda = 4.5 \mu\text{m}$, and take the boundary positions z_i as $z_1 = 3 \mu\text{m}$, $z_2 = 5 \mu\text{m}$, $z_3 = 7 \mu\text{m}$, and $z_4 = 9 \mu\text{m}$, we obtain $\hat{\eta} = 86\%$.

In general, all the tolerances discussed are within the realm of what can be achieved experimentally at the scale required for blue-green generation, and need not pose an unsurmountable limitation to the fabrication of such devices.

2.4.4 Waveguide phasematching

2.4.4.1 Modal dispersion phasematching

When SHG or SFG is carried out in a waveguide, phasematching involves the propagation constants of the guided modes $\beta = 2\pi N_{\text{eff}}/\lambda$, where N_{eff} is an effective refractive index that describes the speed of propagation of the guided mode and is determined by the wavelength, the refractive indices of the materials composing the waveguide, the geometry of the waveguide, whether the polarization of the mode is transverse electric (TE) or transverse magnetic (TM), and the order of the mode.

Since the effective refractive indices depend on a number of factors, several new parameters are available which can be adjusted in an attempt to achieve phase-matching. For example, Figure 2.49 shows the effective refractive index for TE

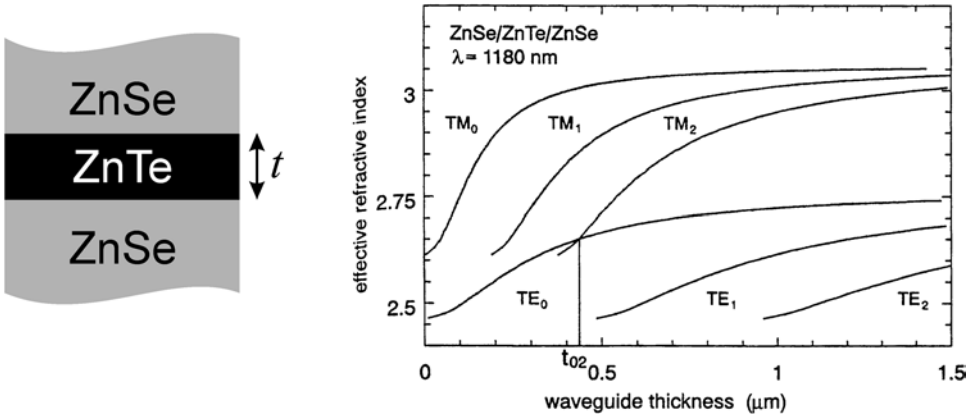


Figure 2.49: Effective refractive indices of a symmetrical ZnSe/ZnTe/ZnSe slab waveguide as a function of the thickness t of the ZnTe layer. At the particular thickness $t_{02} = 0.419 \mu\text{m}$, the effective refractive index of the TE_0 mode at $\lambda_1 = 1180 \text{ nm}$ matches the effective refractive index of the TM_2 mode at $\lambda_3 = 590 \text{ nm}$. (Dispersion curves reprinted by permission from Wagner *et al.* (1995). Copyright: American Institute of Physics.)

modes at 1180 nm and for TM modes at 590 nm for a planar waveguide consisting of a thin layer of ZnTe sandwiched between two thick layers of ZnSe (Wagner *et al.*, 1995). When the thickness of the ZnTe layer is $0.42 \mu\text{m}$, the condition that $N_{\text{TE}_0}(1180 \text{ nm}) = N_{\text{TM}_2}(590 \text{ nm})$ is obtained – that is, 1180-nm light in the TE_0 mode travels at the same speed as 590-nm light in the TM_2 mode of the waveguide. If the materials possess the appropriate component of the nonlinear tensor, we can achieve phasematched SHG of 590-nm light in this way. Note that the constituent materials of the waveguide, ZnSe and ZnTe, are cubic semiconductors and have no birefringence. Thus, modal dispersion in a waveguide allows us to achieve phase-matching using materials for which phasematching cannot normally be achieved.

In this ZnTe/ZnSe example, the second harmonic was generated in the TM_2 mode. When modal dispersion alone is used to achieve phasematching, it is typical for the second-harmonic or sum-frequency beam to emerge in a higher-order mode. The second harmonic tends to travel more slowly than the fundamental because of material dispersion; thus, the second harmonic needs to be sped up by placing it in a higher-order waveguide mode, which travels faster than a lower-order mode. However, it is generally desirable that the blue-green light emerge in the lowest-order mode, particularly in demanding applications like optical data storage, where focusing to a diffraction limited spot is required.

One way that excitation of the lowest-order mode in the blue-green can be achieved is by fabricating the waveguide in a materials like LN, LT, or KTP, in which an additional mechanism such as birefringence or periodic poling can assist in satisfying the phasematching condition. However, even when birefringence or

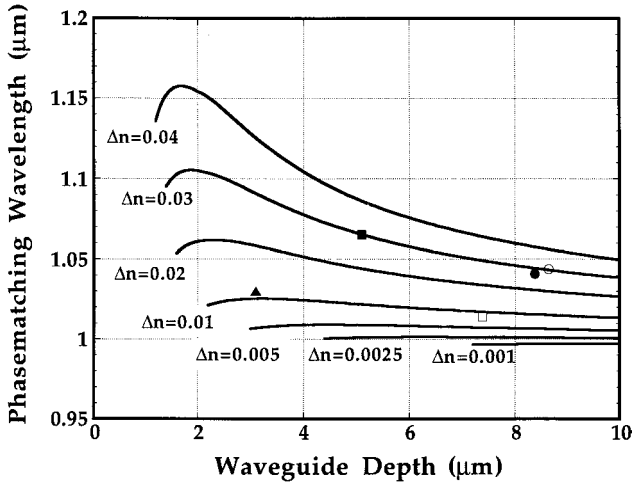


Figure 2.50: Calculated value of the phasematching wavelength λ_{pm} versus waveguide depth d for various values of the surface refractive index change Δn . Individual points show values of Δn and d estimated from measured data for waveguides fabricated in hydrothermally-grown KTP (solid symbols) and flux-grown KTP (open symbols) under the following conditions: ($\blacktriangle$): 100% RbNO_3 , 350 °C, 4 h; ($\blacksquare$): 80% RbNO_3 /20% $\text{Ba(NO}_3)_2$, 350 °C, 2 min; ($\bullet$): 80% RbNO_3 /20% $\text{Ba(NO}_3)_2$, 350 °C, 5 min; ($\square$): 100% RbNO_3 , 350 °C, 45 min; ($\circ$): 95% RbNO_3 /5% $\text{Ba(NO}_3)_2$, 350 °C, 45 min. (Reprinted with permission from Risk *et al.* (1993). Copyright: American Institute of Physics.)

periodic poling is the dominant phasematching mechanism, the properties of the waveguide can still exert a strong influence on the phasematching behavior. Bulk KTP provides Type-II birefringent phasematching for generation of an x -polarized second harmonic from a fundamental wave at 994 nm polarized at 45° to the x - and z -axes, with propagation along the y -axis. If we carry out the analogous interaction in a channel waveguide fabricated in the same KTP crystal by $\text{K} \leftrightarrow \text{Rb}$ ion-exchange, we find that the phasematching wavelength shifts to 1018 nm, even though the blue-green light is generated in the lowest-order spatial mode of the waveguide. Figure 2.50 shows that the value of λ_1 that satisfies the phasematching condition is strongly dependent on the characteristics (such as depth and refractive index profile) of the waveguide, and may vary widely depending on the specifics of the waveguide fabrication process.

Of course, if the blue-green output is not required to have a lowest-order spatial mode, modal dispersion can be used in combination with crystal birefringence to achieve a shorter wavelength than is possible using the lowest-order mode at the second-harmonic. Figure 2.51 shows an example in which interactions of the type $\text{TE}_0(\omega_1) + \text{TM}_0(\omega_1) \rightarrow \text{TE}_q(2\omega_1)$ are considered in a planar waveguide in KTP. Here, the birefringence of the KTP dominates the phasematching condition, but the modal dispersion also plays a significant role. In this waveguide, the excitation of the

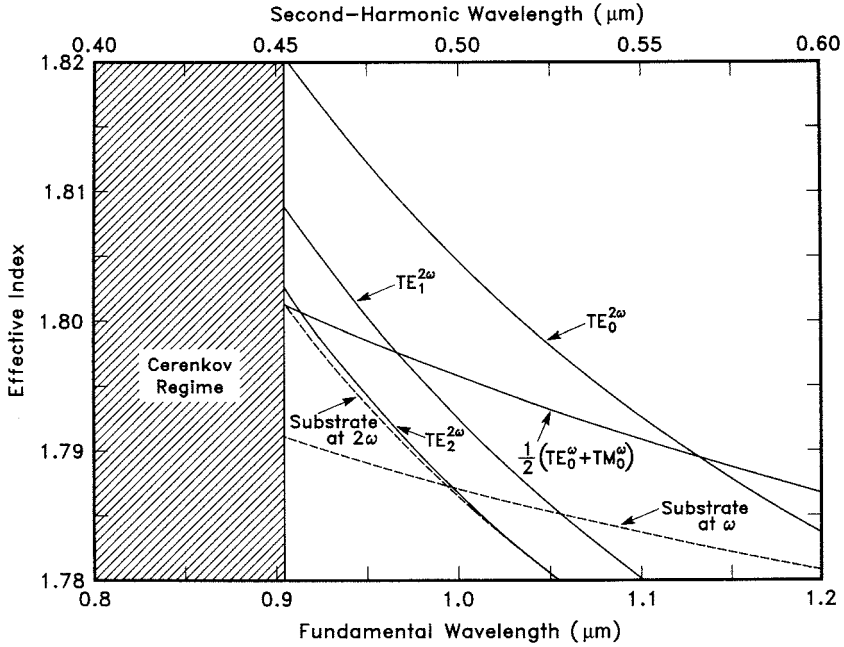


Figure 2.51: Phasematching characteristics for frequency-doubling in a planar KTP waveguide. Lower horizontal scale refers to the fundamental wavelength; upper horizontal scale gives the corresponding second-harmonic wavelength. (Reprinted with permission from Risk (1991). Copyright: American Institute of Physics.)

TE_0 mode at the second harmonic is expected to occur for a fundamental wavelength of 1130 nm. However, excitation of the TE_1 mode at the second harmonic occurs at 966 nm, and of the TE_2 mode at 913 nm. Thus, shorter wavelengths can be achieved by allowing the second harmonic to emerge in a higher-order mode. In addition to the disadvantage in the spatial properties of the output mode, the efficiency generally becomes worse the higher the order of the output mode, because of a reduced overlap integral (Section 2.3.2).

2.4.4.2 Balanced phasematching

In bulk KTP, noncritical phasematching for propagation along the y-axis occurs for a fundamental wavelength of 994 nm, while in a Rb:KTP channel waveguide having certain particular characteristics the phasematching wavelength is pushed out to 1018 nm. For $\lambda_1 > 994$ nm in bulk KTP, we have $\Delta n < 0$, while in the waveguide for $\lambda_1 < 1018$ nm, we have $\Delta N_{eff} > 0$. Therefore, for wavelengths $994 \text{ nm} < \lambda_1 < 1018 \text{ nm}$, propagation for a distance l_1 in a waveguide produces a *positive* phase mismatch $\Delta\beta = 2\pi \Delta N / \lambda_1$ which can be cancelled by allowing propagation through a short section of length l_2 of undoped KTP with *negative* phase mismatch

$\Delta k = 2\pi \Delta n / \lambda_1$, such that $\Delta \beta l_1 + \Delta k l_2 = 0$. This idea is called “balanced phase-matching” and was demonstrated in segmented KTP waveguides by (Bierlein *et al.* (1990)). These “segmented” waveguides are channel waveguides periodically interrupted by gaps $\sim 2 \mu\text{m}$ long. Even though the interacting waves are unguided in these gaps, low-loss waveguiding can still occur through the entire segmented structure (Li and Burke, 1992). Balanced phasematching is useful because it adds flexibility to the design of waveguides that use lowest-order mode interactions. By selecting the segment lengths l_1 and l_2 (and hence the segmentation period $\Lambda = l_1 + l_2$) so that the condition $\Delta \beta l_1 + \Delta k l_2 = 0$ is satisfied, phasematching can be achieved over a wide range of wavelengths, limited at the two extremes by the phasematching wavelength for a continuous (that is, nonsegmented) channel waveguide ($l_2 = 0$) and that of the bulk material ($l_1 = 0$).

2.4.4.3 Čerenkov phasematching

If we examine Figure 2.51 again, we see that as the order of the mode we seek to excite at the second harmonic increases, the effective refractive index of the second-harmonic mode, $N_{TE_q}(2\omega_1)$, comes closer and closer to the refractive index of the substrate at the second harmonic, $n_x(2\omega_1)$. This behavior occurs because higher-order modes are less tightly confined by the guiding structure, and a larger fraction of their energy propagates in the substrate; thus, as the order q of the mode increases, the effective index $N_{TE_q}(2\omega_1)$ approaches the substrate index $n_x(2\omega_1)$.

If we adjust the fundamental wavelength so that

$$\frac{1}{2} [N_{TE_0}(\omega_1) + N_{TM_0}(\omega_1)] < n_x(2\omega_1)$$

the guided fundamental mode can no longer excite a guided second-harmonic mode, but it can excite a radiation mode that freely propagates in the substrate. Since the source at ω_1 is travelling faster than the second-harmonic wave it generates, this process is referred to as “Čerenkov” SHG.

While there is a finite number of guided modes at the second harmonic that are supported by the waveguide structure, there is a continuum of radiation modes. The existence of this continuum ensures “automatic” phasematching for any wavelength shorter than the Čerenkov limit. The blue-green Čerenkov wave is generated at the angle θ_c that satisfies the phasematching condition (Figure 2.52). When a channel waveguide is used, the blue-green radiation typically emerges with a characteristic crescent pattern in the far field (Figure 2.53). This spatial distribution is a disadvantage for most applications, although it is possible to correct it with a suitably designed hemi-conic lens (Tatsuno *et al.*, 1992). However, it also been demonstrated that if the Čerenkov angle is very small, the spatial distribution and

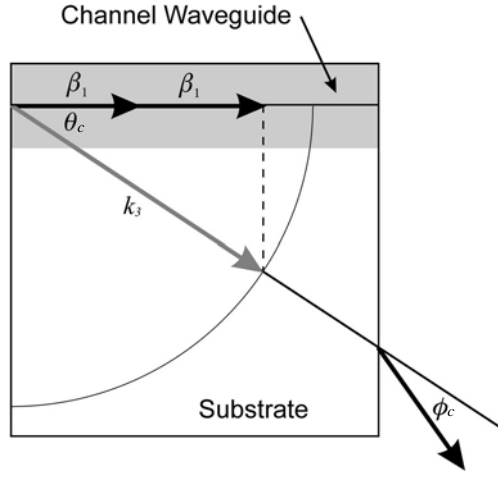


Figure 2.52: “Automatic” phasematching for Čerenkov SHG.

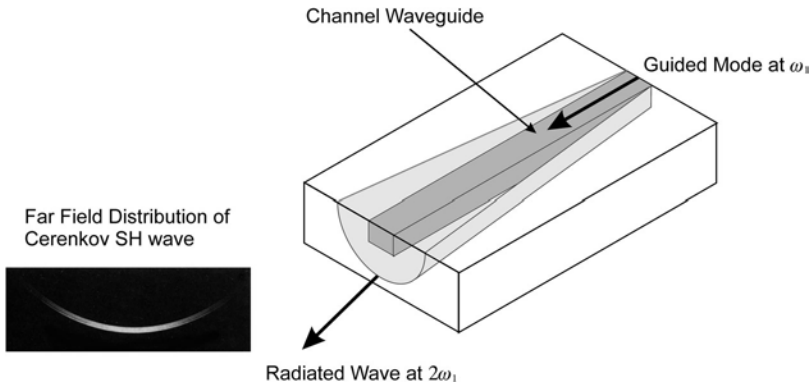


Figure 2.53: The Čerenkov wave emitted due to SHG in a channel waveguide emerges with a characteristic crescent shape, as shown in inset. (Inset reprinted with permission from Yamamoto *et al.* (1991). Copyright: American Institute of Physics.)

conversion efficiency improve dramatically (Doumuki *et al.*, 1994). QPM can be used in combination with Čerenkov phasematching to achieve a low angle of emission (Ito *et al.*, 1993).

One important distinction between guided-mode and Čerenkov phasematching is that for an all guided-mode interaction, the conversion efficiency scales as l^2 , while for a Čerenkov interaction, the conversion efficiency scales as l . This difference arises because, in the guided-mode case, the second-harmonic contribution generated at one section of the guide can constructively interfere with that generated at other points in the guide, whereas, in Čerenkov phasematching, the second-harmonic contribution generated at one section radiates away into the substrate.

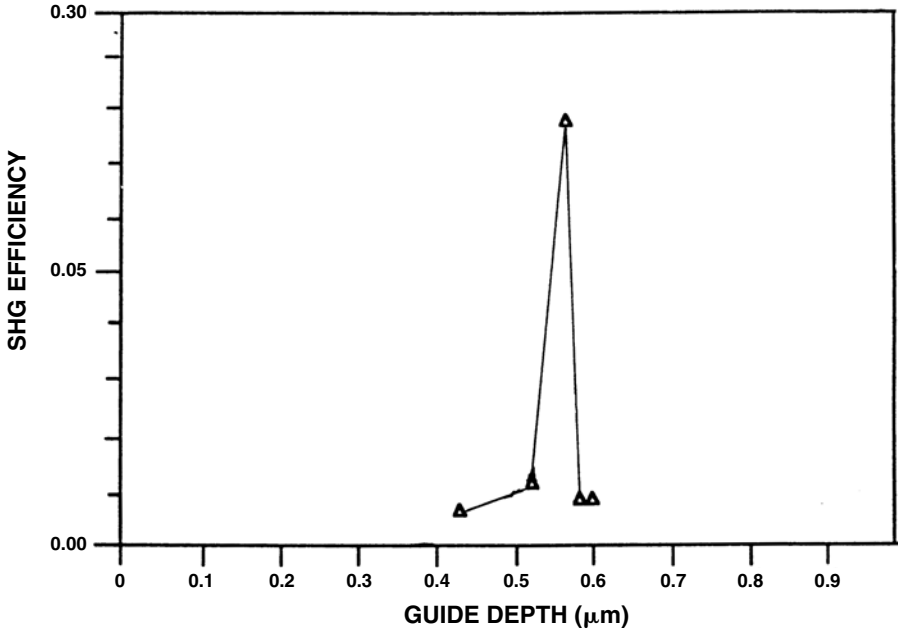


Figure 2.54: Experimental measurement of Čerenkov SHG efficiency as a function of waveguide depth, with the fundamental in the TM_{00} mode at 1064 nm. (Reprinted with permission from Sanford and Connors (1989). Copyright: American Institute of Physics.)

One problem that has plagued Čerenkov SHG is the extreme sensitivity of the conversion efficiency to the thickness of the waveguide. Figure 2.54 shows the measured variation of Čerenkov SHG efficiency with waveguide depth for LN waveguides.

2.4.4.4 Noncritical waveguide phasematching

Sensitivity of the phasematching condition to the geometry of the waveguide is a factor not only in Čerenkov schemes, but in devices involving all-guided-wave interactions. For example, Figure 2.50 shows that relatively small changes in the depth of the waveguide or the index step can cause rather large changes in the phasematching wavelength. Thus, the characteristics of the waveguide must be very uniform over the entire length of the guide in order to maintain phasematching. However, we note that for a given value of Δn , say $\Delta n = 0.04$, the plot of phasematching wavelength λ_{pm} versus depth d achieves a maximum when the depth is approximately 1.8 μm . At that value of d , $\partial\lambda_{pm}/\partial d = 0$, so that variations in the depth of the waveguide have a minimal effect upon λ_{pm} .

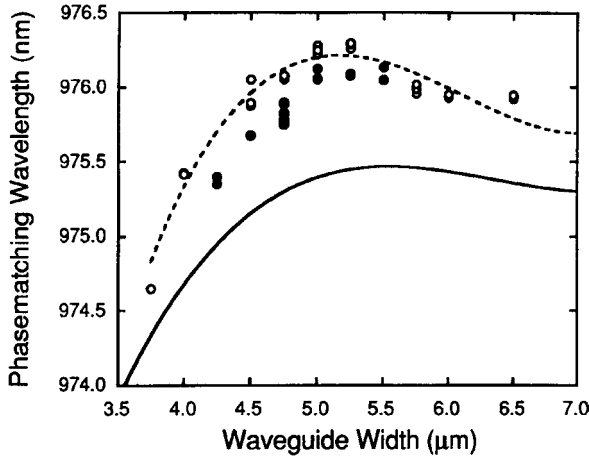


Figure 2.55: Measured SHG phasematching wavelengths versus width of a Ti-in-diffused LN channel waveguide. The open and filled data points were taken at different ambient temperatures. The dashed line is merely a guide for the eye; the solid curve is the result of a modelling calculation. (Reprinted with permission from Bortz *et al.* (1994a). Copyright: IEEE.)

This observation leads to the idea that one can design a waveguide for frequency-doubling such that phasematching is relatively insensitive to perturbations in the geometry of the guiding structure. The existence of noncritical phasematching arrangements in waveguides was pointed out in the early 1970s (Burns and Andrews, 1973, Burns and Lee, 1974) and in the late 1980s and early 1990s, the demand for blue-green sources reinvigorated interest in waveguides for SHG and SFG, stimulating renewed interest in the subject (Lim *et al.*, 1990, Cao *et al.*, 1991a). A good example of this modern work is shown in Figure 2.55. Bortz *et al.* (1994a) fabricated a series of proton-exchanged LN waveguides of different widths. Small perturbations in the width of the guide result from imperfect photolithography or variations in the waveguide fabrication process. The variation of phasematching wavelength with width shows the existence of a noncritical design. The corresponding wavelength tuning curves (Figure 2.56) show that the waveguide with the width corresponding to the maximum in Figure 2.55 has the most sinc^2 -like tuning curve and the greatest conversion efficiency.

2.4.5 Other phasematching techniques

2.4.5.1 Anomalous dispersion

This phasematching technique exploits the perturbation of the refractive index near an absorption peak in the nonlinear material, as illustrated in Figure 2.57. Most

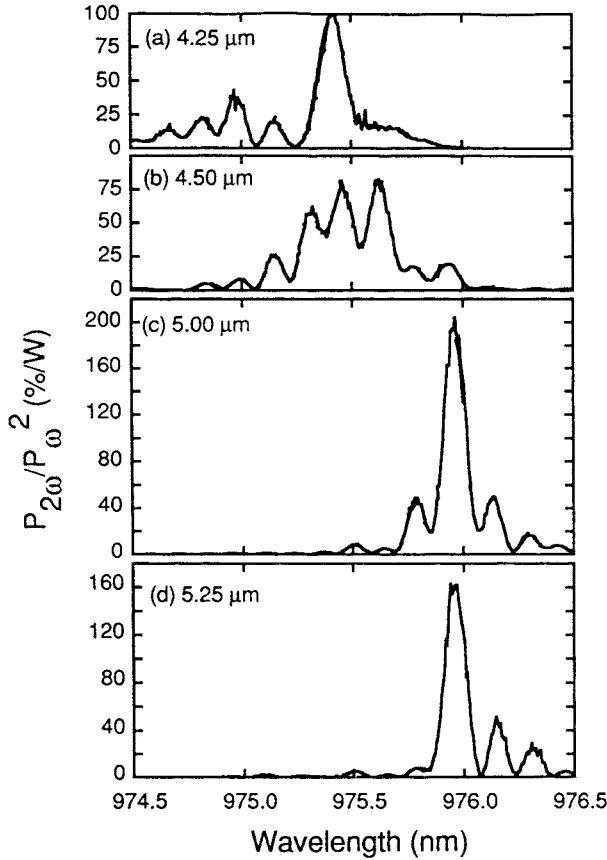


Figure 2.56: Measured wavelength tuning curves for waveguides having the widths shown. The shape of the tuning curve resembles the ideal sinc^2 function only for the width satisfying the dimensionally noncritical condition. (Reprinted with permission from Bortz *et al.* (1994a). Copyright: IEEE.)

typical nonlinear materials exhibit absorption in the deep blue or ultraviolet portion of the spectrum, such that both the infrared fundamental and its second harmonic are at longer wavelengths. In this regime, $\partial n/\partial \lambda$ is negative; this is what we call “normal” dispersion. However, near the absorption there is a region of “anomalous” dispersion where $\partial n/\partial \lambda$ is positive.

Under certain circumstances, this anomalous dispersion can be used to achieve phasematching. For example, the polymeric material PMMA (poly-methyl-methacrylate) has normal dispersion in the visible and near-infrared portions of the spectrum. If an absorbing dye is introduced into PMMA, the anomalous dispersion associated with the absorption feature can increase the refractive index on the long-wavelength side of the absorption while decreasing it on the short-wavelength side (Figure 2.58). Thus, if the absorption wavelength falls between

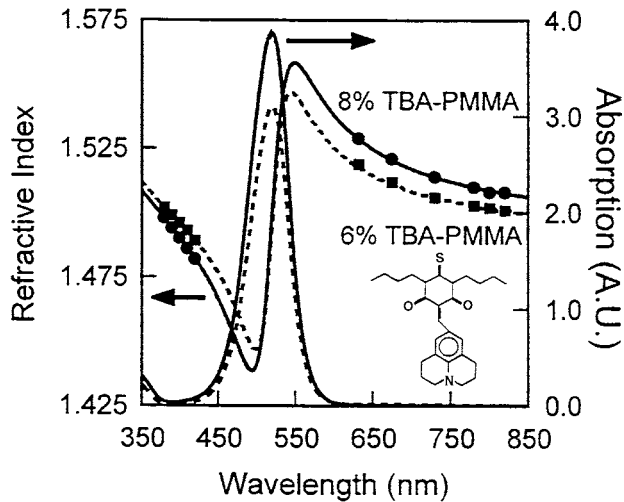


Figure 2.57: Refractive index and absorption of PMMA doped with the nonlinear optical moiety TBA. (Reprinted with permission from Kowalczyk *et al.* (1996).)

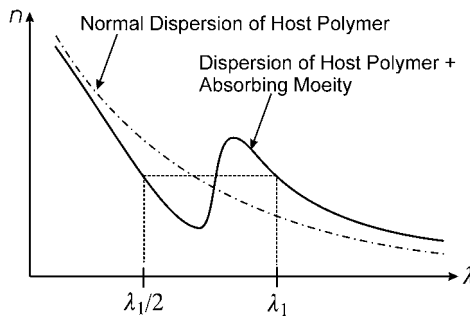


Figure 2.58: How anomalous dispersion can be used to achieve phasematching for SHG using the nonlinear material shown in Figure 2.57.

the fundamental and second-harmonic wavelengths in a frequency-doubling experiment, the anomalous dispersion of the dye can cancel the normal dispersion of the host polymer to produce phasematching. This has been demonstrated experimentally by Kowalczyk *et al.* (1996), who introduced the dye TBA (thiobarbituric acid) into PMMA to achieve anomalous phasematching for SHG of 800 nm light.

2.4.5.2 Counterpropagating waves

Another approach to phasematching uses counterpropagating waves. The momentum of the two infrared waves cancels along the direction of their propagation;

to conserve overall momentum, the second harmonic must emerge in two beams normal to the infrared ones. This approach was demonstrated in LN waveguides by Normandin and Stegeman (1979). More recently, this approach has been used with semiconductor waveguides, which permits convenient integration with a semiconductor laser (Dai *et al.*, 1992). However, because the emitted light emerges from a long, narrow region (essentially a line source), these devices have not found wide application as blue-green light sources for the applications described here, but they do have a number of interesting uses as optical correlators and spectrometers (Normandin *et al.*, 1991, Vakhshoori and Wang, 1991).

2.4.5.3 Total internal reflection

When light propagating inside a transparent dielectric experiences total internal reflection at an interface between that dielectric and another medium of lower refractive index (for example, air), the light wave receives a phase shift. This phase shift can be used to compensate the phase difference accruing from propagation in the nonlinear medium. Thus, phasematching can be achieved by repeatedly reflecting the interacting waves back and forth between the parallel walls of a slab of nonlinear material such that after one passage across the slab and one reflection, the net relative phase shift between the two waves is zero.

This concept was proposed early in the development of nonlinear optics (Armstrong *et al.*, 1962, Boyd and Patel, 1966), and more recently has been revisited (Komine *et al.*, 1998).

2.4.5.4 Form birefringence

A layered medium can exhibit birefringence, even though the materials of which it is composed are optically isotropic. This phenomenon, called “form birefringence”, arises because the boundary conditions experienced by an electromagnetic wave propagating in this structure are different when the electric field vector is perpendicular to the layers versus parallel to them. In 1975, van der Ziel proposed using this effect for phasematching, and presented designs for a layered structure of GaAs and GaP for SHG of 10.6- μm radiation and of ZnS and fused silica for SHG of 1064-nm light (van der Ziel, 1975). However, these designs were not experimentally demonstrated.

More recently, there has been renewed interest in using form birefringence for phasematching. Fiore *et al.* (1997) developed a layered structure of GaAs and AlAs, in which the AlAs is oxidized in order to decrease the refractive index of those layers. The resulting device had a very large birefringence of 0.22. Such a device has been used for phasematched difference-frequency generation of $\sim 5\ \mu\text{m}$ radiation (Bravetti *et al.*, 1998).

2.4.6 Summary

In this section, we have examined a variety of different techniques that can be used in bulk materials or in guided-wave configurations to achieve phasematching. In bulk materials, the two most important techniques are birefringent phasematching and QPM. In waveguides, we saw that in addition to using guided-wave versions of birefringent phasematching and QPM, we can also use modal dispersion phasematching, balanced phasematching, and Čerenkov phasematching. The reader should also be aware of other phenomena, such as anomalous dispersion, counterpropagation, total internal reflection, and form birefringence, that can be used to achieve phasematching, even though they have not yet been extensively exploited for blue-green generation.

Whatever the phasematching technique, the properties of the nonlinear material determine what approaches will work and how well. In the next section, we examine nonlinear materials that are useful for blue-green light generation.

2.5 MATERIALS FOR NONLINEAR GENERATION OF BLUE-GREEN LIGHT

2.5.1 Introduction

In this section, we overview some of the nonlinear materials that have been used in the pursuit of compact blue-green lasers. We will emphasize those materials that have been most important and most widely used, but will also mention a few that are a little more exotic, in order to present the reader with a more complete perspective and suggest areas of ongoing research. For each material, we present an overview of the ways it has been used for blue-green generation and a table summarizing its characteristics. In these tables, space constrains us to give priority to those properties that are most relevant to blue-green light generation – such as refractive index, nonlinear coefficient, etc. – but where possible, we have also included references to more obscure properties. The reader is also referred to the article by Bordui and Fejer (1993) and the book by Dmitriev *et al.* (1997) for further information on the properties of nonlinear materials.

2.5.2 Lithium niobate (LN)

LN has been one of the most widely used and extensively developed nonlinear materials. In the area of blue-green generation, it has been used in both bulk-crystal and waveguide forms with both birefringent phasematching and QPM.

2.5.2.1 Birefringent phasematching

The phasematching properties of LN depend on its composition and growth techniques. LN crystals used in early SHG experiments were grown by the Czochralski

method (Ballman, 1965) from melts containing ~ 50 mol% Li_2O and had nonuniform composition which caused inconsistencies in the phasematching temperature (Fay *et al.*, 1968). In addition, LN exhibits “optical damage”, a photorefractive phenomenon that causes the spatial distortion of an input laser beam and is particularly severe for visible wavelengths (Ashkin *et al.*, 1966). The effects of optical damage can be ameliorated by heating the crystal above the “annealing” temperature ($\sim 180^\circ\text{C}$).

Most of the more recent experiments in blue-green generation have used either *congruent* LN or *MgO-doped* LN. Congruent LN, in which the same concentration (48.38 ± 0.015 mol% Li (Bordui *et al.*, 1991a)) exists in both the melt and the crystallized solid, can be grown in large boules that are extremely uniform in composition and optical properties (Byer *et al.*, 1970, Bordui *et al.*, 1991a). The birefringence of such crystals is sufficient to provide noncritical phasematching for SHG of 1064-nm light at a temperature of about -8°C . Although the phasematching temperature is below the annealing temperature, optical damage has been observed to saturate at sufficiently high powers so that high-efficiency SHG can occur (Ammann and Guch, 1988).

Adding a few percent of MgO to the melt from which LN is grown has been found to significantly reduce optical damage (Zhong *et al.*, 1980), as a result of a hundredfold increase in the photoconductivity (Bryan *et al.*, 1984). Nightingale and coworkers used crystals grown from a congruent melt with 5 mol% of MgO added for SHG and achieved 50% conversion efficiency for SHG of a 1064-nm Nd:YAG laser at a phasematching temperature of 107°C , although they also noted nonuniformities in the crystals which they attributed to variations in the MgO concentration (Nightingale *et al.*, 1986).

Another approach to overcoming some of these problems involves increasing the lithium content of congruent crystals using a vapor transport process. This produces a higher phasematching temperature, while avoiding some of the nonuniformities associated with growth from a noncongruent melt. Luh *et al.* (1987) used this approach to increase the Li content of single-crystal LN fibers to something close to 50 mol%, and achieved a phasematching temperature of 238°C . Jundt *et al.* (1990) applied a similar process to plates of congruent LN to increase the phasematching temperature. Figure 2.59 shows the phasematching wavelengths for different LN crystal compositions, and illustrates that vapor-transport-equilibrated LN (VTE-LN) provides phasematching for a shorter wavelength at a given temperature than do the other compositions.

Birefringent phasematching has also been used for sum-frequency generation in bulk LN. Moosmüller and Vance (1997) mixed the outputs of two Nd:YAG lasers, one operating at 1064 nm and one operating at 1319 nm, in congruent LN heated to 227.5°C to achieve noncritically phasematched generation of 589-nm light.

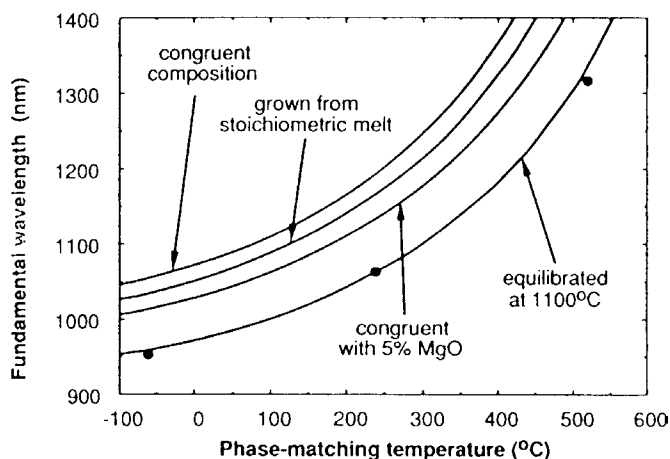


Figure 2.59: Fundamental wavelength for birefringent phasematching vs. temperature for various compositions of LN. (Reprinted with permission from Jundt *et al.* (1990). Copyright: IEEE.)

2.5.2.2 Bulk QPM

Bulk crystals of LN have been periodically poled using electron beam scanning (Haycock and Townsend, 1986, Keys *et al.*, 1990, Yamada and Kishima, 1991, Ito *et al.*, 1991, Fujimura *et al.*, 1992, 1992a, Nutt *et al.*, 1992), application of an electric field to a periodic electrode, and by periodic modulation during growth (Feng *et al.*, 1980, Zheng *et al.*, 1998). The second approach has been the most popular in more recent experiments, and will be described further below. Several groups have reported that periodically-poled crystals exhibit less susceptibility to optical damage than uniformly poled material (Magel *et al.*, 1990, Jundt *et al.*, 1991, Webjörn *et al.*, 1994, Pruner *et al.*, 1995, Taya *et al.*, 1996).

In 1993, Yamada *et al.* (1993) reported producing periodic domain inversion in congruent LN by applying a voltage to a periodic metal electrode deposited on the $+c$ face of the crystal, which also had a uniform electrode on the $-c$ face. By working with a very thin wafer of LN, they were able to apply an electric field that exceeded the coercive value (~ 20 kV/mm) without causing dielectric breakdown of the material. Burns *et al.* (1994) later reported periodic poling of 0.25-mm thick wafers, which were sufficiently thick to be used for bulk SHG, and demonstrated third-order QPM SHG using a focused beam of 937-nm light. A related approach uses an electrolytic liquid, rather than a metal, as an electrode (Webjörn *et al.*, 1994). First-order QPM frequency-doubling of Q-switched 1064-nm light with 55% conversion efficiency has been demonstrated using a crystal prepared by this technique (Webjörn *et al.*, 1995). Miller and coworkers conducted a detailed study of electric field poling in LN (Miller *et al.*, 1996) and demonstrated 42% single-pass

conversion of the 6.5-W cw, 1064-nm output of a Nd:YAG laser using electric-field-poled LN (Miller *et al.*, 1997). Ross *et al.* (1998) reported generation of 450 mW of average power at 473 nm using PPLN for frequency-doubling of a 946-nm Nd:YAG laser. Batchko *et al.* (1999) demonstrated a “backswitching” technique that gives good control over the definition of short-period gratings for blue light generation and produced 60 mW of blue light at 460 nm in a 50-mm long device with a 4- μm period, for a conversion efficiency of 6.1%/W.

Several groups have also explored electric-field poling of MgO-doped LN (Kuroda *et al.*, 1996, Kurimura *et al.*, 1996). Mizuuchi *et al.* (1996) fabricated a periodically-poled grating with a 3.7- μm period for first-order QPM of 874-nm light, and demonstrated generation of 1 mW of 437-nm blue light from 330 mW of infrared in a 10-mm length. Harada and Nihei (1996) used a corona discharge approach to fabricate a 6-mm long domain-inverted grating with a period of 5.2 μm in 0.4-mm thick MgO:LN to provide first-order QPM for SHG of a 980-nm high-power laser diode, and achieved 6.7 mW of second harmonic.

2.5.2.3 Waveguides

While some experiments have investigated modal phasematching in LN waveguides (Fejer *et al.*, 1986), most of the more recent work has concentrated on periodically-poled waveguides. In LN, most such devices are made by a two-step approach. In the first step, a domain-inverted grating is produced, either by one of the bulk poling techniques described above, or by a process such as titanium indiffusion (Miyazawa, 1979, Lim *et al.*, 1989, 1989a, Armani *et al.*, 1992) or Li_2O outdiffusion (Nakamura *et al.*, 1987, Webjörn *et al.*, 1989, 1989a, Fujimura *et al.*, 1991) which produce gratings that are relatively shallow, but deep enough to provide QPM for a guided-wave interaction.

Annealed proton exchange (Veselka and Bogert, 1987) has become established as the preferred method for waveguide fabrication. In this approach, the LN substrate is immersed in a molten bath of (typically) benzoic or pyrophosphoric acid at temperatures around $\sim 200^\circ\text{C}$. Because of the relatively low temperatures involved, this process can be carried out after domain inversion without causing significant depoling. Approximately 70–80% of the lithium ions in a thin layer near the surface are replaced by hydrogen ions, thereby increasing the extraordinary refractive index by $\Delta n_e \sim 0.1$, but lowering the ordinary refractive index by $\Delta n_o \sim -0.04$; thus, for a device fabricated on a z -cut substrate, TM modes are guided while TE modes are not. Such a configuration is therefore compatible with the typical QPM interaction in which the d_{33} nonlinear coefficient is used. Titanium-indiffused waveguides confine both TE and TM modes, but require higher processing temperatures, exhibit significant photorefractive problems at blue-green

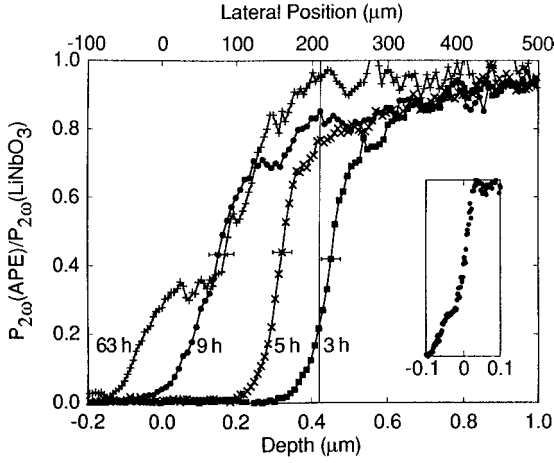


Figure 2.60: Variation of nonlinearity of proton-exchanged layer of LN versus depth, showing recovery of d_{33} near the surface with annealing. (Reprinted with permission from Bortz *et al.* (1993).)

wavelengths (Becker, 1984, Fejer *et al.*, 1986) and are more susceptible to optical damage (Holman and Cressman, 1982, Glavas *et al.*, 1989).

The nonlinear coefficients of LN can be adversely affected by proton exchange. Early SHG measurements on proton-exchanged layers showed a substantial reduction in effective nonlinearity, although the reported magnitude of this reduction varied greatly (a value of essentially zero was reported by Laurell *et al.* (1992a) and by Bortz and Fejer (1992), whereas a value of $\sim 50\%$ that of the original LN was reported by Suhara *et al.* (1989)). Annealing of proton-exchanged samples was reported by some to partially restore the nonlinearity (Keys *et al.*, 1990a, Cao *et al.*, 1991), while others reported that this procedure did not appear to be effective at all (Laurell *et al.*, 1992a), and still others reported recovery under some circumstances but not under others (Hsu *et al.*, 1992).

Careful measurements of the depth profile of the d_{33} coefficient for proton-exchanged waveguides annealed for various times showed that although annealing can cause the recovery of some of the nonlinearity at the depth where most of the energy of a guided mode is confined, it is never fully restored at the surface (Figure 2.60) (Bortz *et al.*, 1993). Bortz and Fejer (1991) have also reported a detailed characterization of the linear properties (such as effective index, mode intensity profiles) for waveguides fabricated by the APE technique.

More recent work has further clarified the complex phase structure of proton-exchanged LN, particularly in regard to SHG response (Korkishko *et al.*, 2000). “Soft” proton-exchange processes that produce waveguides with nonlinear characteristics essentially the same as the bulk material have been investigated and used with PPLN for parametric fluorescence experiments (Chanvillard *et al.*, 2000).

Some properties of LN are given in Table 2.1.

Table 2.1. LN

Refractive indices	
Congruent material (Edwards and Lawrence, 1984)	Stoichiometric material (Hobden and Warner, 1966)
$n_o^2 = 4.9048 + \frac{0.11775 + 2.2314 \times 10^{-8} F}{\lambda^2 - (0.21802 - 2.9671 \times 10^{-8} F)^2} + 2.1429 \times 10^{-8} F - 0.027153\lambda^2$	$n_o^2 = 4.9130 + \frac{1.173 \times 10^5 + 1.65 \times 10^{-2} T^2}{\lambda^2 - (2.12 \times 10^2 + 2.7 \times 10^{-5} T^2)^2} - 2.78 \times 10^{-8} \lambda^2$
$n_e^2 = 4.5820 + \frac{0.099215 + 5.2716 \times 10^{-8} F}{\lambda^2 - (0.21090 - 4.9143 \times 10^{-8} F)^2} + 2.2971 \times 10^{-8} F - 0.021940\lambda^2$	$n_e^2 = 4.5567 + \frac{0.97 \times 10^5 + 2.70 \times 10^{-2} T^2}{\lambda^2 - (2.01 \times 10^2 + 5.4 \times 10^{-5} T^2)^2} - 2.24 \times 10^{-8} \lambda^2$
$F = (T - T_o)(T + T_o + 546)$	Temperature T in kelvins, wavelength λ in μm
Temperature T in $^{\circ}\text{C}$, $T_o = 24.5^{\circ}\text{C}$, wavelength λ in μm	
Lithium-rich material (Jundt <i>et al.</i> , 1990)	MgO-doped material (Zelmon <i>et al.</i> , 1997)
$n_o^2 = 4.913 + \frac{1.163 \times 10^5 + 9.4 \times 10^{-3} F}{\lambda^2 - (2.201 \times 10^2 + 3.98 \times 10^{-5} F)^2} + 1.6 \times 10^{-8} F - 2.73 \times 10^{-8} \lambda^2$	$n_o^2 - 1 = \frac{2.2454\lambda^2}{\lambda^2 - 0.01242} + \frac{1.3005\lambda^2}{\lambda^2 - 0.05313} + \frac{6.8972\lambda^2}{\lambda^2 - 331.33}$
$n_e^2 = 4.546 + \frac{9.17 \times 10^4 + 1.93 \times 10^{-2} F}{\lambda^2 - (2.148 \times 10^2 + 5.3 \times 10^{-5} F)^2} + 2.72 \times 10^{-7} F - 3.03 \times 10^{-8} \lambda^2$	$n_e^2 - 1 = \frac{2.4272\lambda^2}{\lambda^2 - 0.01478} + \frac{1.4617\lambda^2}{\lambda^2 - 0.05612} + \frac{9.6536\lambda^2}{\lambda^2 - 371.216}$
$F = (T - 24.5)(T + 570.5)$	Wavelength λ in μm
Temperature T in $^{\circ}\text{C}$, wavelength λ in μm	

Nonlinear properties

d tensor (Boyd <i>et al.</i>, 1964) Symmetry class $3m$ $\begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$					
d coefficients (Shoji <i>et al.</i> (1997), except where noted: (a): Choy and Byer (1976), (b) Eckardt <i>et al.</i> (1990))					
		1313 nm	1064 nm		852 nm
Congruent LN	d_{31}	3.2	4.6	5.95 (a)	4.8
	d_{33}	19.5	25.2	34.4 (a)	25.7
	Δ_{31}	0.054	0.073	0.1 (a)	0.067
	Δ_{33}				
1% MgO:LN	d_{31}	3.2	4.6		4.8
	d_{33}	20.3	24.9		27.5
	Δ_{31}	0.054	0.073		0.068
	Δ_{33}	0.412	0.473		0.468
5% MgO:LN	d_{31}	3.4	4.4	−4.7 (b)	4.9
	d_{33}	20.3	25.0		28.4
	Δ_{31}	0.058	0.071		0.069
	Δ_{33}	0.419	0.481		0.491

Miscellaneous properties

Spontaneous polarization: (Camlibel, 1969)

$P_s = 0.71 \text{ C/m}^2$

Electro-optic coefficients: (Onuki *et al.*, 1972)
 $r_{13} = 10.9 \pm 1.0 \text{ pm/V}$, $r_{33} = 34.0 \pm 2.5 \text{ pm/V}$
(Evaluated at room temperature)

Thermal expansion coefficients (Kim and Smith, 1969)

$\frac{\Delta l_x}{l_x} = (1.44 \times 10^{-5})\Delta T + (7.1 \times 10^{-9})(\Delta T)^2$
 $\frac{\Delta l_y}{l_y} = (1.59 \times 10^{-5})\Delta T + (4.9 \times 10^{-9})(\Delta T)^2$
 $\Delta T = T - 25^\circ\text{C}$

2.5.3 Lithium tantalate (LT)

2.5.3.1 Birefringent phasematching

Although LT has been widely used for electro-optic and surface acoustic wave applications, its small birefringence ($\sim 4 \times 10^{-3}$ at 1064 nm, compared with $\sim 8 \times 10^{-2}$ for LN) has precluded its use for birefringent phasematching. However, with the advent of periodic poling, LT has received a great deal of attention as a material for QPM. In particular, LT has attracted interest for ultraviolet generation, since it is transparent to a shorter wavelength than LN (short-wavelength absorption edge ~ 280 nm, compared with ~ 330 nm for LN) (Kase and Ohi, 1974). Optical damage has also been reported to be less severe in LT than LN (Tangonan *et al.*, 1977).

2.5.3.2 Bulk periodically-poled LT (PPLT)

Early experiments reported the possibility of creating domain-inverted regions in bulk LT with scanning electron beams (Ballman and Brown, 1972, Haycock and Townsend, 1986, Hsu and Gupta, 1992) or electric fields applied to heated (Matsumoto *et al.*, 1991) or room-temperature crystals (Zhu *et al.*, 1995, Mizuuchi and Yamamoto, 1995). Mizuuchi and Yamamoto used a periodic metal electrode to create periodic domain inversion, but they also subjected the sample to proton exchange between the fingers of the periodic electrode in order to inhibit spreading of the domains. They were able to achieve a very uniform $3.8\text{-}\mu\text{m}$ period grating over 10 mm long in a 0.2-mm thick sample of LT, and generated 3.8 mW of blue power at 425 nm from 300 mW of infrared. Chemical patterning techniques have also been used to pole bulk LT (Mizuuchi and Yamamoto, 1996, Baron *et al.*, 1996).

LT has been of particular interest for frequency-doubling of red lasers, since the transmission is somewhat higher for the second harmonic than for LN. Mizuuchi *et al.* (1997) generated 340-nm ultraviolet light using a 0.15-mm thick sample with a period of $1.7\text{ }\mu\text{m}$ for first-order QPM SHG of a red diode laser ($30\text{ }\mu\text{W}$ at 340 nm from 50 mW at 680 nm, $l = 10$ mm interaction length, $\eta_{\text{norm}} = 1.2\%/W\text{-cm}$). Meyn and Fejer (1997) demonstrated generation of wavelengths as short as 325 nm in 0.2-mm thick LT poled by the electric field method (second-order QPM, $l = 0.3$ mm interaction length, $P_{SH} = 11.5\text{ }\mu\text{W}$, $P_F = 133$ mW of red light, $\eta_{\text{norm}} = 0.22\%/W\text{-cm}$).

2.5.3.3 Waveguides

Waveguides in LT for quasi-phasematched frequency-doubling have generally been made by a two-step process, in which domain inversion is accomplished by either

electric-field poling or proton exchange and then a waveguide is added in a second step. Methods for waveguide fabrication in LT are somewhat limited by the relatively low Curie temperature ($\sim 600^\circ\text{C}$) of the material. Although indiffusion at high temperatures has been explored (summarized in Findakly *et al.* (1988)), as have more exotic approaches (such as strain induction (Eknoyan *et al.*, 1992) and ion implantation (Townsend, 1990)), proton exchange (Spillman *et al.*, 1983, Atuchin and Zakhar'yash, 1984, Tada *et al.*, 1987, Yamamoto *et al.*, 1992a) has emerged as the preferred technique, since it can be carried out well below the Curie temperature and produces waveguides of good optical quality.

Gupta *et al.* (1994) achieved a high conversion efficiency of $\sim 290\%/W\text{-cm}^2$ using an e-beam to produce periodic poling in a room-temperature LT crystal and then adding a waveguide. Baron *et al.* (1996) used a chemical patterning process combined with electric-field poling to produce a grating with $3.6\text{-}\mu\text{m}$ period in 0.5-mm thick LT and used 7-mm long waveguides fabricated on the $-c$ face of the crystal to generate 3 mW of 424 nm light.

Shallow domain inversion can be produced in lithium tantalate by subjecting the $-c$ face to proton exchange, then annealing near the Curie temperature (Nakamura and Shimizu, 1990). Mizuuchi *et al.* (1991) used a similar process to produce a domain-inverted grating with a period of $20\text{ }\mu\text{m}$ for third-order QPM. The inverted domains formed by this process were shallow ($\sim 3\text{ }\mu\text{m}$) and had a semicircular cross-section. They formed a channel waveguide using a second proton-exchange step and demonstrated guided-wave SHG using a Ti:S laser, generating 0.13 mW of light at 421 nm . Åhlfeldt *et al.* (1991) obtained similar results. Continued optimization of this third-order device by the Matsushita group led to generation of 12 mW of blue light at 424 nm from 99 mW of fundamental power in a 9-mm long waveguide, for a normalized conversion efficiency of $37\%/W\text{-cm}^2$ (Yamamoto *et al.*, 1992). By carefully studying the details of the domain inversion process, Mizuuchi *et al.* (1994) were later able to produce a first-order grating ($\Lambda = 4\text{ }\mu\text{m}$) and generate 23 mW of 435 nm light from 115 mW of fundamental power. First-order waveguide devices based on this approach were reported to generate 23 mW of 435 nm light from 115 mW of fundamental power (Mizuuchi *et al.*, 1994).

As with LN, proton exchange degrades the nonlinearity of LT; however, efficient SHG can be restored by annealing (Hsu *et al.*, 1992, Åhlfeldt *et al.*, 1993, Åhlfeldt, 1994). Gradual changes in the refractive indices of proton-exchanged waveguides over time have been noted (Matthews and Mickelson, 1992, Åhlfeldt *et al.*, 1994), and this causes a change in the phasematching wavelength of QPM devices (Åhlfeldt and Laurell, 1995). Degradation of the SHG conversion efficiency over time has also been reported (Yamamoto *et al.*, 1996).

Some properties of LT are given in Table 2.2.

Table 2.2. *LT*

Refractive indices					
Abedin and Ito (1996)					
$n_o^2 = 4.51224 + \frac{0.0847522 - 9.66449 \times 10^{-9} F}{\lambda^2 - (0.19876 + 8.815 \times 10^{-8} F)^2} + 4.25637 \times 10^{-8} F - 0.0239046 \lambda^2$ $n_e^2 = 4.5299 + \frac{0.0844313 + 1.72995 \times 10^{-7} F}{\lambda^2 - (0.20344 - 4.7733 \times 10^{-7} F)^2} - 8.31467 \times 10^{-8} F - 0.0237909 \lambda^2$ $F = (T - T_o)(T + T_o + 546)$ Temperature T in $^{\circ}\text{C}$, $T_o = 25\text{ }^{\circ}\text{C}$, wavelength λ in μm					
Meyn and Fejer (1997) (derived from periodically-poled material, gives more accurate result than that of Abedin and Ito when generated wavelength is blue or ultraviolet).					
$n_e^2 = 4.5284 + \frac{7.2449 \times 10^{-3} + 2.6794 \times 10^{-8}(T + 273.15)^2}{\lambda^2 - [0.2543 + 1.6234 \times 10^{-8}(T + 273.15)^2]^2}$ $+ \frac{7.7690 \times 10^{-2}}{\lambda^2 - (0.1838)^2} - 2.3670 \times 10^{-2} \lambda^2$ Temperature T in $^{\circ}\text{C}$, wavelength λ in μm					
Nonlinear properties					
$\begin{matrix} & d \text{ tensor} \\ & \text{Symmetry class } 3m \\ \begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix} \end{matrix}$		d coefficients (Shoji <i>et al.</i> , 1997)			
			1313 nm	1064 nm	852 nm
		d_{31}	—	0.85	—
		d_{33}	10.7	13.8	15.1
		Δ_{31}	—	0.018	—
		Δ_{33}	0.228	0.278	0.276
Miscellaneous properties					
Spontaneous polarization: (Camlibel, 1969) $P_s = 0.50\text{ C/m}^2$		Thermal expansion coefficients (Kim and Smith, 1969)			
Electro-optic coefficients: (Onuki <i>et al.</i> , 1972) $r_{13} = 8.4 \pm 0.9\text{ pm/V}$, $r_{33} = 30.5 \pm 2\text{ pm/V}$		$\frac{\Delta l_x}{l_x} = (1.61 \times 10^{-5})\Delta T + (7.5 \times 10^{-9})(\Delta T)^2$			
(Evaluated at room temperature, $\lambda = 632.8\text{ nm}$)		$\frac{\Delta l_y}{l_y} = (1.54 \times 10^{-5})\Delta T + (7.0 \times 10^{-9})(\Delta T)^2$			
		$\Delta T = T - 25\text{ }^{\circ}\text{C}$			

2.5.4 Potassium titanyl phosphate (KTP)

2.5.4.1 Birefringent phasematching

The properties of KTP crystals vary, depending on which of three techniques is used to produce them. In the hydrothermal method, the crystal is grown from a seed in an aqueous solution of titanium dioxide and potassium phosphate at high temperature (~ 600 °C) and pressure ($\sim 25\,000$ psi) (Gashurov and Belt, 1985). Because of the high pressure and temperature involved, the growth chamber must be relatively small, and the resulting crystals are limited in size. An alternative

hydrothermal growth process involving lower temperatures (~ 350 – 500 °C) and pressures ($\sim 20\,000$ psi) permits a larger chamber to be used and, hence, somewhat larger crystals can be grown (Laudise *et al.*, 1986). Flux growth techniques cause KTP to crystallize from a high-temperature solution at atmospheric pressure, so that large crystals can be grown (Jacco *et al.*, 1984).

In general, crystals grown hydrothermally tend to have lower ionic conductivity and higher resistance to gray tracking than flux-grown crystals (although there are also differences in ionic conductivity between crystals grown using the high- and low-temperature versions of the hydrothermal approach (Morris *et al.*, 1989)). The ionic conductivity is a particularly important factor for electric-field poling and waveguide fabrication, as discussed below. Sellmeier equations determined from measurements on hydrothermally-grown and flux-grown material also show some differences.

KTP crystals have been widely used for frequency-doubling of 1064-nm Nd:YAG lasers using Type-II birefringent phasematching (Liu *et al.*, 1984, Driscoll *et al.*, 1986). Compared with LN, KTP has a lower nonlinearity but greater resistance to optical damage and phasematching for 532-nm generation can be achieved at room temperature. Type-II phasematching is fairly efficient in KTP, but the effective nonlinearities for Type-I phasematching are so small as to make that approach impractical (except in the QPM case as described below).

KTP can provide noncritical phasematching for frequency-doubling of fundamental wavelengths as short as 994 nm, which is achieved by propagation along the *b*-axis (propagation along the *a*-axis provides noncritical phasematching at 1076 nm) (Anthon and Crowder, 1988). KTP heated to 153 °C provides noncritically-phasematched frequency-doubling of a 1079.6-nm Nd:YALO laser (*a*-axis propagation), with very broad phasematching tolerances with respect to the angle and temperature of the crystal (Garmash *et al.*, 1986). Similar broad tolerances have been reported for noncritically-phasematched frequency-doubling of a 994-nm Styryl-13 dye laser at room temperature, as shown in Figure 2.61 (Risk *et al.*, 1989). The temperature tolerance in particular is remarkably broad – the temperature of the crystal can be changed over a 350 °C range for a fixed wavelength without significantly degrading the SHG efficiency.

Birefringent phasematching in KTP can also be used for noncritically-phasematched SFG. Baumert *et al.* (1987) reported generation of 459-nm light by mixing 809-nm and 1064-nm light. This approach to blue light generation was subsequently developed by several groups, as will be discussed in succeeding chapters. Other SFG combinations that have been demonstrated in KTP include 660 nm + 1319 nm $\rightarrow$ 440 nm (noncritically phasematched), in which the 660 nm was obtained by SHG of the 1319-nm diode laser (Stolzenberger *et al.*, 1989) and 780 nm + 1540 nm $\rightarrow$ 520 nm (critically phasematched), in which both wavelengths were produced by diode lasers (Wang and Ohtsu, 1993).

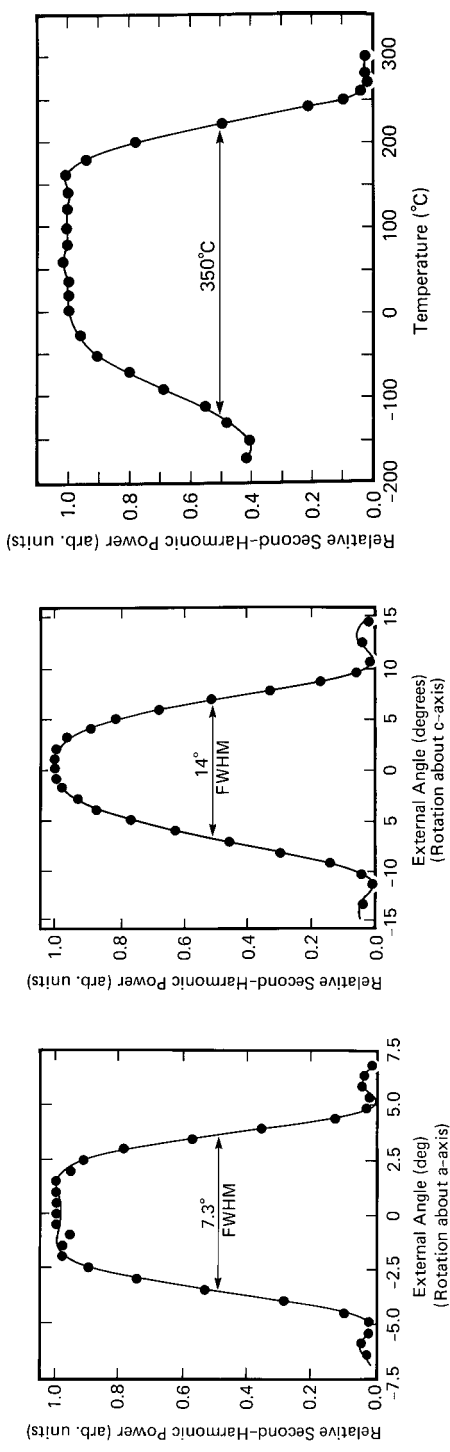


Figure 2.61: Variation of second-harmonic power with angle and temperature for Type-II noncritical phasematching in KTP. Propagation was along the y-axis and the fundamental wavelength was 994 nm. (Reprinted with permission from Risk *et al.* (1989).)

2.5.4.2 Bulk periodically-poled KTP (PPKTP)

Gupta *et al.* (1993) demonstrated that an electron beam could be used for periodic poling of KTP. They produced a 0.5-mm long fifth-order domain-inverted grating (20- μm period) in a 1-mm thick hydrothermally-grown KTP crystal using an electron beam and demonstrated frequency-doubling of Ti:S laser tuned to 840 nm.

Chen and Risk (1994) reported periodic poling of hydrothermally-grown KTP crystal using an electric field applied to a periodic (4- μm period) metal electrode. They observed that the electric-field strength required to produce domain inversion in KTP was only about 2 kV/mm, an order of magnitude lower than the value required for LN. This relatively low electric field permitted poling of thick (~ 1 mm) crystals in the ambient atmosphere without causing breakdown of the surrounding air, making it convenient to monitor the poling process using *in situ* SHG or interferometry.

Flux-grown KTP is less expensive than hydrothermally-grown KTP and is available in larger sizes. However, it has a higher ionic conductivity, which makes electric-field poling more difficult. Rosenman *et al.* (1997) studied the domain inversion process in flux-grown KTP and found conditions under which an applied electric field could produce domain inversion. Oron *et al.* (1997) applied this process to make first-order gratings (6.9- μm period) for SHG of 980-nm lasers in 0.5-mm-thick flux-grown KTP crystals and achieved a bulk SHG efficiency of 4.6%/W-cm. Flux-grown KTP periodically poled by this technique has been used for high-efficiency single-pass SHG of a pulsed, diode-pumped Nd:YAG laser (Englander *et al.*, 1997) and for resonant doubling of a cw Nd:YAG laser (Arie *et al.*, 1998).

Another approach to periodic poling of flux-grown KTP has been reported in which ion-exchange is first used to reduce the conductivity at the surface of a KTP crystal, then an electric field is applied to a periodic electrode in order to create a domain-inverted grating (Karlsson and Laurell, 1997). A grating with a period of 9 μm has been used to demonstrate first-order SHG with a 1064-nm Nd:YAG laser, and this approach generated 2.9 mW of 532-nm light from 890 mW of pump power.

Periodic poling has also been induced by stress using a dielectric cladding (Buritskii *et al.*, 1993).

2.5.4.3 Waveguides

Bierlein *et al.* (1987) first reported fabrication of waveguides in KTP using ion-exchange in molten nitrates of Rb, Cs, and Tl. Of these, ion-exchange in RbNO₃ emerged as the favored process, since it did not require the higher temperatures needed for CsNO₃ exchange or the use of the more hazardous chemical TlNO₃.

The addition of a few percent of a divalent ion salt, such as $\text{Ba}(\text{NO}_3)_2$, to the melt was found to improve the uniformity of the waveguides (Bierlein *et al.*, 1988).

Bierlein *et al.* (1990) demonstrated Type-II frequency-doubling of 1064-nm light using balanced phasematching in a segmented KTP waveguide and achieved an efficiency of $15\%/W\text{-cm}^2$. Later they reported efficient Type-I frequency-doubling of 850-nm light in waveguides with a similar segmented structure and ascribed the high efficiency to domain inversion resulting from the ion-exchange process (van der Poel *et al.*, 1990). The presence of $\text{Ba}(\text{NO}_3)_2$ in the melt seemed to be a crucial factor in producing domain inversion, which was observed in both hydrothermally-grown and flux-grown KTP. They were able to achieve conversion efficiencies of $100\%/W\text{-cm}^2$ for 5-mm long waveguides with a period of 4 μm . Although Khurgin *et al.* (1990) pointed out that effects other than domain inversion could be responsible for Type-I phasematching, further work convincingly established that the ferroelectric domain inversion was indeed responsible for producing QPM (Bierlein *et al.*, 1991, Laurell *et al.*, 1992). Efficiencies as high as $800\%/W\text{-cm}^2$ have been reported in waveguides fabricated by this technique (Eger *et al.*, 1994); however, consistently reproducing high efficiencies has proven difficult because of the sensitivity of the domain inversion process to factors such as crystal properties, melt composition, and processing time and temperature. Laurell *et al.* (1993) later used a combination of balanced phasematching and QPM in the same waveguide to generate ultraviolet (359 nm), blue (421 nm) and green (508 nm) light simultaneously. The physical origin of the domain inversion produced in KTP by the ion-exchange technique has been elucidated by Thomas and Glazer (1991).

Chen and Risk (1996) used the applied electric field method to create periodic domain inversion in a bulk crystal of KTP, then added a waveguide in a second ion-exchange step. They generated 12 mW of blue light from 146 mW of infrared light at 862 nm in a 2.8-mm interaction length, for a normalized conversion efficiency of $720\%/W\text{-cm}^2$. Other guides on the same chip had conversion efficiencies as high as $1100\%/W\text{-cm}^2$.

Risk and Lau (1996) reported a chemical patterning process for KTP, in which the regions of a hydrothermally-grown KTP crystal subjected to ion exchange in a $\text{RbNO}_3/\text{Ba}(\text{NO}_3)_2$ became resistant to poling under the influence of uniform electric field applied to the crystal. They produced a segmented waveguide structure, then used electric-field poling to invert the unexchanged regions between the segments. They used *in situ* waveguide SHG to monitor the poling process and found that it was self-terminating – the SHG output saturated with the number of applied poling pulses, so that precise control of the poling conditions was not necessary.

Other approaches have also been reported for waveguide-based blue-green light generation using KTP. Ion implantation has been used to create waveguides in KTP

for SHG (Zhang *et al.*, 1992, 1992a, 1993). Efficient Čerenkov SHG has been reported in a waveguide formed by depositing a Ta₂O₅ layer atop KTP to form a planar waveguide, then using loading by a SiO₂ ridge to achieve lateral confinement (Doumuki *et al.*, 1994). Type-II modal phasematching involving higher-order modes in KTP planar waveguides has been investigated (Risk, 1991), as has excitation of the lowest-order mode using channel waveguides (Risk *et al.*, 1993).

Some properties of KTP are given in Table 2.3.

2.5.5 Rubidium titanyl arsenate (RTA)

Several isomorphs of KTP have been studied (Stucky *et al.*, 1989), in which the potassium ion has been replaced by another element from Column I of the periodic table and the phosphorus has been replaced by another element from Column V. In particular, substitution of Rb and Cs for K and of As for P have been investigated in an attempt to improve upon the properties of KTP. Of these isomorphs, RTA (RbTiOAsO₄) is perhaps the most interesting for blue-green generation. Flux-grown RTA has a much lower ionic conductivity than flux-grown KTP, comparable to that of hydrothermally-grown KTP (Cheng *et al.*, 1994).

2.5.5.1 Bulk RTA for birefringent phasematching

Cheng *et al.*, (1993) calculated the phasematching wavelengths for Type-II SHG in RTA, and found that the cutoff wavelengths were 1138 nm for propagation along the *b*-axis and 1243 nm for propagation along the *a*-axis (compared with 994 nm and 1076 nm, respectively, for KTP). Thus, RTA is unable to provide phasematching for Type-II SHG of 1064-nm Nd:YAG lasers, and has not been used for blue-green generation with birefringent phasematching.

2.5.5.2 Bulk periodically-poled RTA (PPRTA)

Domain inversion can be produced in RTA using an applied electric field of ~2–3 kV/mm (Hu *et al.*, 1996). Periodic poling can be obtained using a periodic liquid (Karlsson *et al.*, 1996) or metal (Risk and Loiacono, 1996) electrode. Using the former technique, 270 μW of blue light have been generated from 490 mW of fundamental in a 3-mm-long grating with a period of 4.2 μm. Using the latter technique, 184 μW of blue light have been generated from 284 mW of fundamental in a 5-mm-long grating with a period of 3.9 μm. The temperature bandwidth (3.3 °C) and temperature tuning rate of the phasematching wavelength (~0.74 Å/°C) have also been measured.

Table 2.3. *KTP*

Refractive indices	
Flux-grown material (Fan <i>et al.</i> , 1987)	Hydrothermal material (Vanherzeele <i>et al.</i> , 1988)
$n_x^2 = 2.16747 + \frac{0.83733}{1 - 0.04611\lambda^{-2}} - 0.01713\lambda^2$	$n_x^2 = 2.1146 + \frac{0.89188}{1 - 0.04352\lambda^{-2}} - 0.01320\lambda^2$
$n_y^2 = 2.19229 + \frac{0.83547}{1 - 0.04970\lambda^{-2}} - 0.01621\lambda^2$	$n_y^2 = 2.1518 + \frac{0.87862}{1 - 0.04753\lambda^{-2}} - 0.01327\lambda^2$
$n_z^2 = 2.25411 + \frac{1.06543}{1 - 0.05486\lambda^{-2}} - 0.02140\lambda^2$	$n_z^2 = 2.3136 + \frac{1.00012}{1 - 0.05679\lambda^{-2}} - 0.01679\lambda^2$
Wavelength λ in μm	Wavelength λ in μm
Temperature derivatives (Wiechmann <i>et al.</i> , 1993)	
$\frac{\partial n_x}{\partial T} = (1.427\lambda^{-3} - 4.735\lambda^{-2} + 8.711\lambda^{-1} + 0.952)(10^{-6}/^\circ\text{C})$	
$\frac{\partial n_y}{\partial T} = (4.269\lambda^{-3} - 14.761\lambda^{-2} + 21.232\lambda^{-1} - 2.113)(10^{-6}/^\circ\text{C})$	
$\frac{\partial n_z}{\partial T} = (12.415\lambda^{-3} - 44.414\lambda^{-2} + 59.129\lambda^{-1} + 12.101)(10^{-6}/^\circ\text{C})$	

Nonlinear properties		d tensor Symmetry class $mm2$ $\begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{bmatrix}$										d coefficients (Vanherzeele and Bierlein (1992), except where noted. Values at 880 nm are measured, those at 1064 nm are estimated using Miller's rule; (a) Eckardt <i>et al.</i> (1990).) <table><tr><th></th><th colspan="5">d_{ij} (pm/V)</th><th colspan="2">Δ_{ij} (m²/C)</th></tr><tr><th></th><th>d_{15}</th><th>d_{24}</th><th>d_{31}</th><th>d_{32}</th><th>d_{33}</th><th>Δ_{15}</th><th>Δ_{24}</th></tr><tr><td>880 nm</td><td>2.04</td><td>3.92</td><td>2.76</td><td>4.74</td><td>18.5</td><td>—</td><td>—</td></tr><tr><td>1064 nm</td><td>1.91</td><td>3.64</td><td>2.54</td><td>4.35</td><td>16.9</td><td>0.029(a)</td><td>0.025(a)</td></tr><tr><td></td><td>2.6 (a)</td><td>3.3 (a)</td><td></td><td></td><td></td><td></td><td></td></tr></table>											d_{ij} (pm/V)					Δ_{ij} (m ² /C)			d_{15}	d_{24}	d_{31}	d_{32}	d_{33}	$ \Delta_{15} $	$ \Delta_{24} $	880 nm	2.04	3.92	2.76	4.74	18.5	—	—	1064 nm	1.91	3.64	2.54	4.35	16.9	0.029(a)	0.025(a)		2.6 (a)	3.3 (a)					
	d_{ij} (pm/V)					Δ_{ij} (m ² /C)																																																							
	d_{15}	d_{24}	d_{31}	d_{32}	d_{33}	$ \Delta_{15} $	$ \Delta_{24} $																																																						
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	2.6 (a)	3.3 (a)																																																											
Miscellaneous properties																																																													
Spontaneous polarization $P_s = 0.21$ C/m² (Rosenman <i>et al.</i> (1997); from charge transfer during electric field poling. Other measurements giving similar values include those by Bierlein and Arbweiler (1986) and Shaldin <i>et al.</i> (1995))				Thermal expansion coefficients (Bierlein and Vanherzeele, 1989) $\alpha_1 = 11 \times 10^{-6}$ $\alpha_2 = 9 \times 10^{-6}$ $\alpha_3 = 0.6 \times 10^{-6}$				Electro-optic coefficients (Bierlein and Arbweiler, 1986) <table><tr><th>r_{ij} (pm/V)</th><th>Low frequency</th><th>High frequency</th></tr><tr><td>r_{13}</td><td>9.5</td><td>8.8</td></tr><tr><td>r_{23}</td><td>15.7</td><td>13.8</td></tr><tr><td>r_{33}</td><td>36.3</td><td>35.0</td></tr><tr><td>r_{51}</td><td>7.3</td><td>6.9</td></tr><tr><td>r_{42}</td><td>9.3</td><td>8.8</td></tr></table>				r_{ij} (pm/V)	Low frequency	High frequency	r_{13}	9.5	8.8	r_{23}	15.7	13.8	r_{33}	36.3	35.0	r_{51}	7.3	6.9	r_{42}	9.3	8.8																																
r_{ij} (pm/V)	Low frequency	High frequency																																																											
r_{13}	9.5	8.8																																																											
r_{23}	15.7	13.8																																																											
r_{33}	36.3	35.0																																																											
r_{51}	7.3	6.9																																																											
r_{42}	9.3	8.8																																																											

Table 2.4. *RTA*

Refractive indices		Nonlinear properties		Electro-optic properties		
(Cheng <i>et al.</i> , 1994)		d tensor	d coefficients (pm/V) (Cheng <i>et al.</i> , 1994)	r coefficients (pm/V)		
$n_x^2 = 222681 + \frac{0.99616}{1 - 0.048593\lambda^{-2}} - 0.01483\lambda^2$		Symmetry class $mm2$ $\begin{bmatrix} 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 \end{bmatrix}$	d_{31}	r_{13}	13.5	
$n_y^2 = 1.97756 + \frac{1.25726}{1 - 0.041812\lambda^{-2}} - 0.00865\lambda^2$			d_{32}	r_{23}	17.5	
$n_z^2 = 2.28779 + \frac{1.20629}{1 - 0.05515\lambda^{-2}} - 0.01583\lambda^2$			d_{33}	15.8	r_{33}	40.5
Wavelength λ in μm						
Miscellaneous properties						
Spontaneous polarization (Hu <i>et al.</i> , 1996) $P_s = 0.4 \text{ C/m}^2$		Coercive field (Hu <i>et al.</i> , 1996) 2.6 kV/mm				
Curie temperature (Chu <i>et al.</i> , 1993) $T_c = 750^\circ\text{C}$		Thermal conductivity (Ebbers and Velsko, 1996) $\kappa_x = 16 \text{ mW/cm}^\circ\text{C}$, $\kappa_y = 16 \text{ mW/cm}^\circ\text{C}$, $\kappa_z = 17 \text{ mW/cm}^\circ\text{C}$				

2.5.5.3 Waveguides

Fabrication of waveguides in RTA has been reported using ion-exchange in a pure CsNO_3 melt (Risk and Loiacono, 1996). Exchange at 450°C for 30 min produced a waveguide having an exponential refractive index profile, with a depth of $4\text{ }\mu\text{m}$ and a surface index step of 0.023. This process was used to produce channel waveguides in PPRTA and waveguide SHG with a normalized conversion efficiency of $225\%/\text{W}\cdot\text{cm}^2$ was achieved.

Some properties of RTA are given in Table 2.4.

2.5.6 Other KTP isomorphs

The noncritical phasematching wavelength of KTP can be made shorter than 994 nm by altering the composition of the crystal to change the birefringence. $\text{K}_{1-x}\text{Na}_x\text{TiOPO}_4$ crystals can provide noncritical phasematching for frequency-doubling of wavelengths as short as $\sim 981\text{ nm}$ (*b*-axis propagation) (Loiacono *et al.*, 1994). $\text{K}_{1-x}\text{Nb}_x\text{Ti}_{1-x}\text{OPO}_4$ and $\text{K}_{1-x}\text{Ta}_x\text{Ti}_{1-x}\text{OPO}_4$ crystals have also been explored (Cheng *et al.*, 1994a) and used for noncritically phasematched SHG with a 946-nm Nd:YAG laser (Miesak *et al.*, 1995). $\text{KTiOP}_x\text{As}_{1-x}\text{O}_4$ and $\text{Cs}_{1-x}\text{K}_x\text{TiOAsO}_4$ crystals have been studied for non-critically phasematched frequency-doubling of 1064-nm and 1320-nm Nd:YAG lasers (Cheng *et al.*, 1994b).

Other isomorphs of KTP that have been investigated for blue-green light generation include KTA and RTP. Kato (1994) reported that Type-II SHG and sum-frequency mixing in KTA could give wavelengths as short as 517 nm and 419 nm, respectively. Risk and Loiacono (1996a) described waveguide fabrication in KTA, but did not demonstrate frequency-doubling. Pisarevsky *et al.* (1991) used RTP to generate 540 nm light by frequency-doubling of a Nd:YAlO₃ laser. However, since these materials have not been very widely used for blue-green generation, we will not enumerate any further properties here.

Periodic poling of RTP (Karlsson *et al.*, 1999) and KTA (Rosenman *et al.*, 1999) has been reported.

2.5.7 Potassium niobate (KN)

KN (KNbO_3) was one of the earliest nonlinear materials to be exploited specifically for the purpose of frequency-doubling semiconductor diode lasers (Günter *et al.*, 1979). KN is attractive for this application because of a fortuitous match between its room-temperature noncritical phasematching wavelength ($\sim 860\text{ nm}$) and a typical emission wavelength for GaAlAs laser diodes. KN has

large nonlinear coefficients, can be temperature-tuned over a broad range and crystals several millimeters in length are available with good optical quality. Among its disadvantages are susceptibility to the formation of unwanted domains due to stress or temperature gradients, and an increased infrared absorption caused by the presence of blue light that can be particularly deleterious when KN is used inside a resonant enhancement cavity (Mabuchi *et al.*, 1994).

KN is generally grown by the top-seeded flux-growth method (Mizell *et al.*, 1988). This crystal is subject to a number of phase transitions as it cools from growth temperature. It undergoes a transition from cubic structure to tetragonal at $\sim 435^\circ\text{C}$, from tetragonal to orthorhombic at $\sim 225^\circ\text{C}$, and from orthorhombic to rhombohedral at $\sim -50^\circ\text{C}$. These phase transitions impose limitations on the range over which the temperature of the crystal can be adjusted in an attempt to achieve phasematching. Generally, KN crystals are multi-domain as grown and poling is required in order to achieve a uniform single domain. Depoling can be induced by temperature gradients or by mechanical stress, and this characteristic has presented challenges in using heated crystals for SHG and in polishing the input and output surfaces. Mizell *et al.* (1988) reported that KN crystals may be safely heated from room temperature to 180°C in 10 min, provided that there is uniform thermal coupling to the crystal and suggested that the mechanical stress applied to the crystal should not exceed 3 kg/cm^2 . However, Biaggio *et al.* (1992) suggested heating no faster than 80°C/h and cooling no faster than 40°C/h .

2.5.7.1 Bulk KN for birefringent phasematching

The most common use of KN has been in Type-I SHG and SFG configurations. For propagation along the crystallographic a -axis, KN provides noncritical phasematching for 857 nm, using the d_{32} coefficient. For propagation along the b -axis, KN provides noncritical phasematching for 982 nm, using the d_{31} coefficient (Biaggio *et al.*, 1992). Thus, a -axis propagation is well suited to frequency-doubling of GaAlAs laser diodes, and b -axis propagation is well suited to frequency-doubling of InGaAs laser diodes.

The temperature dependence of the phasematching wavelength in KN was shown in Figure 2.28. Frequency-doubling of the 946 nm and 1064 nm emission of Nd:YAG lasers has been of particular interest. For frequency-doubling of 946 nm, a -axis propagation at a temperature of $\sim 180^\circ\text{C}$ can be used, and for SHG of 1064 nm, b -axis propagation at about the same temperature can be used. However, as mentioned earlier, care must be taken as this temperature is close to that of the transition to the tetragonal phase.

KN has also been used for sum-frequency mixing. Propagation along the a -axis at a temperature of 27.2°C provides noncritical phasematching for mixing

of 1064 nm Nd:YAG laser light with 694.3 nm ruby laser light to produce 420.1 nm (Baumert and Günter, 1987). At -4°C , noncritical phasematching occurs for mixing of 1064-nm light with the 676.4 nm light from a krypton laser. Shichijyo *et al.* (1994) mixed 910-nm light from a $\text{Ti:Al}_2\text{O}_3$ laser with 1064-nm light from a Nd:YAG laser to generate 490 nm (*b*-axis propagation). Fluck and Günter (1996) mixed 982 nm light from an InGaAs laser diode with 768–792 nm light from a tunable GaAlAs laser diode in *a*-axis KN to produce a blue output tunable between 431 and 438 nm.

2.5.7.2 Periodically-poled KN (PPKN)

Periodic poling of KN has been reported, using an electric field applied to a periodic electrode (Meyn *et al.*, 1999). Using a domain-inverted grating with a period of 30 μm , appropriate for frequency-doubling of a 926-nm diode laser using the d_{31} coefficient, Meyn *et al.* produced 3.8 mW at 463 nm from 1.9W of fundamental in a 5-mm long crystal.

2.5.7.3 Waveguides

Indiffusion processes for waveguide fabrication have proven to be difficult to implement in KN, because the diffusion coefficients are rather low for temperatures below the phase transition at $\sim 225^{\circ}\text{C}$. Baumert *et al.* (1985) resorted to inducing a waveguiding region electro-optically, using the static fringing field from a coplanar electrode structure to cause an increased refractive index in the region between the electrodes. However, the most successful approach has used implantation of helium ions, as first described by Bremer *et al.* (1988). The progress made on fabrication of these waveguides is reviewed in (Fluck and Günter, 2000) and references therein.

Ion-implanted KN waveguides have been used for Čerenkov frequency-doubling (Fluck *et al.*, 1992, 1994). Blue powers as high as 4 mW have been reported (Fluck *et al.*, 1996). Efficient generation of blue light in the lowest-order guided mode has also been reported (Fluck *et al.*, 1996a). Pliska *et al.* (1998) were able to generate 14 mW of 438 nm light from a fundamental power of 340 mW in a 7.3-mm long waveguide.

Some properties of KN are given in Table 2.5.

2.5.8 Potassium lithium niobate (KLN)

Although it has not been widely used for blue-green generation, KLN deserves to be mentioned briefly here. Previously we saw that KN provides phasematching down

Table 2.5. KN

Refractive indices				Nonlinear properties							
(Jundt <i>et al.</i> (1993); see also Wiesendanger (1970) and Zysset <i>et al.</i> (1992).)				d tensor Symmetry class $mm\ 2$ $\begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{bmatrix}$							
$n^2 - 1 = (S_1 + b_1 F + b_2 G) \frac{\lambda^2 \lambda_1^2}{\lambda^2 - (\lambda_1 + b_3 F + b_4 G)^2} + (S_2 + b_5 F) \frac{\lambda^2 \lambda_2^2}{\lambda^2 - (\lambda_2 + b_6 F)^2} - (D + b_7 F) \lambda^2 + b_8 F \lambda^4$ $F = (T + 273.15)^2 - (22 + 273.15)^2$ $G = (T - 22)$ $\lambda_i = \frac{1.239852\ \mu\text{m}\cdot\text{eV}}{E_i}$ $\lambda\ \text{in}\ \mu\text{m}\quad T\ \text{in}\ ^\circ\text{C}$				$\lambda_1 = 1064\ \text{nm}$		$\lambda_1 = 826\ \text{nm}$		$\lambda_1 = 1064\ \text{nm}$		$\lambda_1 = 826\ \text{nm}$	
				d_{31}	15.8	16.0	Δ_{31}	0.27	0.23		
				d_{32}	18.3	20.5	Δ_{32}	0.28	0.27		
				d_{33}	27.4	33.3	Δ_{33}	0.58	0.44		
				d_{24}	17.1	—	Δ_{24}	0.26	—		
				d_{15}		16.5		—		0.27	
				Miscellaneous properties							
				Spontaneous polarization							
				$P_s = 0.27\ \text{C/m}^2$ (Uematsu, 1974)							
				$P_s = 0.41\ \text{C/m}^2$ (Günter, 1977)							
				Unit cell dimensions: $a = 5.6896\ \text{\AA}$, $b = 3.9693\ \text{\AA}$, $c = 5.7256\ \text{\AA}$							
				Electro-optic coefficients (pm/V) (Günter, 1974)							
				r_{13}	64	$\begin{array}{ c } \hline 64 \\ \hline 28 \\ \hline 1.3 \\ \hline 380 \\ \hline 105 \\ \hline \end{array}$					
				r_{23}	28						
				r_{33}	1.3						
				r_{42}	380						
				r_{51}	105						
				Piezoelectric, dielectric, elastic constants: See Wiesendanger (1974)							

to around 860 nm and shows little optical damage, but is susceptible to depoling by mechanical or thermal shock. LN is more robust mechanically and thermally, but does not provide phasematching for such short wavelengths and can have severe optical damage problems. Investigation into KLN in some sense has been an attempt to combine the advantageous properties of KN and LN by creating an alloy of the two materials.

Smith *et al.* (1971) reported the temperature required to achieve noncritical phasematching for SHG of 1064 nm as a function of crystal composition. They also performed frequency-doubling of a GaAs laser at 900 nm, using a crystal of composition $\text{K}_3\text{Li}_{1.85}\text{Nb}_{5.15}\text{O}_{15.3}$ at 110 °C. Ouwerkerk (1991) showed that by increasing the lithium content of the crystal, the phasematching wavelength at room temperature could be in the 790–920 nm range. Reid *et al.* (1992) used a $\text{K}_3\text{Li}_{1.97}\text{Nb}_{5.03}\text{O}_{15}$ crystal to achieve SHG of 820 nm light at room temperature.

Hu *et al.* (1999) reported fabrication of waveguides in KLN by ion-exchange in a RbNO_3 melt, but they have not been explored for blue-green generation.

2.5.9 Lithium iodate

The large birefringence, high transparency to short wavelengths, and reasonably large nonlinear coefficients of LiIO_3 have caused it to be employed in a wide variety of blue-green generation experiments. The main disadvantage of LiIO_3 has been that the rather large walk-off angle limits the effective interaction length in the most typical angle-tuned, critically-phasematched configurations.

Lithium iodate has been used for generation of 347 nm light by SHG of ruby lasers (Nath *et al.*, 1970) and for generation of 532 nm by SHG of Nd:YAG lasers (Deserno and Nath, 1969). Several groups have reported generating light in the 295–325 nm range using LiIO_3 for SHG of red dye lasers (Majewski, 1983, Johnson and Johnston, 1983, Buesener *et al.*, 1986, Evans and Webb, 1994). Deserno *et al.* (1987) demonstrated direct doubling of a 880-nm GaAlAs laser diode in LiIO_3 , Galat *et al.* (1996) reported generation of 490-nm light by direct doubling of a InGaAs laser diode, and Hayasaka *et al.* (1996) reported generation of 397-nm by resonantly-enhanced SHG of a GaAlAs laser diode. Risk and Lenth (1987) used lithium iodate for intracavity frequency-doubling of a 946-nm Nd:YAG laser to produce blue light at 473 nm. Radiation in the 355–435 nm range has been obtained by resonant SHG of a Ti:S laser (Adams and Ferguson, 1990).

Planar waveguides have been made in LiIO_3 by proton implantation (Rosso *et al.*, 1993, 1997), but they have not yet been extensively used for blue-green generation.

Some properties of lithium iodate are given in Table 2.6.

Table 2.6. *Lithium iodate*

Refractive indices	Nonlinear properties		
(Shen <i>et al.</i> (1991), based on measurements of Umegaki <i>et al.</i> (1971)) $n_o^2 = 3.41442 + \frac{4.6877 \times 10^{-2}}{\lambda^2 - 3.7378 \times 10^{-2}} - 7.6766 \times 10^{-3} \lambda^2$ $n_e^2 = 2.9161 + \frac{3.6550 \times 10^{-2}}{\lambda^2 - 2.1170 \times 10^{-2}} - 2.3194 \times 10^{-3} \lambda^2$ λ in μm For temperature variation, see Gettemy <i>et al.</i> (1988)	<i>d</i> tensor Symmetry class 6 $\begin{bmatrix} 0 & 0 & 0 & d_{14} & d_{15} & 0 \\ 0 & 0 & 0 & d_{15} & -d_{14} & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{bmatrix}$		
	<i>d</i> coefficients (Baumert <i>et al.</i> (1984); see also Uematsu (1974))		
		$\lambda_1 = 1064 \text{ nm}$	
	d_{31}	4.65	Jerphagnon (1970)
		7.3	Choy and Byer (1976)
		-4.1	Eckardt <i>et al.</i> (1990)
	d_{33}	4.8	Jerphagnon (1970)
		$\lambda_1 = 1341.4 \text{ nm}$ $\lambda_2 = 1079.5 \text{ nm}$	
	d_{15}	5.57	Shen <i>et al.</i> (1991)

2.5.10 Beta barium borate (BBO) and lithium borate (LBO)

2.5.10.1 Birefringent phasematching

Two borate crystals – BBO ($\beta\text{-BaB}_2\text{O}_4$) and LBO (LiB_3O_5) – have become popular for generation of wavelengths in the near-ultraviolet to blue portions of the spectrum, because their transparency at these short wavelengths is superior to that of the other popular nonlinear materials discussed above. BBO is transparent down to about 190 nm; LBO is transparent to slightly shorter wavelengths ($\sim 155 \text{ nm}$). Generally, BBO has a larger effective nonlinearity and temperature-tuning bandwidth than LBO, but LBO has a smaller walk-off angle, higher damage threshold, and broader angle- and wavelength-tuning bandwidths than BBO (Chen and Lin, 1993, Rines *et al.*, 1995, Kellner *et al.*, 1996).

BBO has been used for efficient generation of wavelengths as short as $\sim 205 \text{ nm}$ by Type-I SHG of dye lasers (Kato, 1986, Miyazaki *et al.*, 1986) and as short as 191 nm by mixing of the output of a $\text{Ti:Al}_2\text{O}_3$ laser with the frequency-doubled output of an argon laser (Watanabe *et al.*, 1991). Others have generated 194-nm radiation by mixing 257-nm light (obtained by frequency-doubling the 515-nm line of an argon laser) with 792-nm light from a $\text{Ti:Al}_2\text{O}_3$ (Watanabe *et al.*, 1993) or diode laser (Berkeland *et al.*, 1997). In LBO, phasematching permits generation

Table 2.7. BBO

Refractive indices	Nonlinear properties		
(Kato, 1986)	<i>d</i> tensor		
$n_o^2 = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2$	Symmetry class 3m		
$n_e^2 = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01677} - 0.01516\lambda^2$	$\begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$		
λ in μm	<i>d</i> coefficients (Baumert <i>et al.</i> (1984); see also Uematsu (1974).)		
Temperature variation (Eimerl <i>et al.</i> (1987))			
$\frac{dn_o}{dT} = -16.6 \times 10^{-6}/^\circ\text{C}$	d_{22}	1.6	Chen <i>et al.</i> (1986)
$\frac{dn_e}{dT} = -9.3 \times 10^{-6}/^\circ\text{C}$	d_{31}	0.1	Chen <i>et al.</i> (1986)
	d_{33}	0.08	Chen <i>et al.</i> (1986)
Miscellaneous properties			
Electro-optic coefficients: Nakatani <i>et al.</i> (1988)			
Dielectric, mechanical, thermal properties: Eimerl <i>et al.</i> (1987)			

of wavelengths as short as 277 nm by SHG (Lin *et al.*, 1991) and as short as 188 nm by SFG (Wu *et al.*, 1992). BBO can be used for frequency doubling of Ti:Al₂O₃ lasers to produce tunable output between 350 nm and 470 nm (Rines *et al.*, 1995). Kaneda and Kubota (1995) generated 355-nm light for optical disk mastering by mixing the 1064-nm output of one Nd:YAG laser with the 532-nm output obtained by SHG of a second Nd:YAG laser, and Wu (1993) used both LBO and BBO to generate 355-nm output by frequency-tripling of Nd:YAG lasers. Beier *et al.* (1997) generated 403-nm light using BBO and LBO for resonantly-enhanced frequency-doubling of a GaAlAs laser diode. LBO has been used for intracavity frequency-doubling of diode-pumped Cr:LiSAF lasers (Furukawa *et al.*, 1996), producing cw powers at 435 nm as high as 20 mW (Laperle *et al.*, 1997). LBO can be used for noncritically phasematched SHG of 1064 nm (at 148 °C) (Ukachi *et al.*, 1990, Lin *et al.*, 1990), and Martin *et al.* (1997) reported using LBO in this fashion for efficient frequency-doubling of a diode-pumped Nd:YAG laser.

2.5.10.2 Waveguides

Planar waveguides have been fabricated in BBO (Boudrioua *et al.*, 2000) and LBO (Davis *et al.*, 1993) using helium-ion implantation, but they have not yet been studied extensively for blue-green generation.

Some properties of BBO and LBO are given in Tables 2.7 and 2.8.

Table 2.8. *LBO*

Refractive indices	Nonlinear properties		
(Kato, 1990)	<i>d</i> tensor		
$n_x^2 = 2.4542 + \frac{0.01125}{\lambda^2 - 0.01135} - 0.01388\lambda^2$	Symmetry class <i>mm2</i>		
$n_y^2 = 2.5390 + \frac{0.01277}{\lambda^2 - 0.01189} - 0.01848\lambda^2$	$\begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{bmatrix}$		
$n_z^2 = 2.5865 + \frac{0.01310}{\lambda^2 - 0.01223} - 0.01861\lambda^2$	<i>d</i> coefficients (pm/V) (Baumert <i>et al.</i> (1984); see also Uematsu (1974).)		
(<i>a</i> – axis $\parallel x$, <i>c</i> – axis $\parallel y$)	<i>d</i> ₃₁	2.75	Chen <i>et al.</i> (1989)
λ in μm	<i>d</i> ₃₂	2.97	Chen <i>et al.</i> (1989)
Temperature variation (Velsko <i>et al.</i> , 1991)	<i>d</i> ₃₃	0.15	Chen <i>et al.</i> (1989)
$\frac{dn_x}{dT} = [-1.8 \pm 0.2] \times 10^{-6}/^\circ\text{C}$	Miscellaneous properties		
$\frac{dn_y}{dT} = [-13.6 \pm 0.1] \times 10^{-6}/^\circ\text{C}$	See Chen <i>et al.</i> (1989)		
$\frac{dn_z}{dT} = [(-6.3 \pm 0.6) + (2.1 \pm 0.8)\lambda] \times 10^{-6}/^\circ\text{C}$			

2.5.11 Other materials

2.5.11.1 Organic materials

A common figure-of-merit for nonlinear materials is d^2/n^3 . Because of both large nonlinearity and lower refractive index, organic materials can have very high figures-of-merit – for example, the material DAN (4-(N,N-dimethylamino)-3-acetamidonitrobenzene) has a figure-of-merit of about 150, about ten times higher than the figure-of-merit for KN. These large figures-of-merit have spawned the synthesis of a veritable alphabet soup of organic materials for blue-green light generation: DAN, DIVA, PCNB, mNA, CMTC, ABP, ADPA, MHBA, and DMNP, to name just a few.

Despite the potential advantages of organic material and an enormous amount of effort directed toward their development, they have not yet been widely adopted for generation of blue-green light, owing mainly to significant absorption of blue-green wavelengths, concerns about their long-term stability and power-handling capability, and difficulties in growing large crystals of good optical quality.

Here, we mention briefly a few results in which organic materials have been used in device-type configurations.

Looser *et al.* (1989) experimented with using a fundamental wavelength (1000 nm) chosen so that the generated second harmonic fell within the tail of the

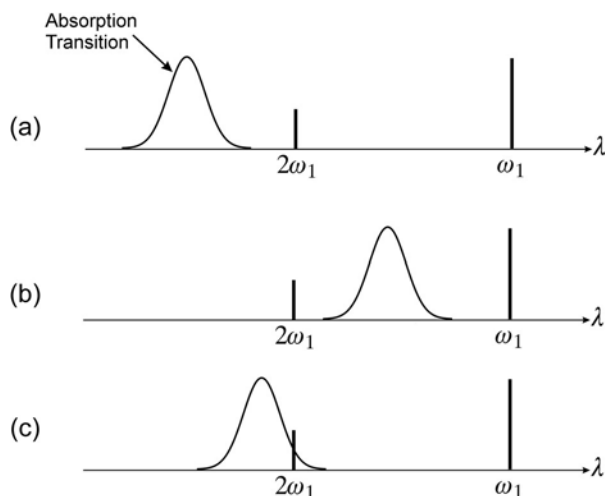


Figure 2.62: Position of fundamental and second-harmonic wave relative to absorption transition. (a) Usual case for most inorganic materials, in which both fundamental and second harmonic are to the “red side” of the transition; (b) case in which second-harmonic wavelength is to the “blue side” of the transition, while fundamental is to the “red side”, as in anomalous dispersion phasematching; (c) case investigated with some organic materials, where second harmonic falls slightly inside the absorption band.

absorption band of the organic nonlinear material DAN (Figure 2.62). In this case, there is a resonant increase in the nonlinearity which competes against increased absorption of the second harmonic. They found that the net effect was a factor-of-2 increase in overall efficiency compared with what they obtained using a fundamental wavelength of 1064 nm, which placed the generated second harmonic outside the absorption band. In another experiment, they compared frequency-doubling of 994-nm light using two different nonlinear materials: a 0.75-mm-thick DAN crystal with an optical density of 1.1 at the second harmonic and a 5-mm long KTP crystal having negligible absorption at the second harmonic. They found that, despite the greater absorption, the DAN crystal was four times more efficient than the KTP one. Chimuka and Umegaki (1994) investigated a similar trade-off in fiber waveguides having an organic crystal core.

Intracavity SHG of neodymium lasers has been investigated using DAN (Ducharme *et al.*, 1990) and thienychalcone (Sasaki, 1992). Sagawa *et al.* (1995) used the material APDA for resonator-enhanced frequency-doubling of a laser diode. Other groups have reported frequency-doubling of a GaAlAs laser diode using MHBA (Yuan *et al.*, 1996), CMTC (Yuan *et al.*, 1997), and ABP (Li *et al.*, 1997).

Organic crystals can be grown inside a glass capillary tube to form a fiber with a crystal core for guided-wave interactions. Harada *et al.* (1991) demonstrated

SHG of a diode laser using a fiber with a core of DMNP and Uemiya *et al.* (1992) demonstrated SHG of a Nd:YAG laser using a fiber with a core of DAN. Other work has included demonstration of waveguide QPM using an organic medium (Suhara *et al.*, 1993) and use of a Langmuir–Blodgett film for guided-wave frequency-doubling (Penner *et al.*, 1994).

2.5.11.2 Poled glasses

One persistent problem with waveguides fabricated in traditional nonlinear materials, like those described above, is that the loss is typically substantially higher than in optical fibers. The observation of efficient SHG in a single-mode optical fiber with a Ge-doped core in 1986 (Österberg and Margulis, 1986) suggested that it might be possible to create a permanent second-order nonlinearity in a low-loss optical fiber and compensate for a small nonlinearity with a long interaction length.

Kester *et al.* (1992) reported producing a permanent periodic $\chi^{(2)}$ in a planar silica waveguide by initially exposing the waveguide to intense beams at both the fundamental and second-harmonic wavelengths. Kashyap (1989) applied an interdigital electrode to an optical fiber that had been polished to expose the evanescent field and showed that he could induce a second-order nonlinearity through the third-order nonlinearity of the glass and the applied static field, although it was not permanent. Weitzman *et al.* (1994) performed a similar experiment in germanium-doped planar silica waveguides.

Myers *et al.* (1991) reported that they could induce a *permanent* second-order nonlinearity of ~ 1 pm/V in a 4- μm thick layer near the surface of fused silica wafer by applying an electric field at an elevated temperature, then allowing the sample to cool to room temperature. Okada *et al.* (1992) used a variation of this technique to produce glass waveguides capable of SHG using modal phasematching. Kashyap *et al.* (1994) showed that a periodic $\chi^{(2)}$ could be induced in fused silica using this approach in conjunction with a periodic electrode.

Kazansky *et al.* (1995) reported producing a periodic nonlinearity (~ 0.1 pm/V) with a period of 20 μm in an optical fiber using their particular implementation of this technique and were able to observe blue light generation. Pruneri *et al.* (1998) reported producing a $\chi^{(2)}$ grating 7.5 cm long. Another advantage of optical fibers is that the dispersion is generally much lower than in crystalline waveguides, so that shorter pulses can be frequency-doubled before efficiency limits imposed by group velocity walk-off are encountered. The Southampton University group reported frequency-doubling of 2.2 ps Ti:Al₂O₃ laser pulses at 844.6 nm (Pruneri and Kazansky, 1997) and 100 fs optical parametric oscillator pulses at 1.5 μm (Pruneri *et al.*, 1998).

2.5.11.3 Semiconductor materials

We previously mentioned the large nonlinearity of GaAs in connection with its use as a nonlinear material for frequency-doubling of 10.6- μm radiation. Other semiconductors – such as ZnSe, ZnTe, and ZnS – also have large nonlinearities and their large bandgaps provide reasonable transparency in the blue-green portion of the spectrum. For example, ZnSe ($d \sim 54$ pm/V, $n \sim 2.5$) has a figure-of-merit d^2/n^3 of 180 – compared with a value of about 15 for KN (Shoji *et al.*, 1997).

The main problem with using these materials has been phasematching. Because of their cubic symmetry they have no inherent birefringence that can be used to achieve phasematching. Diffusion bonding of a properly oriented stack of plates could be used to implement QPM, similar to what has been done with GaAs, and Gordon *et al.* (1994) reported some initial attempts to diffusion bond ZnSe. Angell *et al.* (1994) have demonstrated a clever technique for creating an “orientation template” so that layers of ZnSe and ZnTe can be grown with the periodic orientation changes necessary to implement QPM. In waveguides made from semiconductor materials, modal phasematching or counterpropagation can be used, as described in Section 2.4.6.

2.5.11.4 Self-doubling materials

All the techniques for blue-green light generation described in the first part of this book rely upon nonlinear frequency upconversion of infrared lasers. Thus, there are really at least two materials involved: the nonlinear optical material used for SHG or SFG and the active medium of the infrared laser. In many instances, this active lasing medium will be a rare-earth-doped crystal such as Nd:YAG. Thus, one line of exploration in the quest for compact blue-green lasers has been the search for materials which have suitable nonlinear optical characteristics but also contain rare-earth laser ions. If such materials could be found, both laser emission of the infrared wavelength and frequency conversion could take place in a single crystal.

Unfortunately, it has proven difficult to find a material that advantageously combines these two functions. Neodymium-doped LN, one such material that has been extensively pursued, illustrates this point. When neodymium ions are introduced into this crystal, the symmetry of the sites they occupy leads to luminescence characteristics that are polarization-dependent. The cross-section for lasing with the electric field parallel to the c -axis is considerably higher than that for lasing with the electric field perpendicular to that axis. However, it is this latter polarization that is required for Type-I phasematching. Thus, it proves difficult to make the laser oscillate with the correct polarization for efficient frequency-doubling.

Despite difficulties of this nature, a number of experiments have been done demonstrating the operation of miniature blue-green lasers based on the

self-doubling approach. We will discuss these further in Chapter 5. Here, we simply point to work that is still continuing to perfect the fabrication of nonlinear crystals containing active ions. A number of materials have been, and are being, pursued, including Nd:LiNbO₃ and Tm:LiNbO₃ (Johnson and Ballman, 1969), Nd:MgO:LiNbO₃ (Fan *et al.*, 1986), Nd:Sc₂O₃:LiNbO₃ (Yamamoto *et al.*, 1994), Yb:LiNbO₃ (Foulon *et al.*, 1995), Nd:YAB (yttrium aluminum borate), Nd:Gd₂(MoO₄)₃ (Kaminskii *et al.*, 1996), Nd:LaBGeO₅ (Capmany *et al.*, 1998), Er:LiNbO₃ (Zheng *et al.*, 1998), Nd:KTP (Solé *et al.*, 1996), Nd:Ca₄GdO(BO₃)₃ (Mougel *et al.*, 1998), Yb:Ca₄GdO(BO₃)₃ (Mougel *et al.*, 1999), and Yb:YAB (Wang *et al.*, 1999).

2.6 SUMMARY

In this chapter, we have considered some of the basic elements of blue-green light generation using nonlinear optical processes. In particular, we examined the basic mathematical description of the process, explored important practical considerations such as focusing and the influence of the laser spectrum, considered various methods for achieving phasematching, and looked at the kinds of nonlinear materials that are useful for blue-green generation. In the next four chapters, we will consider how these elements can be put together in the quest for efficient and powerful sources of blue-green light. In Chapter 3, we consider the simple approach of single-pass SHG; in Chapter 4, resonator-enhanced SHG; in Chapter 5, intracavity frequency-doubling of diode-pumped solid-state lasers; and in Chapter 6, waveguide-based blue-green light sources.

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3

Single-pass SHG and SFG

3.1 INTRODUCTION

Perhaps the simplest approach to generating blue-green light using nonlinear frequency conversion is the one shown in Figure 3.1, in which the beam emitted by an infrared laser is focused directly into a suitable nonlinear crystal so that a blue-green beam is produced by SHG in a single pass. In principle, a semiconductor diode laser would be the ideal infrared source for such an application, since these devices are generally rugged and compact, long-lived, and highly efficient. The single-pass arrangement is attractive because of its simplicity and because it allows the blue-green output beam to be modulated simply by modulating the current supplied to the diode laser.

What conditions are necessary in order to make the single-pass arrangement work efficiently? In Chapter 2, we presented the following equation describing the blue-green power produced by cw SHG using a circular gaussian beam focused in a nonlinear crystal, in the low-conversion efficiency regime:

$$P_3 = \frac{16\pi^2 d_{\text{eff}}^2}{\epsilon_0 c \lambda_1^3 n_3 n_1} P_1^2 e^{-\alpha' l} h(\sigma, \beta, \kappa, \xi, \mu) \rho \quad (3.1)$$

This expression has been modified slightly from the form given in Equation (2.29) by including the factor ρ , which accounts for the effect of the laser spectral distribution on the conversion efficiency (as discussed in Section 2.4.3). Examination of this equation leads to some rather obvious conclusions. The nonlinear material should have a high figure-of-merit for SHG, $d_{\text{eff}}^2/n_3 n_1$, the loss should be low in order to make the term $e^{-\alpha' l} \approx 1$, and the interaction length l should be as long as possible, compatible with other constraints such as keeping the loss low. The fundamental power P_1 delivered by the infrared laser to the nonlinear crystal should be large.

In considering the Boyd–Kleinman focusing factor $h(\sigma, \beta, \kappa, \xi, \mu)$ and the factor ρ accounting for the influence of the spectral distribution, it becomes apparent that

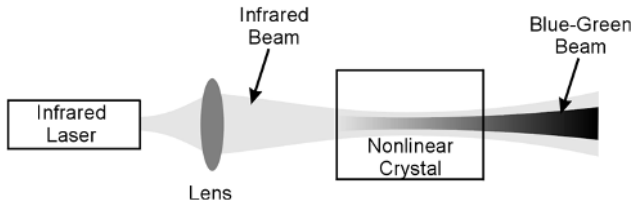


Figure 3.1: Basic configuration for single-pass SHG.

there is an intimate interrelatedness between the choice of the infrared source and the choice of the nonlinear crystal. The Boyd–Kleinman focusing factor involves both beam properties of the laser and material properties of the nonlinear crystal. For example, the behavior of h is strongly dependent on whether the interaction is critically or noncritically phasematched. In practice, the selection of infrared source (and hence, fundamental wavelength) is limited to a relatively small number of options having suitable characteristics; similarly, the choice of nonlinear material is limited to a relatively small number of available crystals having favorable properties for SHG (although the availability of periodically-poled materials has eased this restriction somewhat). Thus, part of the problem of designing a blue-green source based on SHG involves finding suitable combinations of lasers and nonlinear materials with properties that lead to efficient SHG. Part of the solution to this problem has involved trying to invent both new lasers and new nonlinear materials.

Assuming that a suitable combination of nonlinear material and laser is found, what general characteristics should the laser have? The degree of focusing required to optimize P_3 can be calculated from Boyd–Kleinman theory, but the laser must deliver a beam that can be focused to such an extent.¹ Furthermore, the spectral distribution of the laser must be over a narrow range compared with the phase-matching bandwidth in order to optimize ρ . Much of the historical development of blue-green sources based on SHG of diode lasers, as depicted in Figure 3.2, has involved trying to create infrared sources that have high output power as well as

¹ Implicit in the Boyd–Kleinman analysis is the requirement that the infrared beam should have a diffraction-limited gaussian profile. In addition, such a profile is necessary if the blue-green output is to have a beam quality suitable for applications such as optical data storage that demand diffraction-limited focusing of the blue-green beam. A way of characterizing the spatial quality of laser beams – the M^2 factor – has become popular in recent years (Siegman, 1990).

The M^2 formalism is an expression of the Heisenberg uncertainty principle applied to gaussian beams. The uncertainty principle states that the product of our uncertainty about the position of a photon and our uncertainty about its momentum must exceed a certain minimum value. Thus, if we decrease our uncertainty about the transverse position of a photon by confining it to a small waist, we must increase our uncertainty about its transverse momentum. The more tightly we confine a beam by focusing it to a small spot, the more rapidly it will diffract as it propagates away from that spot. The minimum uncertainty product corresponds to a TEM₀₀ gaussian, such as we considered in the Boyd–Kleinman analysis, which is said to be “diffraction-limited”. The M^2 beam quality factor is the ratio between the uncertainty product of a real beam and the minimum uncertainty product for a TEM₀₀ gaussian beam. Thus, $M^2 = 1$ indicates an ideal, diffraction-limited, lowest-order gaussian mode; $M^2 > 1$ indicates a departure from this ideal. In addition to failing to be diffraction-limited, real laser beams may also be asymmetric, astigmatic, or both, and these conditions can also adversely impact the blue-green output power obtained through single-pass SHG or SFG. (Asymmetry and astigmatism are discussed later on in this chapter and depicted in Figure 3.4.)

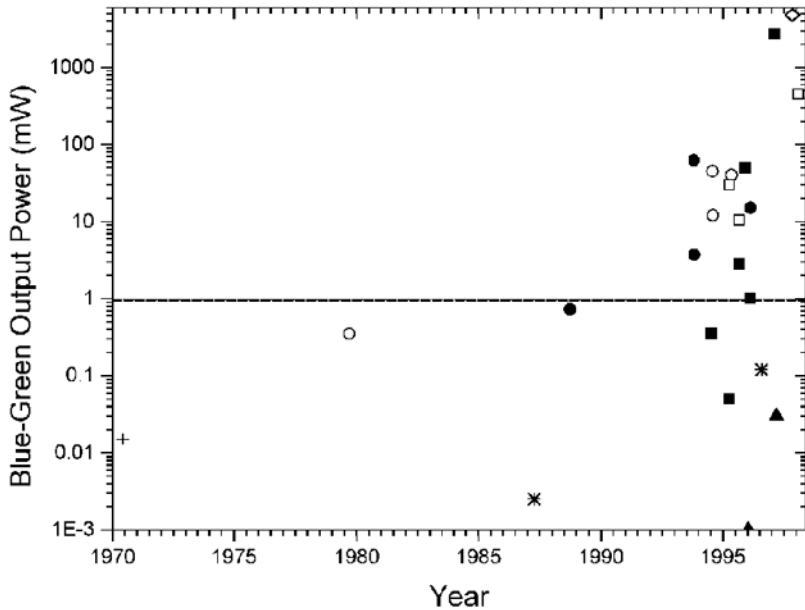


Figure 3.2: Historical development of blue-green sources based on single-pass SHG. Closed points represent cw operation; open points represent pulsed operation. Nonlinear materials used are coded using the following representation: KN – circles; PPLN – squares; PPLT – up triangles; KTP – down triangles; PPKTP – diamonds; alpha-iodic acid – cross; lithium iodate – asterisk. The points plotted here represent a variety of specific output wavelengths, and a variety of different configurations for single-pass SHG, as described in this chapter.

good spatial and spectral mode properties. Dramatic progress has been made in developing new designs for semiconductor diode lasers that have these characteristics. However, many of the semiconductor diode laser designs having the *highest* output powers have not had spatial and spectral mode characteristics suitable for direct SHG, but these devices can be used to pump solid-state lasers that do have suitable spatial and spectral mode characteristics to simultaneously increase the output power available from such lasers while enforcing good spatial and spectral mode behavior. This development, and the improvements that have resulted from it, are the focus of the remainder of this chapter.

3.2 DIRECT SINGLE-PASS SHG OF DIODE LASERS

3.2.1 Early experiments with gain-guided lasers

The first report of frequency doubling of a diode laser² dates to 1970 in the work of Edmonds and Smith from IBM, who reported SHG of pulsed GaAs injection

² Recall that the first GaAs injection laser was demonstrated in 1962. Research groups from IBM (Nathan *et al.*, 1962) and General Electric (Hall *et al.*, 1962) simultaneously published independent reports of stimulated emission from a GaAs *p-n* junction in different journals on November 1, 1962. A group working at MIT's Lincoln Laboratory published a third report exactly one month later (Quist *et al.*, 1962).

lasers, using HIO_3 (α -iodic acid) for angle-tuned Type-I birefringent phasematching (Edmonds and Smith, 1970). Strictly speaking, discussion of their experiment belongs in the next chapter, which deals with resonator-enhanced SHG, since they used this approach in an attempt to increase the second-harmonic power. However, it is appropriate to discuss their work briefly here, since it reports the first attempt to directly frequency-double the output of a semiconductor laser and since their experience illustrates many of the difficulties that have been encountered during the subsequent development of single-pass SHG devices.

In their paper, they mention that they also considered using LiIO_3 as the nonlinear material, but that a crystal with suitable optical quality was not available to them. They also state that they investigated the use of other known nonlinear materials, like LN and $\text{NaBa}_2\text{Nb}_5\text{O}_{15}$, but that these could not provide phasematching at the wavelength of interest (~ 900 nm). Furthermore, no known nonlinear crystals could provide noncritical phasematching at that wavelength, so they were forced to use an angle-tuned configuration, with its attendant disadvantages. These statements provide a good illustration of the problem described above, that of finding a compatible combination of laser and nonlinear material. In addition to the phasematching problem, Edmonds and Smith reported grappling with other difficulties that have remained issues in direct doubling of diode lasers ever since – optimal focusing of the beam in the crystal, spatial mode quality of the diode laser, frequency stability and purity.

The early GaAs lasers used by Edmonds and Smith were pulsed and operated at 77 K. Although stimulated emission at room temperature (Burns and Nathan, 1963) and cw emission (Howard *et al.*, 1963) were reported soon after the first demonstrations of GaAs injection lasers, it was not until nearly a decade later that room-temperature semiconductor lasers began to achieve low enough threshold currents and long enough lifetimes for them to be considered practical. By 1979, the development of the double-heterostructure geometry (Kroemer, 1963) had led to efficient room-temperature $\text{Ga}_{1-x}\text{Al}_x\text{As}$ lasers, with emission wavelengths that could be adjusted by controlling the aluminum fraction x . Günter *et al.* (1979) knew that KN could provide noncritical, Type-I phasematching for SHG of 860 nm light, and recognized that an appropriately designed $\text{Ga}_{1-x}\text{Al}_x\text{As}$ diode laser could be made to emit at that wavelength. They demonstrated SHG of a GaAlAs laser, pulsed to achieve higher peak power (785 mW, 10-ns duration), and achieved a peak blue power of 0.35 mW using the configuration shown in Figure 3.3.

The diode laser they used in these experiments was a “gain-guided” device. As illustrated in Figure 3.4, in such devices the light is confined in the y - z plane by a real refractive index step, while confinement in the x - z plane results only because of the spatial distribution of the gain (resulting from the finite width of the electrical contact or “stripe”). When guiding is provided by a true refractive

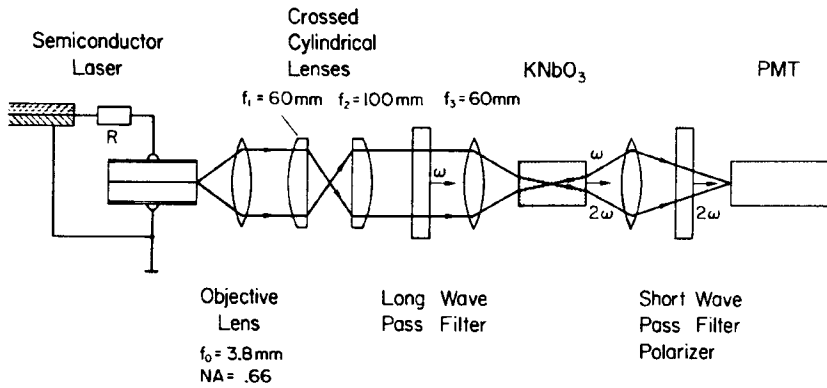


Figure 3.3: Experimental setup used by Günter and colleagues for single-pass SHG of a GaAlAs laser diode. (Reprinted with permission from P. Günter *et al.* (1979).)

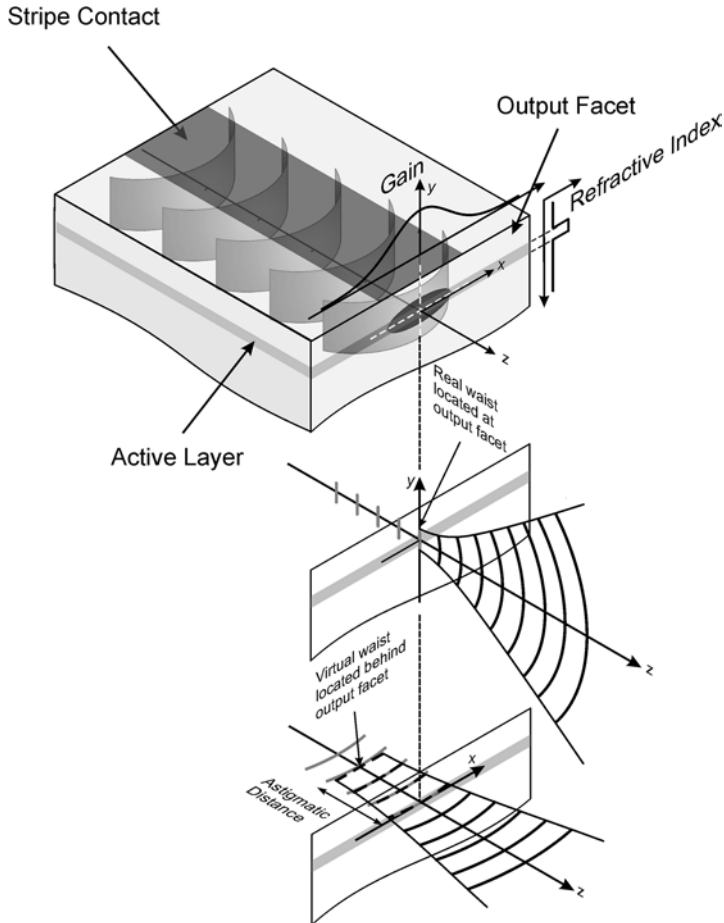


Figure 3.4: Configuration of a gain-guided semiconductor diode laser, showing how gain-guiding leads to astigmatism.

index step, the phase fronts of the guided modes are planar. In contrast, the gain-guiding mechanism produces curved phase fronts (Cook and Nash, 1975). Thus, if we treat the beam emitted by the laser as a gaussian beam and trace it back to its waist – the point at which it is narrowest and has planar phase fronts – we would find that the location of the waist differs depending on whether we are looking at the beam in the plane parallel to $(x-z)$ or perpendicular to $(y-z)$ the junction. If we look at the beam in the $y-z$ plane, it appears to have originated from the output facet; however, if we look at the beam in the $x-z$ plane, it appears to have originated from behind the output facet. The spatial displacement of the two apparent waists is the astigmatism of the laser diode, and can be tens of microns. Since astigmatism results from gain-guiding, it is also generally dependent on the drive current level, and thus may be different for different output powers.

In addition, because the degree of confinement is different in the two planes, the beam has an elliptical shape. In the $y-z$ plane, the real refractive index step is associated with a layer only $0.7\text{ }\mu\text{m}$ thick, while the stripe contact giving rise to gain-guiding in the $x-z$ plane is $7\text{ }\mu\text{m}$ wide. Thus, the beam diffracts much more rapidly in the $y-z$ plane than in the $x-z$ plane, giving rise to an elliptical beam. The presence of astigmatism and ellipticity makes the job of collimating and optimally focusing the light into the nonlinear crystal more complicated. Günter and coworkers used an anamorphic telescope, which has a different optical power for rays travelling in the plane of the junction than for those travelling in the plane perpendicular to it, in order to circularize the beam.

Perhaps the most serious limitation to efficient frequency conversion encountered by Günter and colleagues was the incompatibility of the *spectral emission* of the diode laser with the *spectral acceptance* of the KN crystal. The laser diode emitted in eight longitudinal modes, each with a width of about $0.6\text{ }\text{\AA}$ and separated by about $2.3\text{ }\text{\AA}$. In contrast, the wavelength-acceptance phasematching bandwidth was only about $0.9\text{ }\text{\AA}$ for the 5.74-mm long crystal. Thus, about 75% of the power generated by the diode laser was at wavelengths outside the phasematching band.

3.2.2 Early experiments with index-guided lasers

Within a few years, improved diode laser structures, such as the constricted double heterostructure (CDH) with a large optical cavity (LOC) (Botez, 1980), were available that emitted tens of milliwatts in a single spectral and spatial mode. In 1983, Baumert *et al.* (1983) reported cw blue light generation using KN for frequency-doubling of such a laser diode. The laser diodes used in these experiments were index-guided, so that the astigmatism was substantially lower than in the

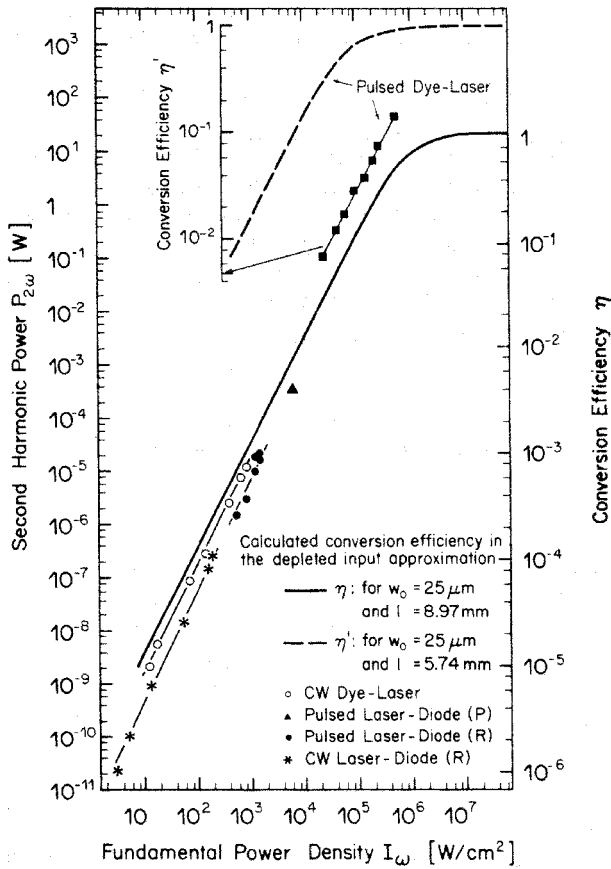


Figure 3.5: Summary of second-harmonic powers obtained using single-pass SHG with different laser sources. (Reprinted with permission from Baumert *et al.* (1983). Copyright: Elsevier Science.)

gain-guided devices used in the 1979 experiments of Günter and coworkers (note that even when a real refractive index step provides lateral confinement, there is generally still some gain-guiding component so that the astigmatism is reduced but not completely eliminated). The improved spectral mode characteristics of these lasers enabled them to achieve $0.2 \mu\text{W}$ of cw emission at 420 nm . Although the diode laser emitted $\sim 28 \text{ mW}$, only 5 mW could be delivered to the nonlinear crystal due to losses in the anamorphic telescope designed to circularize the beam and due to reflection at the uncoated surface of the KN crystal.

Baumert and coworkers' paper also contained Figure 3.5, which summarized the results obtained with KN up to that time using both dye and diode lasers. It can be seen that although the experimentally-obtained powers were approaching what

Boyd–Kleinman theory predicted, substantial increases in laser diode power would be required in order to reach blue-green output powers on the order of 1 mW.

The difficulties encountered in these earlier experiments are instructive, because they helped define the direction that further development of semiconductor diode lasers for SHG would need to take – higher power, narrower spectrum, and better spatial mode properties. These improved devices have found application not only in single-pass doubling, but also in resonator-enhanced SHG, pumping of solid-state lasers, and waveguide frequency-doubling, so that this discussion also serves to lay the groundwork for subsequent chapters describing those configurations.

3.2.3 High-power index-guided narrow-stripe lasers

One pathway for the development of semiconductor lasers for use in blue-green light generation has involved the improvement of index-guided, narrow-stripe lasers to permit reliable operation at high output powers. By the early 1990s, lasers capable of emitting ~ 500 mW cw before failing had been demonstrated, with lifetimes at the 100–200 mW cw level exceeding 5000 hours (at wavelengths ~ 860 nm, suitable for SHG in KN) (Welch *et al.*, 1990, Jaeckel *et al.*, 1991). These devices incorporate a waveguide only 3–4 μm wide to provide transverse (i.e., the x – z plane in Figure 3.4) guiding in order to enforce single-spatial-mode operation. Although this output power was still somewhat too low for efficient *single-pass* SHG, their moderate output power combined with single-spatial- and longitudinal-mode characteristics made these devices very attractive for resonator-enhanced SHG (Chapter 4) and waveguide SHG (Chapter 6).

The advantages associated with the superior spectral and spatial mode properties of index-guided narrow-stripe lasers can be illustrated by experiments in which they were used for sum-frequency mixing. Risk and Lenth (1989) reported generating 459-nm light by sum-frequency mixing of the 1064-nm emission from a Nd:YAG laser with 809-nm radiation from a GaAlAs laser diode. They compared the mixing efficiency obtained when an index-guided narrow-stripe laser was used with that obtained when a broad-area, multi-longitudinal-mode gain-guided laser was used instead, and showed that the superior spectral and spatial mode properties of the index-guided narrow-stripe laser yielded a mixing efficiency about four times higher than when the broad-area laser was used.

Another avenue worth mentioning is the development of index-guided narrow-stripe lasers at other wavelengths. Although the combination of GaAlAs lasers and KN has many attractive advantages, it is not without some disadvantages. As we discussed in Chapter 2, KN suffers from a number of practical problems that detract from its utility, despite its high nonlinear coefficient and ability to provide noncritical phasematching at a wavelength conveniently obtained from GaAlAs

diode lasers. In particular, the phasematching tolerances with respect to angle, temperature, and wavelength are rather narrow, even for the noncritically phasematched case. On the other hand, KTP has very broad tolerances, particularly with regard to temperature, as we also saw in Chapter 2. Unfortunately, the shortest wavelength for which KTP can provide phasematching at room temperature is 994 nm; in the late 1980s, when these properties of KTP were being explored, semiconductor diode lasers were not well developed at such wavelengths. However, the advantageous nonlinear properties of KTP motivated an effort to develop strained-layer InGaAs lasers that could emit at 994 nm in order to investigate noncritically phasematched Type-II SHG in KTP as a possible mechanism for a blue-green source based on single-pass SHG (Risk *et al.*, 1989).

Another advantageous combination of laser diode and nonlinear crystal again involves KN. As pointed out by Günter *et al.* (1979) and Kato (1979), in addition to providing noncritical phasematching for SHG at 860 nm, KN can also provide noncritical phasematching for SHG using a fundamental wavelength near 986 nm. This wavelength happens to be very close to the wavelength required to pump erbium-doped fiber amplifiers (EDFAs). There has been a tremendous effort to develop high-power InGaAs lasers at that wavelength for pumping EDFAs, and the availability of high-powered, frequency-stable lasers at ~ 986 nm (Moser *et al.*, 1991) has stimulated interest in generation of 493 nm blue-green light by single-pass SHG of these lasers using KN.

Despite their excellent spatial and spectral mode characteristics, index-guided narrow-stripe laser diodes have found limited use for single-pass nonlinear generation of blue-green light, because their output powers have been restricted by facet damage to a few hundred milliwatts. While this power level is adequate for efficient blue-green generation in resonator-enhanced and waveguide schemes, it simply is not enough for efficient single-pass SHG.

3.2.4 Multiple-stripe arrays

One way to overcome the power limitations of narrow-stripe lasers is to use several of them together in a monolithic array (Figure 3.6). Placing the individual stripes close together maintains a relatively narrow overall emitting aperture, which is advantageous for focusing the emitted beam to a small spot. However, close proximity also causes the optical modes localized under each stripe to couple through their evanescent fields. The oscillation of such an array is thus described in terms of a “supermode” – that is, a mode characteristic of the entire array structure, rather than of each individual emitter. The gain is highest in the pumped regions directly under the stripe contacts; in between, the unpumped regions are lossy. If the array elements all oscillate in phase with each other, the supermode has a significant

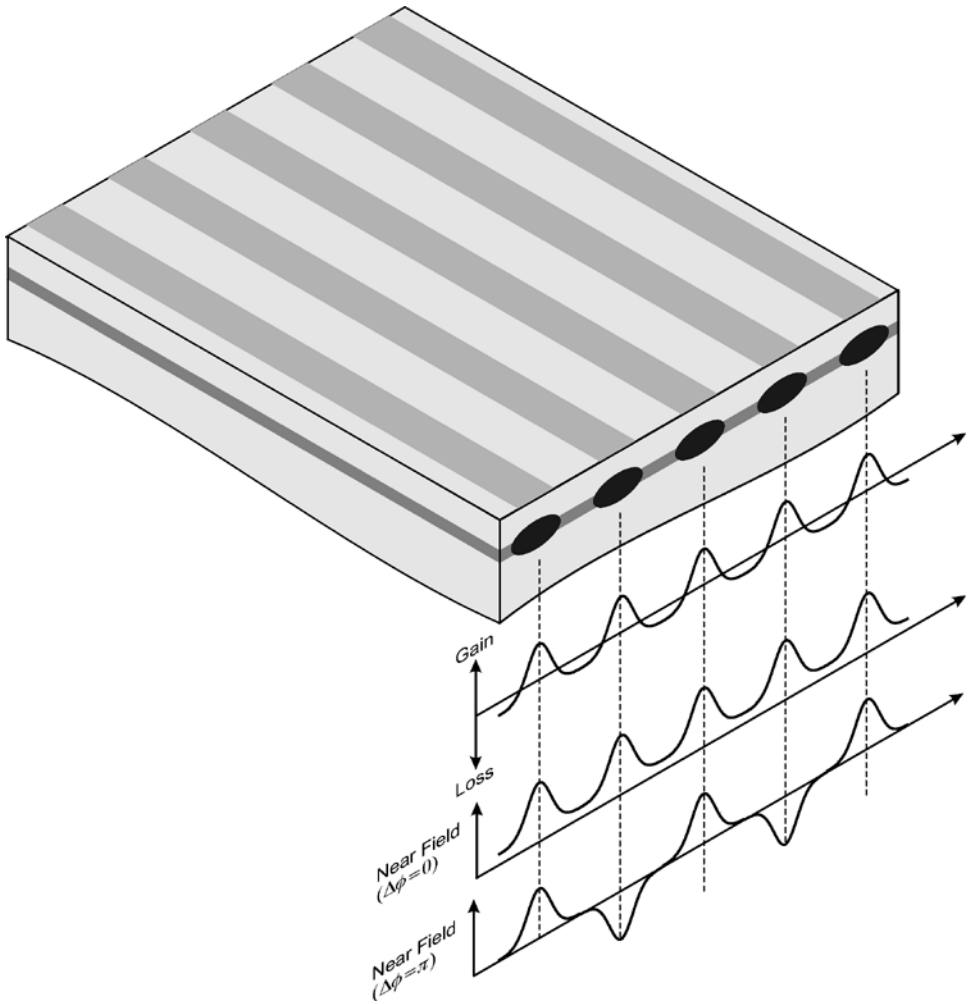


Figure 3.6: Monolithic array of multiple narrow-stripe emitters.

electric field strength in the in-between areas, and can experience significant loss. However, if adjacent elements oscillate π out of phase with each other, their electric fields cancel in between the stripes, thereby reducing the loss for that supermode. Since the supermode experiencing the lowest loss has the lowest threshold for lasing, such arrays tend to oscillate in this “ π out of phase” supermode. The far-field distribution from such an array consists of two lobes, since the contributions from neighboring emitters cancel along the center-line due to the π phase shift (Figure 3.7).

This two-lobed far-field pattern and the tendency of these devices to oscillate in numerous longitudinal modes at high power (Figure 3.7) have been drawbacks which have limited the application of multiple stripe arrays to single pass SHG.

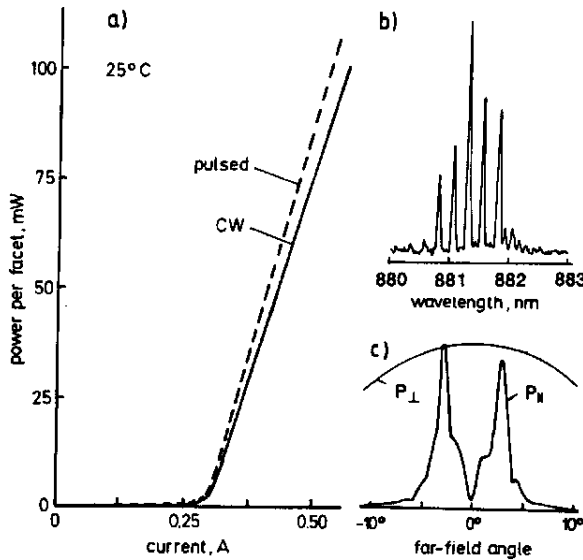


Figure 3.7: Characteristics of an eleven-stripe phase-locked array used by Deserno *et al.* (1987) for single-pass SHG: (a) light/current characteristics, (b) longitudinal mode spectrum under pulsed operation, (c) lateral and transverse far field patterns, showing dual-lobed distribution typical of multiple-stripe arrays. (Reprinted with permission from Deserno *et al.* (1987).)

For example, Deserno *et al.* (1987) demonstrated SHG in LiIO_3 using lasers of this variety emitting at 880 nm. Lithium iodate has a broader wavelength acceptance than KN, but can provide only critical phasematching at 880 nm, which is accompanied by substantial walk-off. In addition, the relevant nonlinear coefficient is only about one-third as large as that of KN. The combination of these limitations was such that although they were delivering over 20 times more power to the nonlinear crystal than in Günter *et al.*'s experiment, they were able to produce only slightly more power at the second-harmonic wavelength ($\sim 0.3 \mu\text{W}$) than Günter and colleagues achieved.

Another approach to using phase-locked arrays for frequency-doubling is shown in Figure 3.8. Here, light from a narrow-stripe, single-mode laser is injected into the high-power phase-locked array. Injection locking causes the output from the high-power laser to emit a diffraction-limited beam in a single-longitudinal mode (Goldberg and Weller, 1987). Chun *et al.* (1988) used such a source for frequency-doubling, and were able to obtain 0.72 mW of 421 nm light using 270 mW of cw infrared light from the injection-locked array, and using a 5-mm long KN crystal.

While sources better suited to direct doubling than multiple-stripe arrays have since been developed, high-power arrays have proven to be extremely important for pumping solid-state lasers, as we shall see in Section 3.3.

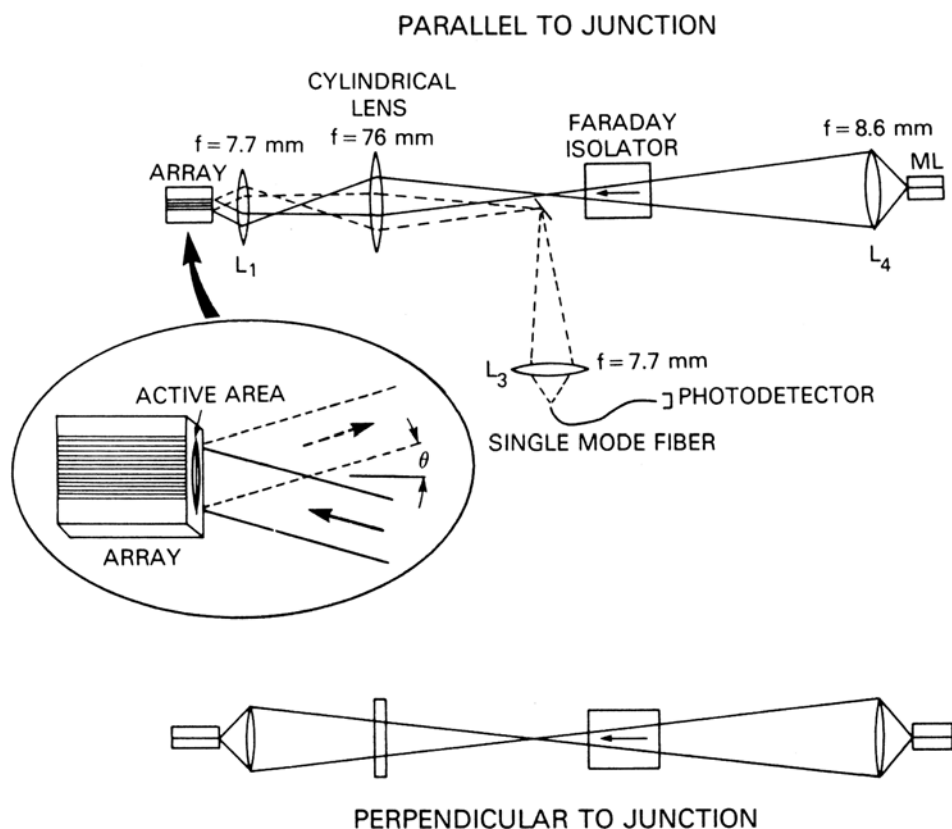


Figure 3.8: Injection locking arrangement of Goldberg and Weller using a low-power single-mode laser diode as the master oscillator. (Reprinted by permission Goldberg and Weller (1987).)

3.2.5 Broad-area lasers

Another direction that has been pursued to reach higher power is simply to increase the width of the lasing region, ultimately leading to a “broad-area” laser. For example, Chinone *et al.* (1976) reported achieving 390 mW cw power in a device with an 80- μm wide stripe, and, more recently, Shigihara *et al.* (1991) reported generating 2.6 W cw from a single-quantum-well, separate-confinement heterostructure (SQW-SCH) with a 150- μm wide stripe. However, it is difficult to maintain a uniform optical power density over the entire width of the lasing region in such lasers – instead, the beam tends to develop complex spatial distributions due to self-focusing in the gain medium. This process, known as “filamentation”, results in a near-field radiation pattern consisting of a number of bright spots, which is generally unsuitable for use in nonlinear upconversion.

As mentioned above, broad-area lasers have been used in sum-frequency mixing experiments. They have also been investigated for single-pass SHG. Galat *et al.* (1996) reported frequency-doubling of the emission from a broad-area (stripe width $\sim 100\text{ }\mu\text{m}$) InGaAs laser diode emitting at 980 nm, using LiIO_3 as the nonlinear crystal. With 1 W of power delivered to the nonlinear crystal at the fundamental wavelength, they generated only 120 μW of second-harmonic power.

Although the poor spatial and spectral mode characteristics of broad-area lasers have largely precluded their use as sources for blue-green light generation, they have proven more useful in two related applications. As is the case with multiple-stripe arrays, the imperfect spectral and spatial mode characteristics of broad-area diode lasers are less of a disadvantage when they are used to pump solid-state lasers. In addition, while they suffer from numerous disadvantages as oscillators, the same basic structure used for broad-area lasers has proven useful as an amplifier, which can be used to boost the power of a single-spatial-mode, single-frequency master oscillator.

3.2.6 Master oscillator–power amplifier (MOPA) configurations

3.2.6.1 MOPAs using discrete devices

While broad-area diode laser structures have numerous disadvantages when used as oscillators, many of these disadvantages are less severe when they are used as amplifiers to boost the power of a second laser which has low output power but otherwise desirable characteristics, such as a stable, single-frequency spectrum and a good spatial mode. This configuration, known as “MOPA”, has been extensively explored as a source for single-pass SHG.

In 1992, Goldberg *et al.* (1992) reported using a broad-area GaAlAs amplifier to increase the power emitted by a single-frequency Ti:S laser (Figure 3.9). The amplifier was a separate-confinement heterostructure 600 μm wide and 1 mm long. The end facets were anti-reflection-coated to produce a reflectivity of about 10^{-3} , which was necessary to suppress lasing and allow the device to operate as

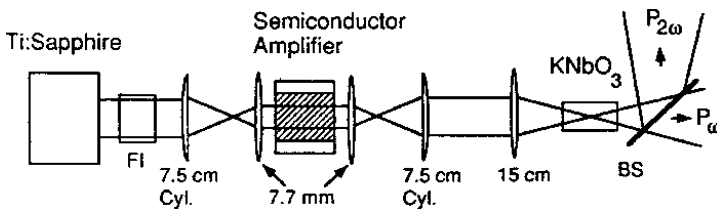


Figure 3.9: Experimental setup used by Goldberg and colleagues for single-pass SHG of a Ti:S master oscillator augmented by a semiconductor power amplifier. (Reprinted with permission from Goldberg *et al.* (1992).)

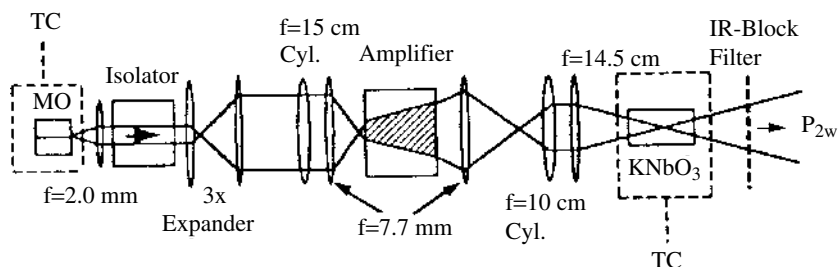


Figure 3.10: Experimental setup used by Goldberg and coworkers for single-pass SHG using a semiconductor diode laser as the master oscillator. (Reprinted with permission from Goldberg *et al.* (1993).)

a travelling-wave, single-pass amplifier. When driven by 15 A of current (in 5 μs long pulses, with a duty cycle of 10^{-3}), it was able to deliver a maximum power output of 7.7 W. When this amplified beam was focused into a 14-mm long KN crystal, 400 mW of blue output was generated (from 6.4 W of amplified 860 nm power incident on the crystal).

Later, Busse *et al.* (1993) reported increasing the length of the amplifier to 2.2 mm, which enabled them to amplify 400 mW of Ti:S output to a power level of 15.6 W when the amplifier was driven by 32 A of current (in pulses of 0.3 μs duration). With this amount of infrared power focused into a 20-mm long KN crystal, they were able to generate 3.0 W of second-harmonic power at 430 nm, giving a single-pass conversion efficiency of 19%.

Using a Ti:S laser as the master oscillator is fine for demonstrating the basic principle, but practical operation demands the use of a single-frequency semiconductor diode laser instead. Continuous-wave operation is also desirable for many applications. In 1993, Goldberg and coworkers used a single-mode semiconductor diode laser as the master oscillator (Figure 3.10) (Goldberg *et al.*, 1993). The power available from their diode laser was lower (150 mW) than the output of the Ti:sapphire laser used in their earlier experiments, and cw pumping of the amplifier introduced thermal limitations that were not significant in their earlier experiments where the amplifier was pumped with low-duty-cycle ($\sim 1\%$) pulses.

In this experiment, they used a tapered, or flared, amplifier structure, in which the width of the pumped region increases along the length of the device. Tapering the amplifier in this way improves the ability of the device to deliver the energy contained in the electrically-pumped gain region to the optical signal. In order to see this, consider an amplifier with a uniform width. The amount of energy that is available to be delivered to the optical signal is proportional to the pumped volume; thus, an amplifier with a wide stripe has more energy available for extraction than one with a narrow stripe. However, extracting as much of this energy as possible requires developing a large enough optical power density to drive the amplifier into

saturation. In an amplifier with a wide stripe, the input power required to produce this power density is larger than in an amplifier with a narrow stripe. Hence, design of the amplifier is driven by conflicting goals: on one hand, it is desirable to make the stripe narrow so that less input power is required to achieve saturation and efficient energy extraction; on the other hand, it is desirable to make the stripe broad in order to increase the total output power that can be delivered by the amplifier. The tapered amplifier design allows both goals to be achieved. The narrow width of the stripe at the input allows the amplifier to become saturated with a relatively low input power, so that energy is efficiently extracted from the pumped region, which is narrow at that location. The total power delivered by the amplifier is then increased by gradually flaring the width of the gain region, maintaining a uniform optical density and degree of saturation across that width for efficient energy extraction.

In the experiments of Goldberg and coworkers, the master oscillator emitted a power of 130 mW, of which 100 mW was incident upon the amplifier and 38 mW was actually coupled into the amplifier. The maximum output of the amplifier was 2.3 W, obtained with a current of 7 A, and this led to a blue-green output power of 62 mW when doubled using a 12.4-mm long KN crystal.

In another variation on this configuration, Goldberg and Mehuys (1994) demonstrated single-pass SHG of a mode-locked MOPA. In this configuration, shown in Figure 3.11, a “compound cavity” laser was used, in which the output facet of the narrow-stripe master oscillator was anti-reflection coated ($R \sim 1\%$), and a grating was placed between the master oscillator and the power amplifier. The laser cavity extended from the highly-reflective back facet of the narrow-stripe amplifier to the front facet of the tapered amplifier, as evidenced by the appearance of a sharp threshold in output power as a function of amplifier current (Goldberg *et al.*, 1994). With both devices present inside the lasing cavity, one can be used to

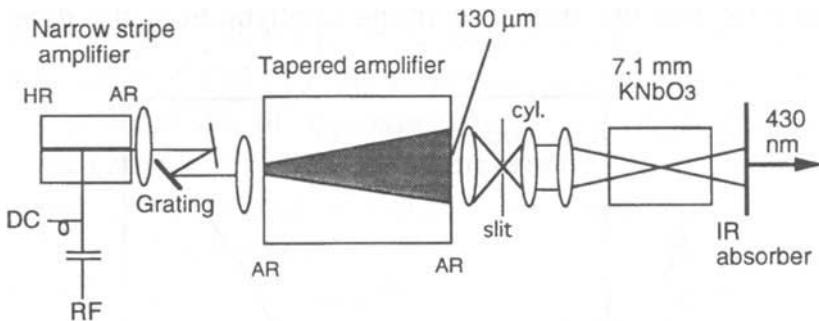


Figure 3.11: Experimental setup used by Goldberg and Mehuys for single-pass SHG of a mode-locked compound-cavity laser. (Reprinted with permission from Goldberg and Mehuys (1994).)

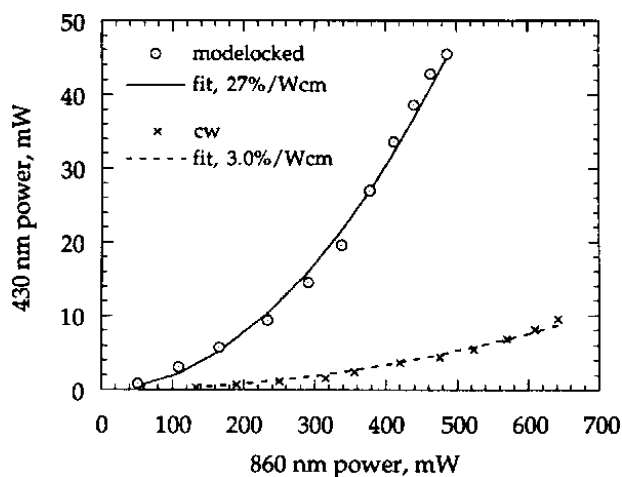


Figure 3.12: Second-harmonic power obtained by SHG of the source shown in Figure 3.11, comparing mode-locked and cw operation. (Reprinted with permission from Goldberg and Mehuys (1994).)

modulate the gain in order to produce active mode-locking, while the other can be used for power amplification. The presence of the grating permitted tuning of the center wavelength and narrowing of the spectral emission, which resulted in shorter mode-locked pulse lengths. Mode-locking was achieved by modulating the current supplied to the narrow-stripe amplifier at 1.544 GHz, a frequency that corresponds to the third harmonic of the longitudinal mode spacing for a cavity with an optical length of 30 cm. The blue output at 430 nm consisted of pulses ~ 12.5 ps long and spaced by 650 ps. The high peak power of the mode-locked pulses combined with the quadratic dependence of second-harmonic power on fundamental power produced a substantially higher average blue output power than when the laser was run with cw excitation alone (Figure 3.12), even though mode-locking makes the frequency spectrum broader (~ 1.0 Å) than the phasematching bandwidth (0.7 Å). A maximum average blue output power of 45.5 mW (1.6 W peak power) was obtained with mode-locked operation, compared to 9.5 mW with cw operation.

In both curves in Figure 3.12, the same DC current was applied – the only difference was in whether the rf modulation was turned on or off. Thus, the blue output can be switched on and off by corresponding switching of the rf modulation, leading to a 5:1 contrast in terms of average power or a 170:1 contrast in instantaneous power. Goldberg and Mehuys (1994a) developed this idea further, demonstrating a contrast of 8:1 in average power and investigating the cavity dynamics governing the build up and shut off of the pulses.

Later, Goldberg *et al.* (1995) used essentially the same source for frequency-doubling in PPLN. They used a 6-mm PPLN crystal with $\Lambda = 3.3$ μm and obtained

an average power at 432 nm of 2.8 mW when the laser was operated cw and 10.5 mW when the laser was mode-locked. When the crystal was used at room temperature, they observed that the generated blue beam was distorted by photorefractive effects at powers of only a few hundred microwatts; however, if the crystal was heated to 150 °C, and used at the corresponding phasematching wavelength, photorefractive effects were eliminated.

The mode-locked 430 nm source demonstrated by Goldberg and coworkers has been further exploited to generate up to 15 μ W average power of ultraviolet light, tunable around 215 nm, by using a second single-pass SHG step to convert the 430 nm radiation to its second harmonic (Goldberg and Kliner, 1995). Goldberg and Kliner also demonstrated mixing of the 858-nm fundamental generated by a mode-locked compound-cavity laser with the 429-nm light generated by single-pass SHG of that same laser to produce ultraviolet radiation tunable around 286 nm (Goldberg and Kliner, 1995a).

As new types of diode lasers become available, the device configurations discussed above, which were implemented with GaAlAs lasers, can be implemented with other kinds of lasers. Knappe *et al.* (1998) reported generation of 30 μ W of 340-nm radiation by single-pass frequency-doubling of a red AlGaInP laser diode in LiIO₃ using a MOPA configuration. Their configuration was similar to that of Figure 3.11 in using an external grating for tuning, but differed in the addition of an optical isolator between the master oscillator and power amplifier.

3.2.6.2 Monolithically-integrated MOPA devices

One feature of the tapered amplifier is that the width of the narrow input end can be made comparable with the width of the active stripe of the master oscillator, which is convenient for coupling the light emitted by the oscillator into the amplifier. This compatibility, together with the fact that they are generally similar in their epitaxial structure and in their constituent materials suggests fabricating both the master oscillator and the power amplifier on the same chip rather than as separate devices, as was done in the experiments we have just described.

In those experiments, the master oscillator was a separate Fabry–Perot laser diode, in which a cleaved facet serves as the output reflector of the laser cavity. When the master oscillator and power amplifier are integrated onto the same chip, this facet is no longer physically present, so a grating must be used as the output reflector. Figure 3.13 shows the structure of a monolithically-integrated MOPA (or M-MOPA) (Parke *et al.*, 1993). Light in the master oscillator is guided by a single-mode waveguide. Two reflective gratings bound the resonant cavity. The grating that is further away from the power amplifier is made longer in order to provide higher reflectivity; the length of the grating nearer the power amplifier is chosen to provide

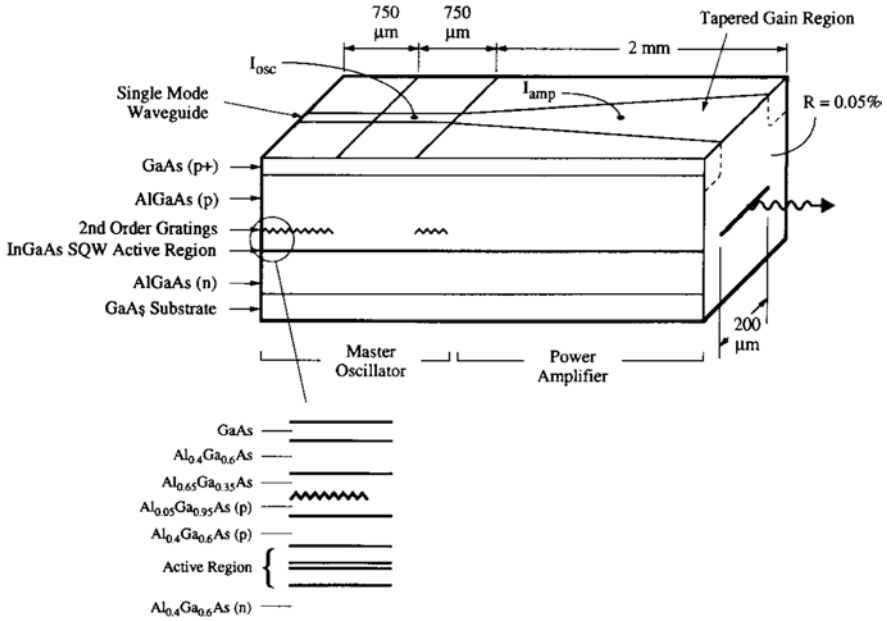


Figure 3.13: Structure of a monolithic semiconductor MOPA. (Reprinted with permission from Parke *et al.* (1993). Copyright: IEEE.)

a particular degree of coupling out of the master oscillator and into the power amplifier. The frequency selectivity of the gratings used in this distributed Bragg reflector (DBR) geometry aids in forcing stable single-longitudinal-mode oscillation of the device, even at high powers. Once the light leaves the master oscillator, it is no longer guided in the lateral direction, and spreads by diffraction as it propagates through the power amplifier. The width of the gain region in the power amplifier section is flared at an angle slightly larger than the diffraction angle of the beam. The cleaved output facet of the power amplifier section is anti-reflection-coated.

Waarts *et al.* (1993) used such a M-MOPA with a single-quantum-well InGaAs active region for single-pass frequency-doubling, using a *b*-cut KNbO₃ crystal (8.2 mm long) for noncritically phasematched SHG of 983 nm light. The experimental configuration is shown in Figure 3.14. The output of the M-MOPA was characterized by large astigmatism ($\sim 600 \mu\text{m}$) and a high degree of asymmetry, so that use of an anamorphic optical system was essential for achieving optimum focusing into the KN crystal. They generated 3.7 mW of 491-nm light using a M-MOPA that delivered a maximum of 1 W cw to the KN crystal. The current supplied to the master oscillator was 100 mA, and the power amplifier was driven with 3 A of current. The generated blue-green beam was nearly diffraction-limited in both the plane defined by the active layer of the M-MOPA ($M^2 = 1.15$) and in the plane perpendicular to it ($M^2 = 1$). The beam quality of the blue-green beam

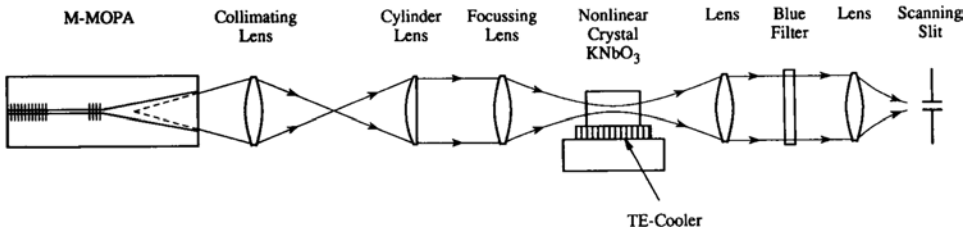


Figure 3.14: Experimental setup used by Waarts and colleagues for single-pass frequency doubling of a monolithic semiconductor MOPA. (Reprinted with permission from Waarts *et al.* (1993). Copyright: IEEE.)

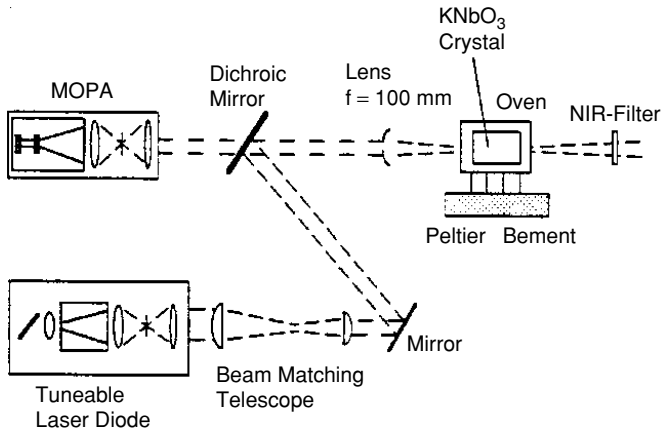


Figure 3.15: Experimental setup for single-pass SFG used by Fluck and Günter. (Reprinted by permission from Fluck and Günter (1997). Copyright: Elsevier Science.)

was somewhat better than that of the infrared beam ($M^2 \sim 1.5$ (Parke *et al.*, 1993)), because the high-spatial-frequency components that increase the M^2 value of the infrared beam do not contribute effectively to the blue-green beam, owing to the limited angular acceptance for SHG in KN. Waarts and coworkers also showed that they could modulate the blue-green output beam by modulating the current of the master oscillator, and demonstrated this by imposing a 20-MHz square wave modulation on the blue-green output beam. Note that the configuration shown in Figure 3.14 does not include an isolator. One feature of M-MOPA devices is a decreased sensitivity to optical feedback, so that, if the reflected light can be reduced to a certain level by very good anti-reflection coatings, the use of a bulky and expensive isolator may not be necessary. Fluck and coworkers used a very similar arrangement, with a 17-mm long KN crystal, to generate 10 mW of 491-nm radiation from 760 mW of infrared light (Fluck *et al.*, 1996).

Fluck and Günter (1997) have also used M-MOPAs for generation of blue-green light by sum-frequency mixing. In their experiment (Figure 3.15), they used a

M-MOPA (982.6 nm) as one infrared source; as the second source they used an extended-cavity laser that was tunable from 772 to 789 nm. The gain element in the extended-cavity laser source was a GaAlAs tapered amplifier. The light emitted from the narrow end of the gain element illuminated a grating, which efficiently retroreflected only a particular wavelength – thus, the lasing wavelength could be set by changing the angle of the grating. For any wavelength in this range, they could achieve phasematching by adjusting the temperature of the *a*-cut KN crystal to the appropriate value, which was in the range 54–76 °C. The infrared light was polarized along the *b*-axis and the resulting blue-green light, which was tunable over the range 432–438 nm, was polarized along the *c*-axis.

3.2.6.3 Master-oscillator–fiber-amplifiers

Another variation on the MOPA theme, described by Guskov *et al.* (1998), is shown in Figure 3.16. Here, an ytterbium-doped fiber amplifier was used to boost the power of a strained-layer InGaAs laser diode oscillating at 1062 nm. A 20-mm long PPLN crystal was used for quasi-phasematched SHG. In cw operation, this system produced 440 mW of power at 531 nm, corresponding to 6 W of 1062-nm light delivered by the MOPA to the crystal. In pulsed operation, the current supplied to the laser diode master oscillator was modulated to produce a train of 2-ns long optical pulses with a repetition rate of 64 MHz. A conversion efficiency of 41% was obtained with an average fundamental power of 3.2 W (peak fundamental power of ~25 W) and an average second harmonic power of 1.3 W.

Hart and colleagues (1999) reported a similar experiment in which a fiber amplifier was used for power boosting. They performed sum-frequency mixing of 1064 nm and 1550 nm in a PPLN crystal to generate 630 nm red light. A Nd:YAG (neodymium-yttrium aluminum garnet) laser generated the 1064 nm signal, which was used to pump an erbium/ytterbium codoped fiber amplifier. This amplifier increased the power of the 1550 nm source, which consisted of a laser diode pre-amplified by an erbium-doped fiber amplifier. The 9-m length of the Er/Yb fiber was chosen as a compromise between amplification of the 1550-nm signal and

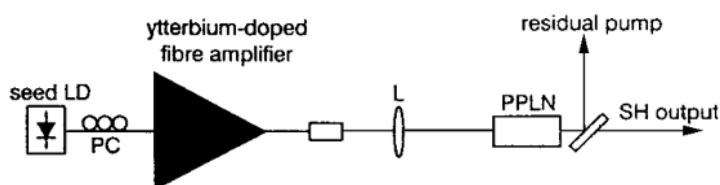


Figure 3.16: Experimental setup used by Guskov and colleagues for single-pass SHG of a diode laser augmented by an ytterbium-doped fiber amplifier. (Reprinted with permission from Guskov *et al.* (1998).)

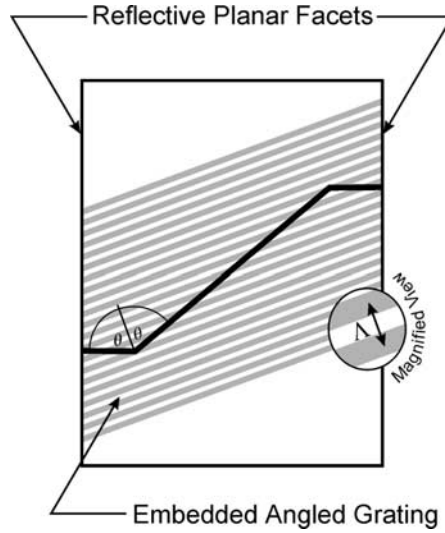


Figure 3.17: Schematic top view of the α -DFB laser.

absorption of the 1064-nm signal, in order to maximize the product of the powers emerging from the fiber at the two wavelengths. With 5.6 W at 1064 nm and 1.4 W at 1550 nm, they were able to generate 0.45 W at 630 nm, using a 2.3-cm long PPLN crystal.

3.2.7 Angled-grating distributed feedback (DFB) lasers

Another type of single-mode, high-power semiconductor diode laser has been investigated that seems to have great potential for single-pass SHG (Dzurko *et al.*, 1995). The basic principle of this laser, denoted the “angled-grating distributed-feedback laser” or “ α -DFB”, is shown in Figure 3.17. In this design, the active layer is bounded by layers of lower refractive index; thus, Figure 3.17 shows a view looking down on a two-dimensional planar waveguide. An angled grating is embedded in the epitaxial structure so that it interacts with the mode guided by the planar waveguide. Two planar facets act as mirrors that define the laser cavity. These two flat reflectors force the lasing mode to have planar phase fronts at these two surfaces. So, imagine a ray corresponding to such a laser mode that leaves the left-hand reflector at normal incidence. The ray encounters the grating and is diffracted. The only way it can reach the right-hand reflector at normal incidence is if it is again diffracted by the grating. Thus, the ray corresponding to a lasing mode must experience an even number of diffractions, following a zig-zag path such as the one shown in Figure 3.17.

In order for the lasing mode to be diffracted by the grating, it must satisfy the Bragg condition, $m\lambda = 2N_{\text{eff}}\Lambda \cos \theta$, where m is the order of diffraction, λ is the wavelength, N_{eff} is the effective index of the guided mode, Λ is the spatial period

of the grating, and θ is the angle of incidence. For a fixed angle, period, and order, the Bragg condition specifies the wavelength and effective index that the lasing mode must have in order to be efficiently diffracted by the grating. The sensitivity of the Bragg condition to wavelength thus favors single-frequency oscillation of the laser. Furthermore, the dependence of the Bragg condition on the mode index N_{eff} produces modal selectivity, since other modes that might propagate in the planar waveguides would have different values of N_{eff} . (This modal selectivity can also be viewed in terms of the angular selectivity of the grating. A higher-order mode would be associated with a pair of rays which emerge from the left-hand facet at a slight angle from normal incidence. The angular selectivity of the grating is such that they would not be diffracted efficiently.) DeMars *et al.* (1996) have demonstrated a 980-nm α -DFB laser that provides up to 1 W of output power in a single spatial mode with a single-frequency spectrum.

DeMars and colleagues have also reported single-pass frequency-doubling using α -DFB lasers emitting light at 1060 nm and 920 nm. Using a 1-cm long PPLN crystal as the doubling element, they produced 2.4 mW of 530-nm light and 1 mW at 460 nm. They found that, compared with other diode laser designs, the α -DFB structure is relatively tolerant to optical feedback, so that use of an isolator was not necessary.

3.3 SINGLE-PASS SHG OF DIODE-PUMPED SOLID-STATE LASERS

Earlier in this chapter, we indicated that some approaches to achieving high-power operation in semiconductor diode lasers have produced devices that are unsuitable for direct frequency-doubling, because of their spectral and spatial mode properties. In particular, we mentioned that both broad-area lasers and multiple-stripe arrays can have broad spectra and complex spatial distributions that are not well suited for efficient nonlinear frequency upconversion.

However, rather than attempting to use such lasers directly as the source of infrared radiation for SHG or sum-frequency mixing, they can instead be used for optical pumping of lasers based on rare-earth ions doped into a solid-state host. Lasers using trivalent neodymium ions doped at atomic concentrations of around 1% into a YAG crystal as the active medium and pumped by GaAlAs laser diodes have been one of the most popular and successful such systems. The energy-level diagram for Nd:YAG is shown in Figure 3.18, which compares two infrared lasing transitions that have been exploited for generation of blue-green light. The $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition at 1064 nm constitutes a four-level laser system, in which the lower level of the lasing transition has no appreciable equilibrium population. The transition from the $^4F_{3/2}$ level to uppermost component of the $^4I_{9/2}$ ground-state manifold at 946 nm constitutes a “quasi-three-level” laser, since the equilibrium population in the lower

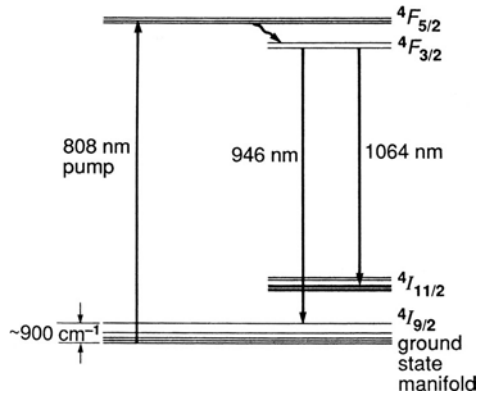


Figure 3.18: Energy level diagram for Nd:YAG, showing the two laser transitions of main interest for blue-green light generation, at wavelengths of 1064 nm and 946 nm, and the pumping transition at 809 nm. (Reprinted by permission from Byer (1988).)

laser level due to thermal excitation is nonnegligible, and this leads to reabsorption of the emitted 946-nm photons. This reabsorption loss causes the threshold pump power to be higher and the slope efficiency to be lower for a 946-nm laser than for a comparable 1064-nm laser. In addition, the gain cross-section for the $^4F_{3/2} \rightarrow ^4I_{9/2}$ is about an order-of-magnitude lower than for the $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition. However, when sufficient pump power is available and when the laser resonator is appropriately designed, the performance of a 946-nm laser can nearly match that of its 1064-nm counterpart. Interest in diode-pumped 946-nm sources for generation of 473-nm light by frequency-doubling has spawned several papers that explore the optimization of the laser design to overcome the adverse effects of reabsorption loss (Fan and Byer, 1987, Risk, 1988, 1997, Beach, 1996 Taira *et al.*, 1997).

In both lasers, population is pumped from the $^4I_{9/2}$ ground state into the $^4F_{5/2}$ manifold, then decays nonradiatively from that level to the $^4F_{3/2}$ upper laser level. The $^4I_{9/2} \rightarrow ^4F_{5/2}$ pump transition corresponds to wavelengths in the vicinity of 800 nm. Figure 3.19 shows an absorption spectrum for 1% doped Nd:YAG, in which it is clear that an absorption peak occurs at ~ 809 nm, a wavelength which is accessible with GaAlAs laser diodes.

Compared with the flashlamps which are sometimes used for pumping Nd lasers, semiconductor diode laser pumps have many advantages. Spectrally, the energy is concentrated in a relatively narrow range, in contrast to the broad emission spectrum of a typical flashlamp. Furthermore, the emission wavelength of the GaAlAs laser light can be tuned by adjusting the temperature of the laser diode, in order to maximize the absorption. Spatially, the pump energy is confined to a beam with high brightness (intensity divided by far-field solid angle), which can be efficiently directed into the laser medium. With regard to both criteria – spatial mode and

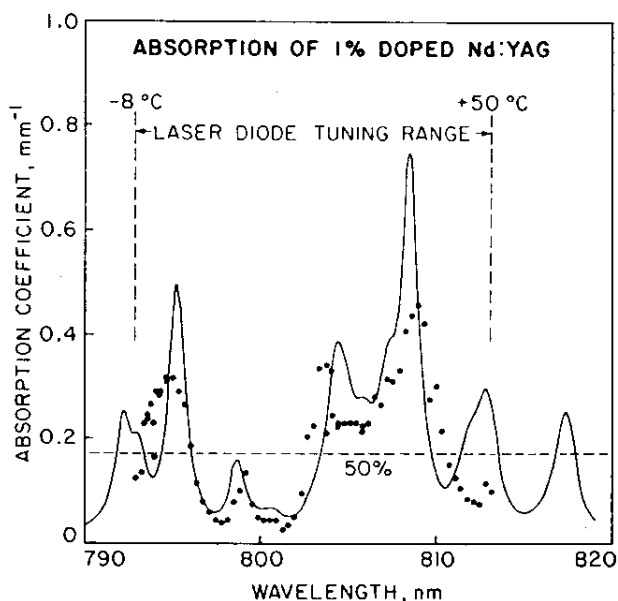


Figure 3.19: Bulk absorptivity of Nd:YAG near 810 nm. The data points correspond to measurements made by temperature tuning a particular diode laser. (Reprinted with permission from Zhou *et al.* (1985).)

spectral mode – the demands placed upon a laser diode intended for pumping a solid-state laser are considerably less stringent than those placed upon a laser diode intended for direct frequency-doubling. Thus, although broad area diode lasers and multiple-stripe arrays have spatial and spectral mode properties which are not compatible with efficient direct doubling, they can be used very effectively for pumping solid-state lasers, which can then be efficiently doubled in a single-pass configuration. Since solid-state lasers can be made to oscillate in a TEM₀₀ gaussian mode with a very narrow spectral distribution, the diode-pumped solid-state laser is, in a sense, a “mode transformer” which converts the poor spatial and spectral characteristics of the diode laser to something more compatible with frequency-doubling and mixing. Although the advantages of pumping a neodymium laser with a GaAs diode were recognized as early as 1963 (Newman, 1963), it was some two decades later before diode pump sources with sufficient power to make this idea practical became available.

Since the first demonstrations of Nd:YAG lasers pumped by “modern” diode lasers in the early 1980s, an impressive variety of laser geometries have been developed, each tailored in some way to a specific purpose or application. Discussion of this wide range of devices is beyond the scope of this text, but we will describe some specific geometries relevant to blue-green light generation as they become appropriate in this and subsequent chapters. Here, we will begin by drawing

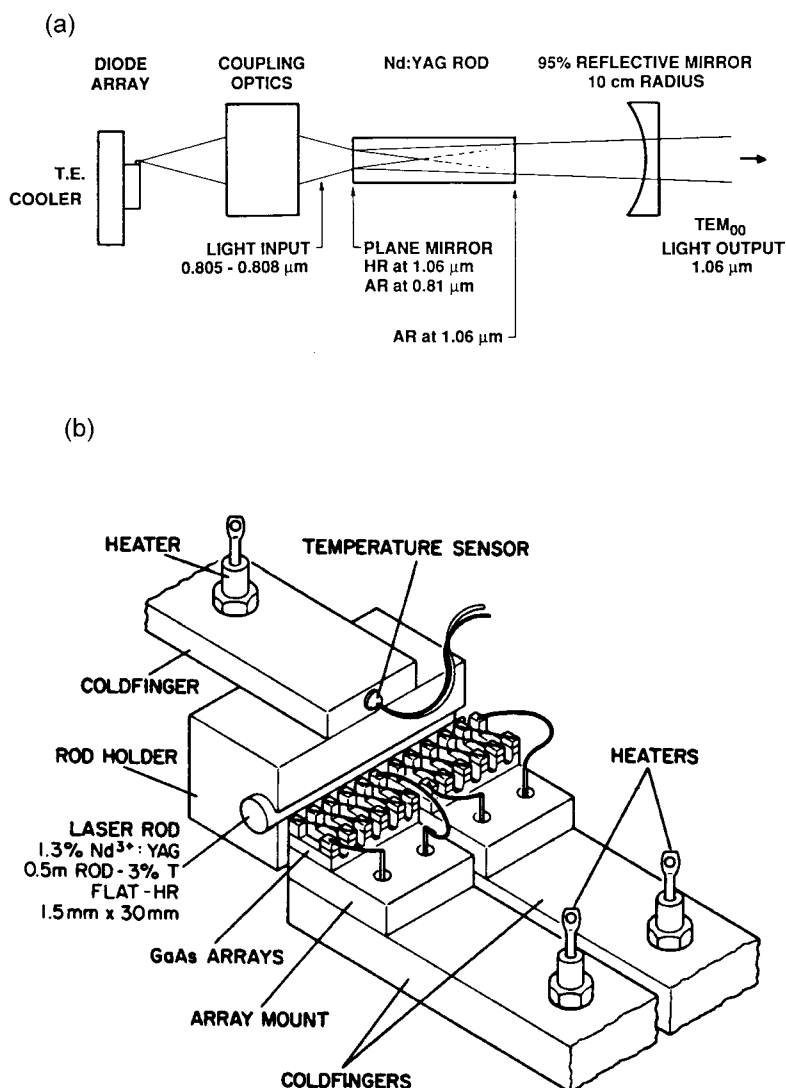


Figure 3.20: (a) An example of an end-pumped Nd:YAG laser. (Reprinted with permission from Berger *et al.* (1987).) (b) Example of a side-pumped Nd:YAG laser. (Reprinted with permission from Conant and Reno (1974).)

a rather broad distinction between the two principal geometries that have been used for pumping, and between two types of laser resonator design.

The two types of pumping we will distinguish are “end-pumping” (also called “longitudinal pumping”) and “side-pumping” (also called “transverse pumping”). In end-pumping, the pump energy emitted by the diode laser enters the solid-state laser along the axis of laser emission, through the end of the lasing medium (Figure 3.20(a)); in side-pumping, the pump energy enters the lasing medium

though the side (Figure 3.20(b)). Both approaches have advantages and disadvantages. End-pumping generally results in a higher efficiency and a better beam quality, since the pump energy can be spatially concentrated within the volume of the TEM₀₀ gaussian mode of the solid-state laser; however, this places demands on the spatial quality of the beam that are more severe than in side-pumping. Side-pumping is thus more easily scalable to higher powers, since it is relatively simple to add additional pump lasers in this geometry; however, it may be more difficult to ensure that the solid-state laser oscillates in a clean TEM₀₀ gaussian mode. In most of the examples given in this text, end-pumping has been used.

While a complete discussion of end-pumping geometries cannot be given here, it is worth pointing out that the advantages of this approach have led to a number of schemes designed to overcome the limitations to scalability. We mention two here, to give the reader an awareness of the ideas that have been investigated. The first is the approach of Fan *et al.* (1989), who considered the requirements that the end-pumping geometry places on the spatial-mode properties of the pump laser (Fan and Sanchez, 1990). They realized that since diode lasers typically emit beams that are diffraction-limited in the direction perpendicular to the junction but not in the direction parallel to it, it is possible to “stack” multiple beams in the diffraction-limited direction, so that more pump energy is delivered to the mode volume of the solid-state laser (Figure 3.21). The second approach is that of Clarkson and Hanna (1996), who used two highly-reflective planar mirrors, parallel to each other but with a lateral offset between them, to reshape the beam so that its M^2 value is approximately the same in both transverse directions, with only a slight decrease in the overall brightness of the beam (Figure 3.22).

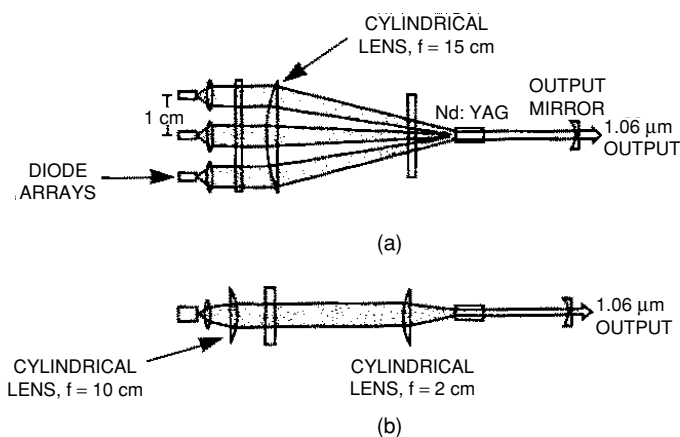


Figure 3.21: Schematic diagram of a Nd:YAG laser end-pumped by multiple diode laser arrays: (a) the plane perpendicular to the diode laser junction; (b) the plane parallel to the diode laser junction. (Reprinted with permission from Fan *et al.* (1989).)

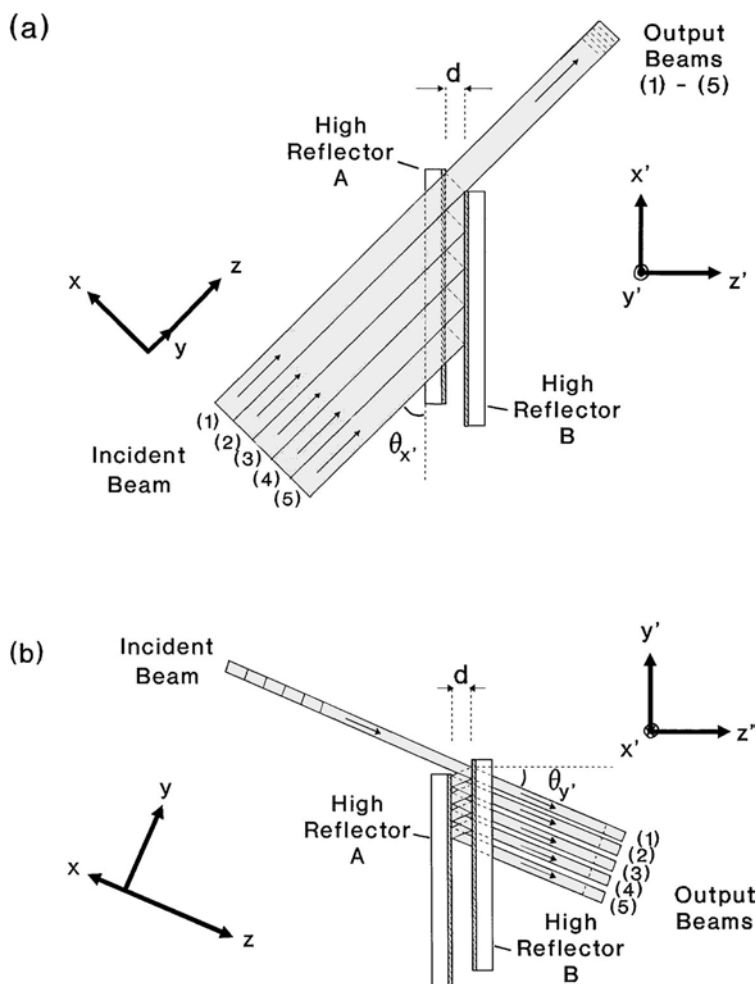


Figure 3.22: Two-mirror beam-shaping technique for high-power diode laser bars described by Clarkson and Hanna: (a) plan view; (b) side view. (Reprinted by permission from Clarkson and Hanna (1996).)

The other broad distinction which is useful to make at this point concerns the geometry of the solid-state resonator. Most of the resonator designs discussed here consist of a number of discrete elements, such as the Nd:YAG crystal, mirrors, lenses, polarizing elements, and possibly additional elements required for pulsed or single-frequency operation; an example is shown in Figure 3.23. However, for some purposes, a monolithic design is preferable, in which the reflectors that define the laser resonator are deposited directly on polished surfaces of the laser crystal. The example shown in Figure 3.24 is a particularly elegant version of this approach, a monolithic single-frequency, unidirectional ring laser.

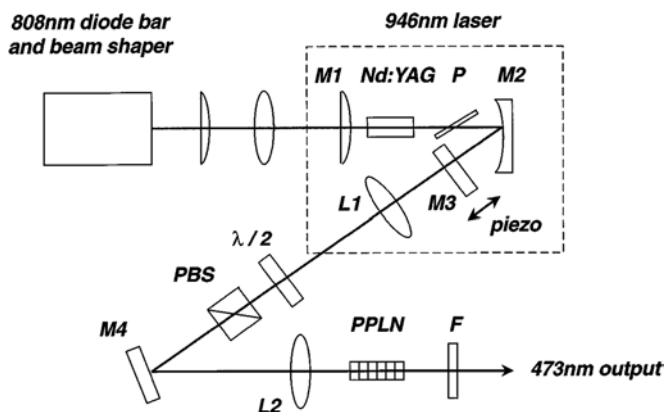


Figure 3.23: Experimental arrangement for generation of blue light by SHG of an end-pumped 946-nm Nd:YAG laser used by Ross and coworkers: $M1$, $M2$ are mirrors with radii of curvature of 100 and 25 mm, respectively; $M3$, $M4$ are planar mirrors; $L1$, $L2$ are lenses with focal lengths of 150 and 50 mm, respectively; P is a polarizing slide; $\lambda/2$ is a half-waveplate; PBS is a polarizing beamsplitter; F is an infrared filter. (Reprinted with permission from Ross *et al.* (1998).)

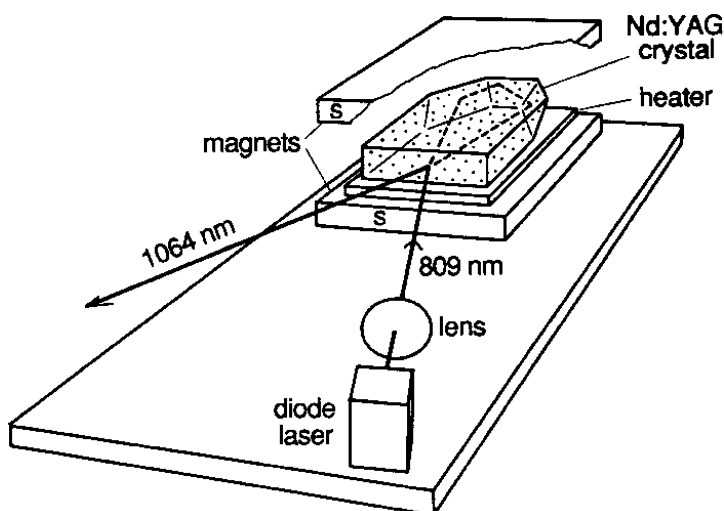


Figure 3.24: Schematic diagram of diode-laser pumped monolithic nonplanar Nd:YAG ring oscillator. (Reprinted by permission from Byer (1988).)

Most of the more recent work on direct, single-pass SHG of diode-pumped solid-state lasers has exploited new periodically-poled materials, like PPLN and PPKTP, the advantages of which were discussed in Chapter 2. Here, we review a few of these results in order to show what can be achieved using this approach.

3.3.1 Frequency-doubling of 1064-nm Nd:YAG lasers

Webjörn *et al.* (1995) reported achieving a 55% conversion efficiency using a 4-mm long PPLN crystal (period $\Lambda = 6.8 \mu\text{m}$, phasematching temperature $T_{pm} = 35^\circ\text{C}$) which provided first-order QPM for single-pass SHG of a Q-switched Nd:YAG laser (18.5-ns long pulses at a repetition rate of 2 kHz, with a longitudinal mode structure resulting in a factor $\rho = 1.8$). They achieved an average green output power of 30 mW using an average infrared power of 55 mW. The M^2 value of the second harmonic was 1.2, indicative of excellent beam quality.

Later, Pruneri *et al.* (1996) reported using a 3.2-mm long PPLN crystal ($\Lambda = 6.35 \mu\text{m}$, $T_{pm} = 102^\circ\text{C}$) for frequency-doubling of the picosecond pulses from an amplified mode-locked Nd:YLF laser. They achieved $\sim 52\%$ efficiency, corresponding to an average green power of 330 mW, obtained from 640 mW of average infrared power (in the form of pulses of ~ 2 ps duration with a repetition rate of 105 MHz).

More recently, Englander *et al.* (1997) reported using PPKTP for efficient single-pass SHG of a Nd:YAG laser. They used a 10-mm long PPKTP crystal ($\Lambda = 9.01 \mu\text{m}$, $T_{pm} \sim 30^\circ\text{C}$) and achieved $\sim 64\%$ conversion efficiency, yielding 4.8 W of average green power from 7.5 W of average infrared power (in the form of pulses of 120 ns duration with a repetition rate of 50 kHz, running in multiple longitudinal modes).

In the cw regime, Miller *et al.* (1997) have reported achieving 42% conversion efficiency using a 53-mm long PPLN crystal ($\Lambda = 6.5 \mu\text{m}$, $T_{pm} = 199.5^\circ\text{C}$). They obtained a green power of 2.7 W from an infrared power of 6.5 W, using a single-frequency Nd:YAG laser.

3.3.2 Frequency-doubling of 946-nm Nd:YAG lasers

Pruneri *et al.* (1995) reported efficient cw generation of 473-nm blue light by single-pass SHG of a diode-pumped Nd:YAG laser operating at 946 nm. They used a 6-mm-long PPLN crystal ($\Lambda = 4.6 \mu\text{m}$, $T_{pm} = 68.5^\circ\text{C}$). They used a 20-W diode laser bar combined with the two-mirror beam-shaping technique described above to pump the Nd:YAG laser, which produced a polarized output of 1.5 W at 946 nm with an $M^2 \leq 1.5$, and oscillated in several longitudinal modes. They produced 49 mW of cw blue power from 1.28 W of infrared light. At these high blue power levels and at the temperature used, the M^2 value of the second-harmonic beam increased to ~ 3 , indicating some photorefractive effects.

More recently, Ross *et al.* (1998) reported improvements to this system (Figure 3.23) which have resulted in generation of 450 mW of average blue power.

They used a 15-mm-long PPLN crystal ($\Lambda = 4.5 \text{ } \mu\text{m}$, $T_{pm} = 140 \text{ } ^\circ\text{C}$). The higher phasematching temperature helped mitigate the photorefractive effects observed by Prunerì and colleagues. By operating the laser in a driven-relaxation-oscillation mode, they were able to achieve pulses of ~ 300 -ns duration with a repetition rate of 160 kHz. The longitudinal mode structure of the laser was such that ρ was estimated to be ~ 1.7 . The average power internal to the uncoated PPLN crystal was 1.13 W, and this produced an internal average second-harmonic power of 450 mW.

3.3.3 Sum-frequency mixing

Although most of the more recent work on blue-green light generation using single-pass nonlinear upconversion of diode-pumped solid-state lasers has involved SHG, some work has also been done using SFG. For example, Moosmüller and Vance (1997) reported generating light at 589 nm for high-resolution spectroscopy of the sodium D_2 line by mixing 1064-nm light from one diode-pumped Nd:YAG laser with the 1319-nm light from a second diode-pumped Nd:YAG laser in a 50-mm long crystal of congruent LN (phasematching temperature = $227.5 \text{ } ^\circ\text{C}$). They generated 3.4 mW cw using 0.7 W at 1064 nm and 0.35 W at 1319 nm. In this experiment, both lasers were of the monolithic ring design shown in Figure 3.24.

3.4 SUMMARY

In this chapter, we have considered single-pass SHG and SFG. Although this approach offers the most straightforward way to generate blue-green light using nonlinear upconversion of infrared lasers, achieving milliwatt and higher levels of blue-green power requires both powerful lasers and efficient nonlinear materials. Only since 1995 have these requirements been met. Diode lasers having high power levels combined with good spatial-mode quality and a narrow spectrum are now available, and the infrared output of these lasers can be frequency-doubled directly. Diode-pumped solid-state lasers have many advantages and have also been used for efficient single-pass SHG and SFG. The improvement of growth techniques for crystals like KN and the development of periodically-poled nonlinear materials have also enabled successful demonstrations of single-pass doubling.

In the next chapter, we will consider enhancing the single-pass conversion efficiency by placing the nonlinear crystal in an optical resonator. This approach can increase the efficiency of nonlinear conversion, but generally creates a more complicated system than is demanded by single-pass conversion.

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4

Resonator-enhanced SHG and SFG

4.1 INTRODUCTION

In the preceding chapter, we considered single-pass SHG and SFG. There, we saw that efficient frequency upconversion from infrared to blue-green wavelengths is generally possible only when the power at the fundamental wavelength is several watts. The approach to achieving such powers that we considered in Chapter 3 was a very direct and “brute force” one: build a more powerful laser. We examined several approaches that have been used for increasing the infrared power available for the nonlinear interaction, including:

- using a power amplifier to boost the output of a master oscillator;
- using high-power diode lasers that have poor spectral and spatial characteristics for pumping solid-state lasers which then act as sources for frequency-doubling;
- using pulsed, rather than cw, operation in order to achieve higher peak powers.

While these brute force approaches have the advantage of being conceptually straightforward, it has only been since about 1995 that they have succeeded in producing blue-green powers sufficient for some of the applications described in Chapter 1. In addition, these approaches suffer from a number of practical disadvantages. The powerful lasers required for efficient single-pass conversion tend to be complicated and expensive, and since they generate high powers they require substantial electrical power and thermal management. Furthermore, although pulsed configurations have succeeded in producing large average blue-green powers, the power generated by cw operation has been too low for many applications.

Given these disadvantages, several alternatives have been pursued with the goal of producing high cw blue-green powers using more modest infrared sources than are demanded by single-pass frequency upconversion. In this chapter, we will consider one such approach: resonator-enhanced SHG harmonic and SFG. In subsequent chapters, we will consider intracavity and waveguide configurations.

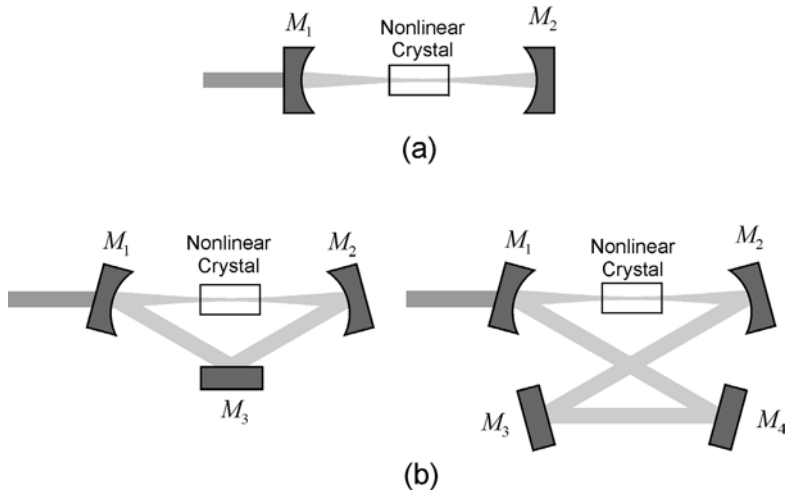


Figure 4.1: The two basic configurations of resonators for enhancement of SHG and SFG: (a) standing-wave resonator; (b) ring resonators.

The basic idea behind resonator-enhanced SHG and SFG is simple: since only a small fraction of the fundamental energy is depleted in a single pass through the nonlinear crystal, the remaining infrared energy can be redirected by mirrors so that it interacts with the crystal again. For example, Figure 4.1(a) shows a nonlinear crystal placed between two mirrors which repeatedly redirect and refocus the infrared energy so that it can interact many times with the nonlinear crystal. Since cw light is most often used with this approach, successive passes of the light through the crystal interfere with each other, and it is appropriate to describe this approach in terms of an optical resonator. From this point of view, we would say that the mirrors define a stable optical resonator, in which successive passes of the fundamental reinforce each other, so that the infrared intensity circulating inside the resonator becomes many times higher than the intensity incident upon the resonator. Resonating the fundamental wavelength has been the most widely used approach for enhancing the efficiency of SHG or SFG; however, it is also possible to increase the efficiency by resonating the second harmonic, or by resonating both the fundamental and second harmonic, although this can be difficult in practice.

Two basic resonator geometries are shown in Figure 4.1 – standing-wave resonators and ring resonators.¹ Ring resonators can be further divided into two

¹ We note in passing that such resonators are often also called “cavities”. While this nomenclature is an accurate description of a resonator consisting of an arrangement of mirrors with nothing but air or vacuum between them, it has also been applied – somewhat inaccurately, from a semantic point of view – to resonators in which the “cavity” is partially or completely filled with dense material, such as a nonlinear crystal. Because this nomenclature is so thoroughly entrenched, and for the sake of variety, we will occasionally use the term “cavity” interchangeably with “resonator”, even to describe monolithic resonators, in which the space between the mirrors is entirely filled with crystal.

principal configurations: the three-mirror “triangle” configuration and the four-mirror “bow tie” configuration (Figure 4.1(b)).

While standing-wave resonators are simpler than ring resonators, they have two main disadvantages. The first is that the fundamental light passes through the nonlinear material in both the forward and backward directions, so that two oppositely-travelling second-harmonic beams are generated, only one of which is generally usable. (It is possible to retroreflect the backward-travelling second-harmonic beam, so that it joins the forward-travelling beam; however, care must be taken to adjust the phase so that it adds to the power in the forward-travelling beam, rather than subtracting from it.) The second is that some fraction of the incident fundamental light is reflected directly back towards the source. Such feedback can induce instabilities in the source, particularly if the source is a semiconductor diode laser. Because of these disadvantages, ring resonators have generally been preferred for enhancement of SHG and SFG. In addition, ring resonators also have the advantage that they are relatively insensitive to misalignments of the mirrors in the plane of the ring, since the ring path can simply change slightly to accommodate the misalignment. However, one disadvantage of ring resonators is that astigmatism is introduced when curved mirrors are used for oblique-incidence reflection, and this can complicate the problem of achieving optimum focusing in the nonlinear crystal (Hanna, 1969). Various approaches to cancelling astigmatism (Kogelnik *et al.*, 1972) or eliminating it through symmetry (Johnston *et al.*, 1972) can be used.

Both standing-wave and ring resonators have been implemented in two forms. In the implementations shown in Figure 4.1, the mirrors and nonlinear crystal are discrete elements. However, it is possible to combine the mirrors and nonlinear crystal into one element, by polishing and coating appropriate surfaces of the nonlinear crystal so that they act as reflectors. Monolithic resonators have the advantage that they contain fewer loss-inducing surfaces than discrete designs. This is important because reducing the loss encountered by a wave circulating inside the resonator is a key design objective for efficient blue-green light generation. In addition, monolithic resonators generally have better frequency stability than resonators assembled from discrete components. Both standing-wave and ring resonator designs have been implemented in monolithic form; Figure 4.2 shows a few ring resonator designs, which are discussed in greater detail below.

In this chapter, we will consider how an optical resonator should be designed in order to provide the maximum enhancement of SHG and SFG. We will discover the following considerations to be crucial:

- The loss within the resonator must be minimized.
- A number of “matching” conditions must be satisfied in order to maximize the build up of the fundamental power within the resonator:

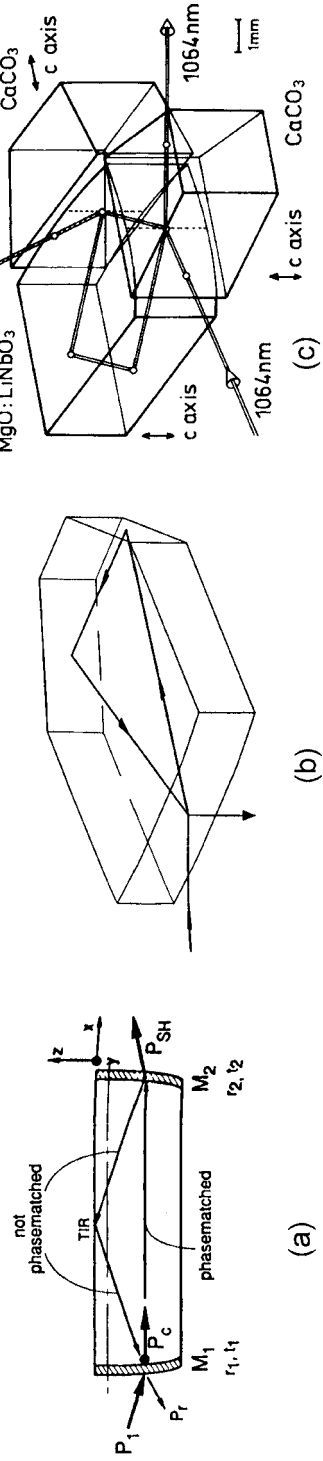


Figure 4.2: Three designs of a monolithic ring resonator for enhancement of SHG: (a) monolithic ring resonator configuration with three reflections, two from surfaces having mirror coatings and one from total internal reflection (TIR); (b) monolithic ring resonator configuration with three reflections, one from a mirror-coated surface and two from TIR; (c) monolithic ring resonator configuration with four reflections, all from TIR. ((a) Reprinted with permission from Kozlovsky *et al.* (1988). Copyright IEEE. (b) Reprinted with permission from Gerstenberger *et al.* (1991); (c) Reprinted with permission from Fiedler *et al.* (1993).)

- The input mirror reflectivity must be “impedance matched” to the resonator (analogous to impedance-matched termination of an electrical transmission line).
- The frequency of the input beam must be matched to a resonant frequency of the cavity.
- The input beam must be spatially mode-matched to the resonator.

We begin by developing a mathematical description of the build up of intensity in a resonator, from which the need to maintain these conditions will emerge naturally.

4.2 THEORY OF RESONATOR ENHANCEMENT

Given a fundamental power P_{inc} incident upon a resonator, what circulating power develops inside it? The analysis of this problem was treated in a classic paper by Ashkin *et al.* (1966). Kozlovsky *et al.* (1988) later extended this analysis to include the loss at the fundamental wavelength induced by conversion to the second-harmonic. In this section, we will follow the general outline of these treatments in order to develop the basic equations describing resonator SHG. However, the notation we will use here departs somewhat from that used by Ashkin *et al.* and Kozlovsky *et al.* and follows more closely that used by Siegman (1986), which makes explicit the phase relationship between the beams transmitted through and reflected from the resonator mirrors and provides a clear way to distinguish between amplitude coefficients and power coefficients.

We assume the resonator configuration shown in Figure 4.3, in which the mirror M_1 has an amplitude reflection coefficient of r_1 and an amplitude transmission coefficient of jt_1 , where r_1 and t_1 are complex numbers. This particular representation of the reflection and transmission coefficients results from the choice of a specific reference frame, one in which the coefficients are the same no matter which side of the mirror the light is incident upon. The $\pi/2$ phase shift inherent between the reflected and transmitted waves is also expressed in a very explicit way in this reference frame. The corresponding power reflection coefficient is $R_1 = |r_1|^2$

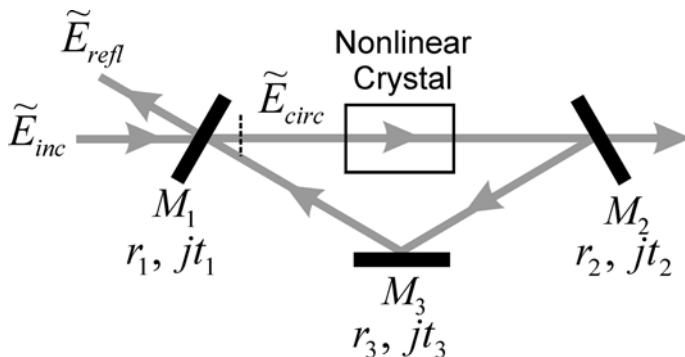


Figure 4.3: Geometry for analysis of resonator enhancement.

(where we will consistently use lower-case letters to represent amplitude coefficients and upper-case letters to represent power coefficients) and the corresponding power transmission coefficient is $T_1 = |t_1|^2$, where $R_1 + T_1 = 1$ (we assume that there are no losses due to scattering or absorption in the mirrors). What we actually measure in practice are *power* transmission and reflection coefficients, but in developing the theory we deal with *amplitudes*; thus, we need a notation that easily allows us to move between the two. Similar notation applies for mirror M_2 , and we assume mirror M_3 to be perfectly reflecting ($R_3 = 1$).

When cw light is coupled into the resonator, the interference of successive passes of the light through the resonator gives rise to a steady-state “circulating” electric field. If we follow this circulating field through one round trip in the resonator, we find that it decreases in energy due to the losses present in the resonator. In order for the circulating field to be self-consistent, the lost energy must be replenished by energy coupled into the resonator through mirror M_1 .

What losses does the circulating field encounter? First, it encounters the nonlinear crystal, and may suffer loss as a result of reflection or scattering from its input and output surfaces, absorption or scattering from within the bulk of crystal, or conversion to the second harmonic. We will account for all the losses related to the crystal, other than the loss due to SHG, through a power transmission factor T_x . The SHG loss we will account for through a transmission factor $T_{SH} = (1 - \gamma_{SH} P_{circ})$, where $\gamma_{SH} P_{circ} = \eta_{SH}$, the single-pass conversion efficiency. From the results for SHG from a focused gaussian beam given in Chapters 2 and 3, we can write γ_{SH} in the form given by Kozlovsky *et al.* (1988):

$$\gamma_{SH} = \left(\frac{2\omega^2 d_{eff}^2 k_\omega}{\pi n^3 \epsilon_0 c^3} \right) Lh(B, \xi) \quad (4.1)$$

where these quantities are as defined previously. As a rule of thumb, for LN with $L = 1$ cm and optimum focusing, $\gamma_{SH} \sim 0.003/\text{W}$.

Thus, after passing through the crystal, the electric field is $t_x t_{SH} \tilde{E}_{circ}$, where t_x and t_{SH} are the amplitude coefficients corresponding to T_x and T_{SH} , and $\tilde{E}_{circ}$ is the complex amplitude of the circulating electric field (that is, the actual field is $\frac{1}{2}(\tilde{E}_{circ} e^{j\omega_1 t} + \tilde{E}_{circ}^* e^{-j\omega_1 t})$). After the circulating field exits the nonlinear crystal, it is reflected from mirror M_2 , so that the field is $r_2 t_x t_{SH} \tilde{E}_{circ}$. We assume mirror M_3 to be perfectly reflective, so that no change in amplitude occurs as a result of reflection from that element.

What we have determined so far is the fraction of power leaving mirror M_1 that returns to M_1 after traversing the resonator – we might therefore think of this as the fraction of power that has been “reflected” by the remainder of the resonator. Thus, it is convenient to define a “resonator reflectance parameter”, $r_m = r_2 t_x t_{SH}$ that expresses this fraction.

Continuing our trip around the resonator, the field returning to M_1 is then reflected from mirror M_1 , so that the field amplitude after one complete round trip is $r_1 r_m \tilde{E}_{circ} e^{j\phi}$, where we have included $e^{j\phi}$ to account for the accumulated round-trip phase associated with propagation around the resonator and reflection from the mirrors. Clearly, this amplitude is lower than what we started out with. In order to maintain a steady-state condition, the externally incident field must replenish the circulating field. Thus, we must have the self-consistent condition that

$$\tilde{E}_{circ} = r_1 r_m \tilde{E}_{circ} e^{j\phi} + j t_1 \tilde{E}_{inc} \quad (4.2)$$

We can solve Equation (4.2) to obtain

$$\frac{\tilde{E}_{circ}}{\tilde{E}_{inc}} = \frac{j t_1}{1 - r_1 r_m e^{j\phi}}$$

or, in terms of the corresponding powers:

$$\frac{P_{circ}}{P_{inc}} = \frac{t_1^2}{1 - 2r_1 r_m \cos \phi + r_1^2 r_m^2} = \frac{1 - R_1}{1 - 2\sqrt{R_1}\sqrt{R_m} \cos \phi + R_1 R_m} \quad (4.3)$$

where the latter expression has been written in terms of power reflection and transmission coefficients. When ϕ is an integer multiple of 2π , then $\cos \phi = 1$, the resonance condition is satisfied, and the ratio of circulating to incident power takes on its maximum value:

$$\left. \frac{P_{circ}}{P_{inc}} \right|_{res} = \frac{1 - R_1}{(1 - \sqrt{R_1}\sqrt{R_m})^2} \quad (4.4)$$

With this basic equation, we can begin to examine how to maximize the blue-green power generated by SHG or SFG.

4.2.1 The impact of loss

First, for simplicity, we will assume that the loss due to SHG can be neglected, so that R_m depends only on fixed, nonintensity-dependent losses. We can then plot $P_{circ}/P_{inc}|_{res}$ as a function of R_1 for various values of R_m , as shown in Figure 4.4. Two important properties of power-enhancement resonators are evident from this figure. The first is that the larger R_m is (that is, the lower the round-trip losses are), the greater will be the power enhancement on resonance; thus, we would like to make the round-trip loss in the cavity as low as possible.

Reduction of the round-trip loss has been one of the factors motivating the development of monolithic nonlinear resonators for efficient SHG. In these devices, polished surfaces of the nonlinear crystal itself are used as reflectors. These surfaces

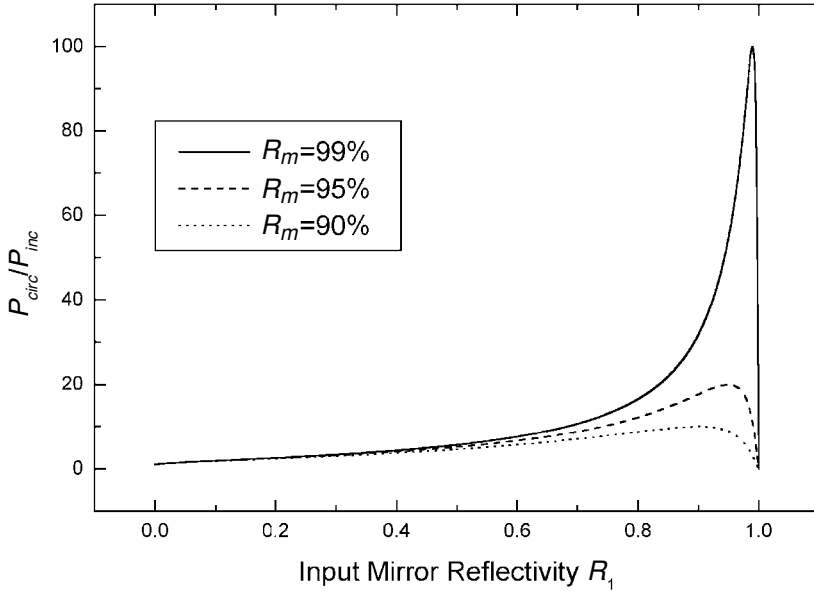


Figure 4.4: Ratio of circulating power to incident power as a function of input mirror reflectivity for different values of the cavity reflectance parameter.

may be flat, or may have some curvature; they may reflect by total internal reflection (TIR) or may be covered with multi-layer dielectric mirror coatings. Filling the resonator with the nonlinear crystal in this way eliminates interfaces which would otherwise need to be anti-reflection-coated and which could give rise to loss through scattering or reflection. Both standing-wave and ring resonator designs have been developed as shown in Figure 4.2. The design used by Kozlovsky *et al.* (1987, 1988, 1990) in both LN and KN uses two curved, coated surfaces and a third flat, uncoated surface, which reflects by TIR (Figure 4.2(a)). Gerstenberger *et al.* (1991) used a design that employs a single curved, coated surface and two flat, TIR surfaces (Figure 4.2(b)). Fiedler *et al.* (1993) used a design consisting of four reflecting surfaces, two curved and two flat, all of which employ TIR (Figure 4.2(c)). Coupling into and out of the resonator was accomplished using prisms for evanescent-wave excitation.

As illustrated in Figure 4.2(a), one feature of monolithic ring resonators is that generally only one leg of the ring path lies along the phasematching direction. In the triangular resonator shown in Figure 4.2(a), the second harmonic is generated only along the leg connecting the two curved mirrors, so that only a single blue-green beam is emitted from the device. The design of monolithic ring resonators can be subject to further constraints of a problem known as “bireflection”, which is described in Section 4.3.3.

4.2.2 Impedance matching

The second important property of power-enhancement resonators evident from Figure 4.4 is that $P_{circ}/P_{inc|res}$ is maximized when $R_1 \approx R_m$, a condition known as “impedance matching.” Thus, in order to maximize the power enhancement produced by the resonator, we would like to be able to accurately estimate the cavity reflectance parameter R_m and then choose the reflectivity of mirror M_1 to match. In practice, it is sometimes difficult to accurately determine R_m . Figure 4.4 indicates that the variation of $P_{circ}/P_{inc|res}$ with R_1 is less rapid when $R_1 < R_m$ than when $R_1 > R_m$. Thus, depending on how much confidence we have in our estimate of R_m (and in the ability of the mirror manufacturer to achieve the specified value of R_1), we may wish to design our resonator so that R_1 is slightly smaller than our estimated optimum, since the margin for error is larger in that direction.

Now let us consider the beam reflected from the resonator. It consists of two parts: a portion of the incident beam that is directly reflected from mirror M_1 , and a portion of the circulating beam that is leaked through mirror M_1 . Thus, we can write its amplitude as

$$\tilde{E}_{refl} = r_1 \tilde{E}_{inc} + jt_1 r_m e^{j\phi} \tilde{E}_{circ}$$

or

$$\frac{\tilde{E}_{refl}}{\tilde{E}_{inc}} = r_1 - \frac{t_1^2 r_m e^{j\phi}}{1 - r_1 r_m e^{j\phi}} = \frac{r_1 - r_m e^{j\phi}}{1 - r_1 r_m e^{j\phi}}$$

On resonance, we obtain

$$\left. \frac{\tilde{E}_{refl}}{\tilde{E}_{inc}} \right|_{res} = \frac{r_1 - r_m}{1 - r_1 r_m}$$

or, in terms of powers,

$$\left. \frac{P_{refl}}{P_{inc}} \right|_{res} = \left(\frac{\sqrt{R_1} - \sqrt{R_m}}{1 - \sqrt{R_1 R_m}} \right)^2 \quad (4.5)$$

We see that when the impedance-matching condition $R_m = R_1$ is met, the amplitude of the reflected field is zero, indicating that all the power incident upon the resonator is coupled into it. Thus, making $R_m = R_1$ is very similar to the familiar problem of matching the impedance Z_L of an electrical load to the characteristic impedance Z_0 of a transmission line. When $Z_L = Z_0$, there is no reflection from the termination, and the maximum possible power is delivered to the load. In a similar way here, when $R_m = R_1$, no light is reflected from the resonator (on resonance), and maximum power is coupled into the cavity.

Earlier, we assumed for convenience that we could neglect the loss due to SHG; thus, in evaluating R_m , we assumed that $T_{SH} = 1$. However, if the resonator is

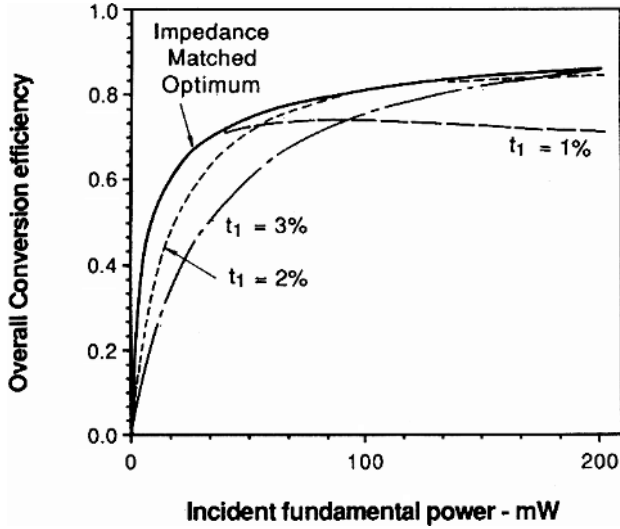


Figure 4.5: Plot of nonlinear conversion in a monolithic LN ring resonator versus incident power at the fundamental wavelength. (Reprinted with permission from Kozlovsky *et al.* (1988). Copyright: IEEE.)

performing its intended function of increasing the conversion efficiency, then the loss due to SHG cannot be neglected. If we write out the full expression for the cavity reflectance parameter, we have $R_m = R_2 T_x^2 (1 - \gamma_{SH} P_{circ})^2$, which shows that R_m depends on P_{circ} ; thus, Equation (4.1) becomes a cubic equation in P_{circ} . Although this equation can be solved analytically, it is perhaps most conveniently solved numerically for specific cases of interest. Figure 4.5 shows conversion efficiency as a function of incident pump power obtained by solving Equation (4.1) for a monolithic LN ring resonator, 1-cm long, with $T_x = 0.997$ and $R_2 = 1$. Three of the curves in the figure assume a fixed value for the power transmission coefficient of the input mirror M_1 (namely, 1%, 2% or 3%), as would be the case in practice. The fourth curve shows the best performance we could possibly hope to obtain if the input mirror reflectivity could somehow be tuned to its optimum value for each incident power level. Suppose we examine the curve corresponding to 2% transmission ($R_1 = 0.98$) as a specific example. At very low levels of incident power, where SHG is negligible, $R_m \approx T_x^2 = 0.997$, so that $R_m > R_1$. At some value of the incident fundamental power, however, the conversion of the fundamental to the second harmonic will be such that the additional loss due to SHG will make $R_m = R_1$, and at that specific power level, the resonator will be impedance-matched. At a higher value of fundamental power, $R_m < R_1$. Thus, the resonator will again not be impedance-matched, and the conversion efficiency will be lower than the impedance-matched optimum, as is shown in the figure.

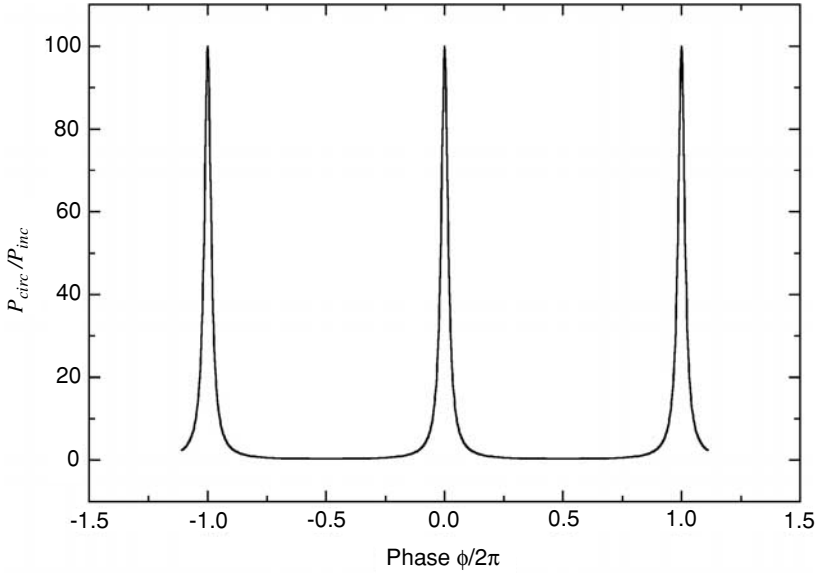


Figure 4.6: Resonance peaks in $P_{\text{circ}}/P_{\text{inc}}$ as a function of round-trip phase ϕ .

4.2.3 Frequency matching

What must we do in order to achieve the resonant condition? If we plot $P_{\text{circ}}/P_{\text{inc}}$ as a function of the accumulated round-trip phase ϕ , we obtain a graph like the one in Figure 4.6, which assumes $R_1 = R_m = 0.99$. We see that near $\phi = 0$, there is a sharp peak, and that such a peak occurs whenever $\phi \approx 2m\pi$, where m is an integer. From Equation (4.3), we can calculate the amount $\Delta\phi$ by which ϕ can deviate from such a resonance value before $P_{\text{circ}}/P_{\text{inc}}$ drops to half of its on-resonance value. The result is (assuming a fairly sharp resonance, so that a small-angle approximation in $\Delta\phi$ can be used):

$$\Delta\phi = \frac{1 - \sqrt{R_1 R_m}}{(R_1 R_m)^{\frac{1}{4}}} \quad (4.6)$$

The phase ϕ accumulated in a round trip is $2\pi l_{\text{opt}}/\lambda$, or $2\pi l_{\text{opt}}f/c$, where $l_{\text{opt}} = \sum_i n_i l_i$ is the optical length of the cavity, given by the sum of the products of refractive index and length of each of the elements present in the cavity – including, of course, the empty space between elements. (For simplicity, we have left out of this expression fixed phase shifts which may be associated with reflection from the cavity mirrors.) Thus, by adjusting the cavity length or the frequency of the laser, we can satisfy the resonant condition. If we recast the resonance condition $\phi = 2m\pi$ in terms of frequency, we obtain $f_{\text{res}} = mc/l_{\text{opt}}$, and the spacing between such resonance frequencies is thus $\Delta f_{\text{res}} = c/l_{\text{opt}}$, which is usually called the “free

spectral range” (FSR) or the “longitudinal (or axial) mode spacing” of the resonator. If we recast Equation (4.6) in terms of a frequency difference, and double the value to obtain a full-width-at-half-maximum bandwidth, we obtain:

$$\Delta f_{FWHM} = \frac{c}{\pi l_{opt}} \frac{1 - \sqrt{R_1 R_m}}{[R_1 R_m]^{\frac{1}{4}}} = \frac{\Delta f_{res}}{\pi} \frac{1 - \sqrt{R_1 R_m}}{[R_1 R_m]^{\frac{1}{4}}} \quad (4.7)$$

Note that this bandwidth depends strongly on the loss present in the resonator, expressed by R_m . The “finesse” $\mathcal{F} = \Delta f_{res} / \Delta f_{FWHM}$ of a resonator is often used to express the “sharpness” of the resonant peak.

As an example, the standing-wave LN resonator described by Kozlovsky and coworkers had a physical length of 25 mm and a refractive index of 2.23; thus we would expect

$$\Delta f_{res} = \frac{3 \times 10^8 \text{ m/s}}{2 \times 2.23 \times (25 \times 10^{-3} \text{ m})} = 2.7 \text{ GHz}$$

The reflectivity of the mirror M_1 was $R_1 = 0.997$, that of mirror M_2 was $R_2 = 0.997$ and the round-trip transmission was $T_x = 0.992$; thus, when no second harmonic is being generated we have $R_m = R_2 T_x = 0.989$, and $R_1 R_m = 0.986$. If this value is inserted into Equation (4.7), we calculate $\Delta f_{FWHM} = 6.1 \text{ MHz}$, and $\mathcal{F} \approx 450$. Thus, in order to achieve significant build up of the fundamental intensity, we must be able to tune the laser frequency to within $\pm 3 \text{ MHz}$ of the exact resonance frequency and hold it there. In addition, the linewidth of the laser oscillation must be narrow in comparison with this value. Thus, the spectral purity, frequency tunability, and frequency stability of the infrared laser are very important issues for resonator-enhanced SHG and SFG. In addition, we saw that the value of the resonant frequency we are trying to match is a sensitive function of the round-trip optical path length in the resonator. If this optical path length changes – which might result from the physical expansion of the resonator, or a temperature-induced change in refractive index – the value of f_{res} will also change. Thus, the frequency stability of the resonator is also a concern.

4.2.4 Approaches to frequency locking

For a practical device, the coincidence between the laser frequency and a resonant frequency of the cavity must be established and maintained by some sort of “locking” technique. The techniques can be grouped into two broad categories: electronic (active) locking and optical (passive) locking. In electronic locking, either the frequency of the laser or the resonant frequency of the cavity is actively tuned in order to bring the two into coincidence. Generally, an electrical error signal is generated (we shall see how below) that indicates the magnitude of the discrepancy between the laser frequency and cavity resonant frequency and the direction in which the

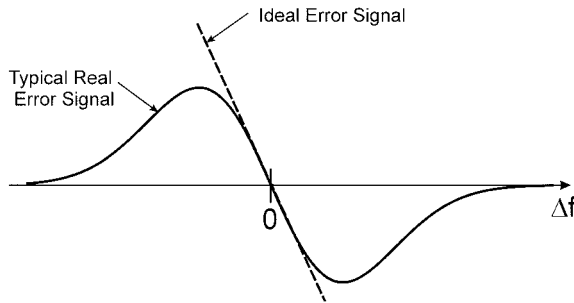


Figure 4.7: Characteristics of the error signal for electronic locking.

frequency of the tunable element must be changed in order to achieve coincidence. In response to this signal, the frequency of either the laser or resonator is tuned until the error becomes zero. In optical locking, frequency-dependent optical feedback is supplied to the laser in order to force it to oscillate at a resonant frequency of the build-up cavity. In Section 4.2.4.1 we will consider some techniques that have been used for electronic locking; in Section 4.2.4.2 we will consider some optical locking techniques.

4.2.4.1 Electronic locking techniques

In order for electronic locking to be used, some method must be found to generate an error signal which has a magnitude proportional to the deviation between the laser frequency and the resonant frequency of the cavity, and a sign that indicates whether the laser frequency is above or below the resonant frequency. Ideally, the signal should look something like that shown in Figure 4.7. In actual systems, the appearance of the error signal is more complex, and resembles the ideal shape only over a limited range. Thus, actual locking systems have a finite range near resonance over which they can maintain frequency matching; if the deviation exceeds this range, frequency coincidence between the laser and resonator will be lost.

Dither locking Perhaps the most straightforward approach to matching the laser frequency to a resonant frequency is dither locking. In this technique, the relative frequency between the laser and resonator is subjected to a small-amplitude, time-varying modulation, which can be accomplished either by moving the laser frequency relative to the resonant frequency or vice-versa. This frequency variation is converted to a variation in the amplitude of the reflected and transmitted beams, as is shown in Figure 4.8 for the reflected beam. Suppose that the laser is offset from the center of the resonance feature by Δf_0 . Inspection of Figure 4.8 shows that the magnitude of the induced amplitude variation is proportional to the slope of the resonance curve at $\Delta f = \Delta f_0$. Since this slope is zero for $\Delta f_0 = 0$,

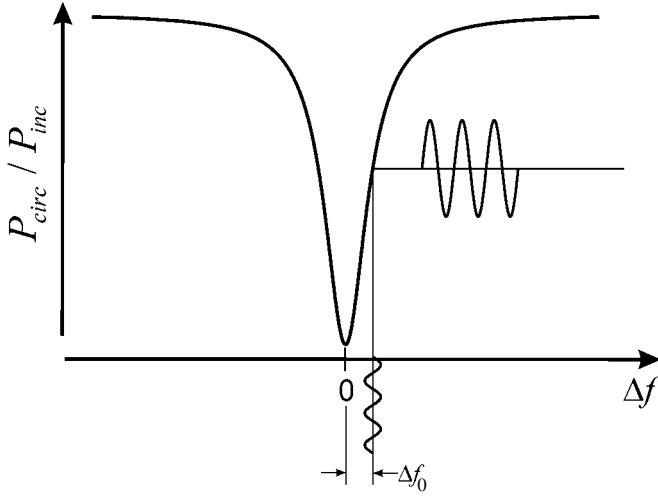


Figure 4.8: Origin of error signal in dither locking.

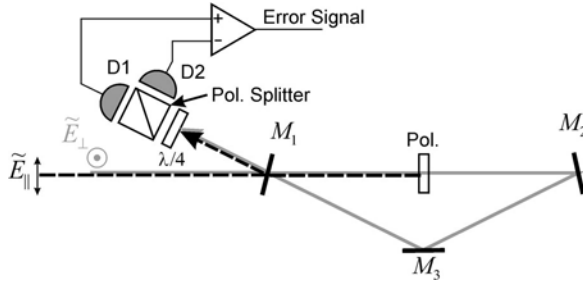


Figure 4.9: Configuration for Hänsch–Couillaud locking.

dithering provides a signal which is zero on resonance. Furthermore, since the slope is negative on one side of the resonance and positive on the other, the phase of the induced amplitude variation relative to the modulating signal changes by 180° as the laser is tuned through resonance.

In practice, the dither is often provided by moving one of the resonator mirrors with a piezoelectric element, or by applying the dither signal to the nonlinear crystal in order to change the resonant frequency electro-optically. Typically, a lock-in amplifier is used to provide phase-sensitive detection of the dither signal and to produce a dc error signal that can be fed back to tune either the laser or the resonator.

Hänsch–Couillaud locking Another approach that has been used for frequency matching is called Hänsch–Couillaud locking (Hänsch and Couillaud, 1980). The idea behind this approach is illustrated in Figure 4.9, for the case of a ring resonator. The light supplied by the laser is in a linear polarization state at some angle with respect to the plane defined by the ring; thus, it consists of a component $\tilde{E}_{inc, \parallel}$ that is

parallel to the plane of the ring, and a component $\tilde{E}_{inc,\perp}$ that is perpendicular to the plane of the ring. Since the light is linearly polarized, these two components are in phase with each other, although they may have different magnitudes. Within the ring resonator is a polarizer which blocks the $\tilde{E}_{\parallel}$ component and allows only the $\tilde{E}_{\perp}$ component to circulate around the ring. The reflected beam thus consists of a component

$$\tilde{E}_{refl,\parallel} = r_1 \tilde{E}_{inc,\parallel}$$

and a component

$$\tilde{E}_{refl,\perp} = r_1 \tilde{E}_{inc,\perp} + jt_1 r_m e^{j\phi} \tilde{E}_{circ,\perp} = \left(\frac{r_1 - r_m e^{j\phi}}{1 - r_1 r_m e^{j\phi}} \right) \tilde{E}_{inc,\perp}$$

On resonance, $\phi = 2m\pi$, and this latter component becomes

$$\tilde{E}_{refl,\perp} = \left(\frac{r_1 - r_m}{1 - r_1 r_m} \right) \tilde{E}_{inc,\perp}$$

Since the quantity in parentheses is real, $\tilde{E}_{refl,\parallel}$ and $\tilde{E}_{refl,\perp}$ are in phase with each other, so that the beam reflected from the resonator is linearly polarized at an angle to the ring plane determined by r_1 and r_m . Off resonance, there is a phase shift between $\tilde{E}_{refl,\parallel}$ and $\tilde{E}_{refl,\perp}$, the sign of which depends on the sign of $\Delta\phi$. Thus, the polarization of the reflected wave is slightly left-hand elliptical or slightly right-hand elliptical, depending on the sign of $\Delta\phi$ (Figure 4.10(a)). The reflected beam is now passed through a quarter-wave plate followed by a polarizing beamsplitter, which (in this example) transmits the component of the beam that is polarized parallel to the ring plane and reflects the component polarized perpendicular to

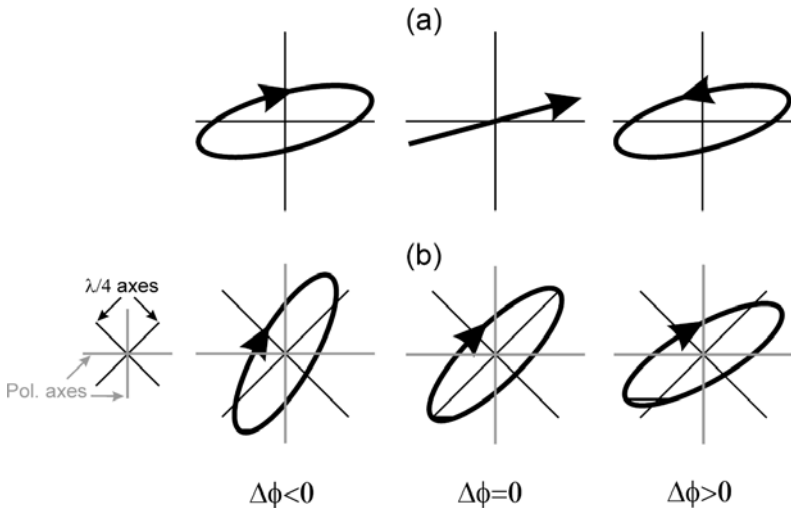


Figure 4.10: Polarization states involved in generating the error signal for Hänsch–Couillaud locking: (a) reflected from resonator; (b) after $\lambda/4$ plate.

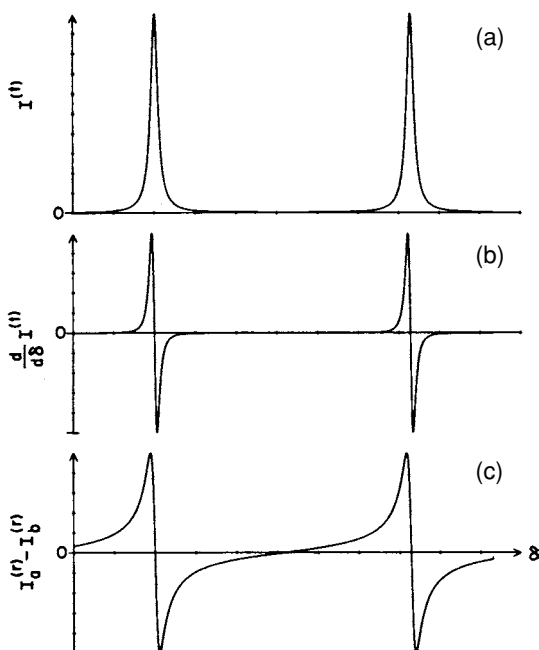


Figure 4.11: Comparison of error signals for dither locking (b) and Hänsch–Couillaud locking (c). Transmission through the resonator is shown in (a). (Reprinted with permission from Hänsch and Couillaud (1980). Copyright: Elsevier Science.)

that plane. The fast and slow axes of the waveplate are aligned at 45° to the axes of the polarization splitter. When the reflected beam passes through the quarter-wave plate, the effect of the $\lambda/4$ retardation is to change the variation of the *handedness* of the ellipse with frequency detuning into a variation of the *tilt* of the ellipse (Figure 4.10(b)). On resonance, the reflected beam has equal components along the two axes of the polarization splitter. When the laser frequency is above the resonant frequency of the cavity, one component exceeds the other, and this relationship is reversed when the laser frequency is below resonance. If the signals from the two detectors are subtracted, the result will be the error signal shown in Figure 4.11, which has the characteristics required for active locking.

Figure 4.11 also shows the error signal resulting from dither locking, illustrating the broader locking range associated with the Hänsch–Couillaud technique. This approach also has the advantage that it does not require active dithering of the laser frequency or resonator frequency, but it does require the presence of a polarizing element within the resonator (which, as described later, can be the nonlinear crystal itself).

Pound–Drever–Hall locking One of the most popular schemes for locking a laser to a SHG-enhancement resonator has been the Pound–Drever–Hall method.

The basic approach was developed by Pound for use in the microwave regime (Pound, 1946), and was extended by Drever and coworkers to the optical regime (Drever *et al.*, 1983). In this approach, the laser is weakly frequency-modulated, so that its spectrum contains a carrier component at frequency f_0 and two frequency-modulated sidebands at $f_0 \pm f_m$ which are equal in magnitude, but are 180° out-of-phase with each other. When purely frequency-modulated light is incident upon a detector, beating between the upper sideband and the carrier exactly cancels beating between the lower sideband and the carrier, so that the detected signal contains no amplitude modulation. However, if the balance between the sidebands is disrupted, this cancellation will no longer be perfect, and the signal will acquire an amplitude modulation at frequency f_m . This principle forms the basis for the technique of frequency-modulation spectroscopy (Bjorklund, 1980), in which one sideband is used to probe a spectral feature and the phase and amplitude of the resulting beat signal provide information about the absorption and dispersion associated with that feature.

This principle also provides the basis for generating the error signal in Pound–Drever–Hall locking. Suppose that a frequency-modulated signal is incident upon an impedance-matched resonator (Figure 4.12). If the carrier frequency is centered on resonance, it will be completely coupled into the cavity, and no component at frequency f_0 will be present in the reflected beam. Now suppose that the carrier

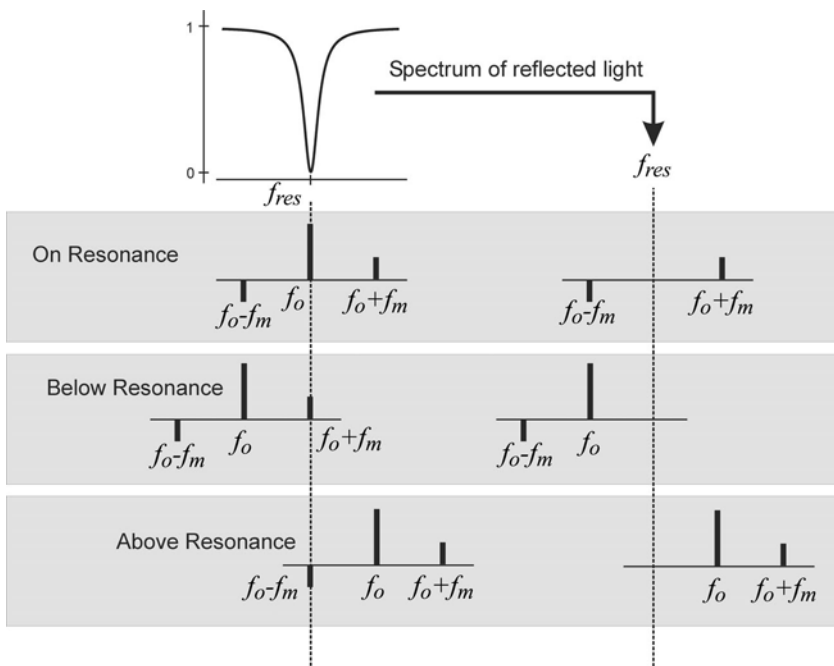


Figure 4.12: Basic principle involved in frequency-modulated locking.

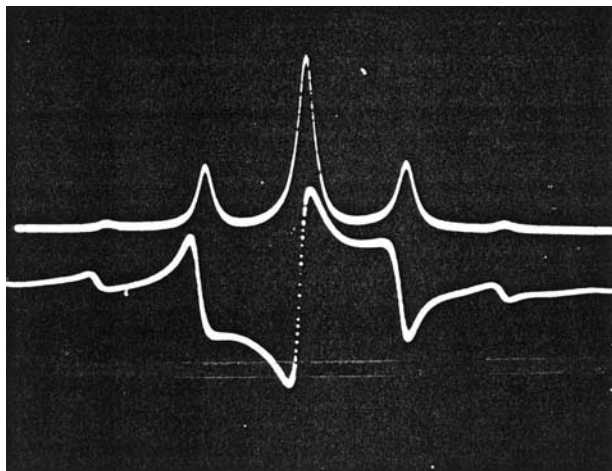


Figure 4.13: Light transmitted through resonator (above), showing existence of frequency-modulated sidebands; error signal derived from detecting the quadrature component of the reflected beam (below). (Reprinted with permission from Drever *et al.* (1983).)

frequency is detuned above resonance to such an extent that the lower sideband falls on resonance, and thus is no longer present in the reflected signal. Thus, the reflected beam will now contain components at f_0 and at $f_0 + f_m$, and the electrical current produced by the detector will contain a component at frequency f_m , corresponding to beating between the carrier at f_0 and the sideband at $f_0 + f_m$. If the carrier is detuned in the opposite direction, so that the reflected beam contains only frequencies f_0 and $f_0 - f_m$, the detector current will again contain a beat signal at f_m , but because the upper and lower frequency-modulated sidebands are 180° out of phase with each other, the phase of the resulting beat signal will also differ by 180° from the case where f_0 is detuned above resonance. Thus, if a phase-sensitive technique is used for detection (such as heterodyne detection), an error signal can be generated which satisfies the requirements for locking (Figure 4.13). A closer examination of the current produced in the detector shows that it consists of a component that is in phase with the modulating signal and a component that is in quadrature with it (i.e., is 90° out of phase) (Bjorklund, 1980). The quadrature component (shown in Figure 4.13) has a steeper slope in the vicinity of resonance and is preferred as the error signal for active locking.

A particularly elegant implementation of Pound–Drever–Hall locking is shown in Figure 4.14, in which the frequency of a GaAlAs laser diode is locked to a monolithic KN resonator (Kozlovsky *et al.*, 1990). In this case, the frequency-modulated sidebands can be conveniently generated by direct amplitude modulation of the diode laser drive current (Lenth, 1984). The modulation frequency was 325 MHz, somewhat larger than the 200 MHz width of the resonance. A doubly-balanced mixer was used for heterodyne detection, and the dc signal generated was

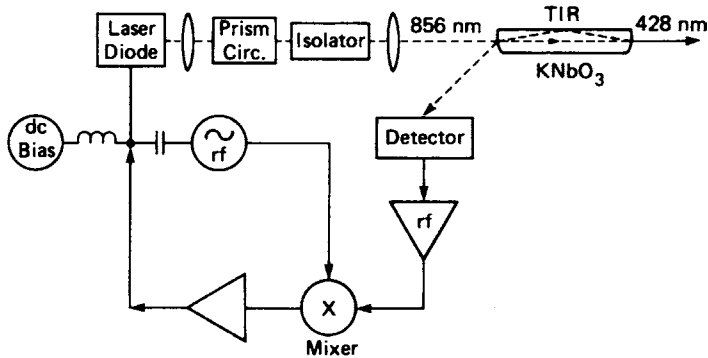


Figure 4.14: Arrangement used for frequency-modulated locking of a GaAlAs laser diode to a monolithic KN ring resonator. (Reprinted with permission from Kozlovsky *et al.* (1990).)

fed back to the diode laser to adjust the frequency. They obtained 41 mW of cw blue light using this approach.

4.2.4.2 Optical locking techniques

Another approach to matching the laser frequency to a resonant frequency of the build-up cavity and maintaining it there is referred to as “optical locking”, “passive locking”, or “self-locking”. In this approach, the idea is to inject light at the desired resonance frequency into the laser in order to force oscillation at that frequency. Light at a resonant frequency is obtained by using the resonator itself to filter out the appropriate spectral component from the emission of the laser. This approach has proven particularly effective when semiconductor laser diodes are used as the source of infrared light, because the typical output facet reflectivity is sufficiently low to render them especially sensitive to feedback. In addition to automatically locking the emission frequency of the diode laser to a cavity resonance, such frequency-selective feedback tends to narrow the linewidth of the emission, which is advantageous for nonlinear conversion. Frequency matching using optical locking techniques can be maintained for relatively large excursions of the diode laser current (Dahmani *et al.*, 1987).

Several implementations of this approach have been explored for use with resonator-enhanced SHG. One of these, reported by Dixon *et al.* (1989), exploits the frequency selectivity of the transmission characteristics of a resonator. They coupled light from a diode laser into a standing-wave resonator, then redirected some of the light transmitted through the resonator back into the diode laser (Figure 4.15). Since this transmitted signal is maximum on resonance, the diode laser receives the greatest feedback at a resonance frequency and tends to emit at that frequency.

In order for the injected light to cause stable locking, unwanted feedback resulting from spurious reflections or scattering must be prevented from reaching the

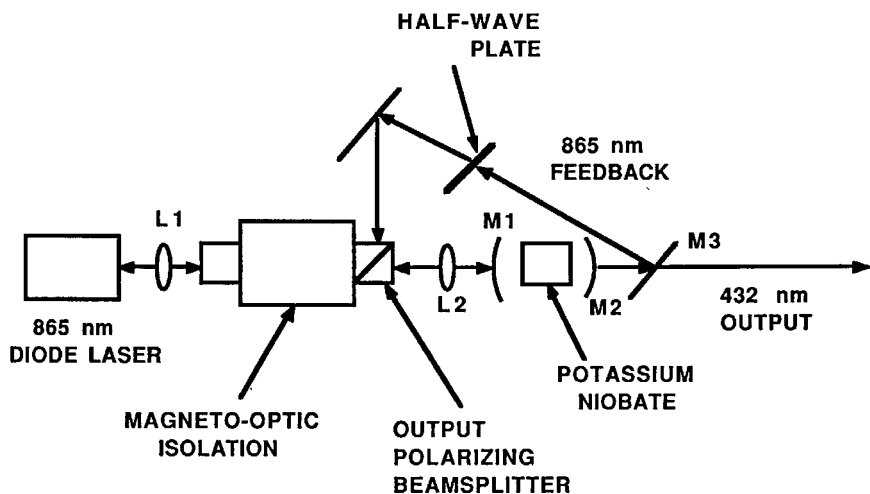


Figure 4.15: Arrangement used by Dixon and coworkers for optical locking. (Reprinted with permission from Dixon *et al.* (1989).)

laser diode. For this purpose, Dixon and colleagues used an optical isolator based on the combination of input and output polarizers and a magneto-optic Faraday rotator. In normal operation, the isolator provides low attenuation for light in the forward direction, but high attenuation for light travelling in the reverse direction. It accomplishes that unidirectional behavior in the following way. The input polarizer transmits to the Faraday rotator only light in a specific linear polarization. The Faraday element rotates this linear polarization state by 45° . The output polarizer is aligned at 45° to the input polarizer, so that it efficiently transmits this state. Now suppose some of the light emerging from the output polarizer is retroreflected (without altering its polarization state). This light passes through the output polarizer, but then is rotated by the Faraday element 45° in the sense opposite to the rotation imparted to forward-propagating light. Thus, when this backward-travelling light encounters the input polarizer it is rejected (greatly attenuated or directed away from the diode laser) because it has been rotated by 90° with respect to the original forward-travelling light.

In order to direct the desired frequency-dependent feedback to the diode laser, it is necessary to defeat the isolating properties of the Faraday device, and Dixon and coworkers did this by rotating the polarization state of the light transmitted through the resonator by 90° , so that it could be injected into the normally unused port of the output polarizing beamsplitter. When this polarization state is rotated by the Faraday element, it is rotated to match the polarization of the original diode laser light and thus is passed by the input polarizer. This 90° rotation was accomplished using a half-wave plate, the angle of which could be used to control the amount of feedback reaching the diode laser (typically ~ 0.1 – 1%).

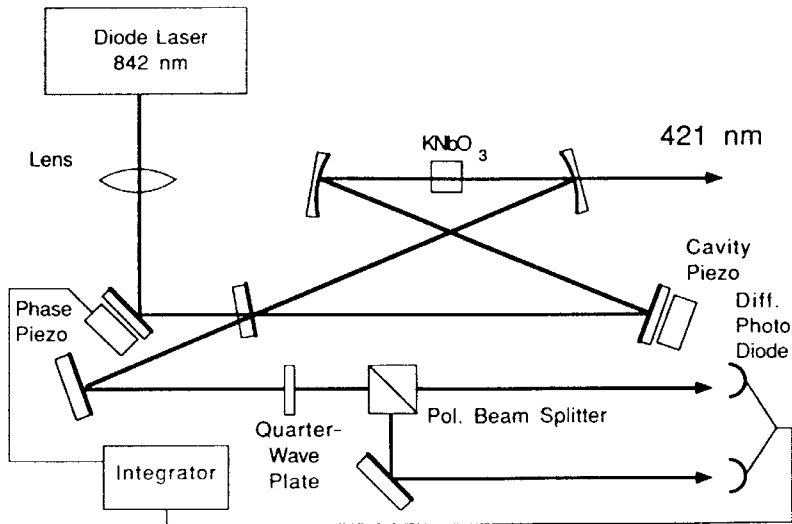


Figure 4.16: Arrangement used by Hemmerlich and coworkers for optical locking, using light scattered into the counterpropagating resonator mode. (Reprinted by permission from Hemmerlich *et al.* (1990).)

They used a 5-mm-long KN crystal placed inside a standing-wave resonator consisting of a flat input mirror ($R_1 = 98\%$) and a concave output mirror (radius-of-curvature = 9.375 mm, $R_2 = 99.7\%$) with a separation between the two mirrors that could be adjusted over the range 6–8 mm. They obtained an output power in the forward-propagating blue beam of 0.215 mW (and roughly equal power in a backward-propagating beam) from a circulating infrared power of 248 mW that was enhanced by the resonator from the input level of 12.4 mW.

Hemmerlich *et al.* (1990) used a ring resonator containing a KN crystal to enhance SHG of a 842-nm GaAlAs laser diode (Figure 4.16). Although the propagation of infrared light in a ring resonator is predominantly unidirectional, a small amount of light is coupled to the counterpropagating mode through reflection and scattering. This counterpropagating mode leaks through the input mirror and impinges on the diode laser; since this counterpropagating signal is maximized on resonance, it acts to force the diode laser to oscillate at the resonant frequency.

In addition to the frequency-dependent feedback, the diode laser receives a reflection from its output facet. The frequency-dependent feedback must be in phase with the reflection from the facet in order to properly cause the diode laser frequency to lock to a cavity resonance. The phase of the frequency-dependent feedback relative to the facet reflection is a function of the separation between the diode laser and the resonator; thus Hemmerlich and colleagues found that the stability of the optical locking was greatly improved if they also actively controlled the separation using a mirror mounted on a piezoelectric element.

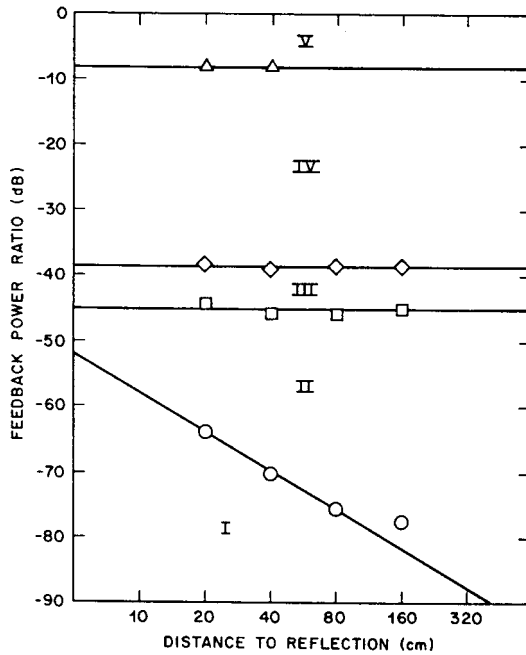


Figure 4.17: Regimes of feedback effects for a diode laser. (Reprinted with permission from Tkach and Chraplevy (1986). Copyright: IEEE.)

Hemmerlich and colleagues generated 6.7 mW of 421-nm light using this locking scheme and the bow-tie resonator configuration shown in Figure 4.16. Senoh *et al.* (1992) reported similar results using a triangle resonator. Subsequently, Hemmerlich *et al.* (1994) used this frequency-locking technique with a monolithic KN resonator to generate 22 mW of blue light.

In the configurations described so far, the fraction of the output power of the diode laser that is returned to it in the form of frequency-selective feedback is quite small ($\sim 10^{-3}$ – 10^{-4}). The behavior of laser diodes subjected to various degrees of feedback was studied by Tkach and Chraplevy (1986), who identified the five regimes of operation, as depicted in Figure 4.17. Although their work was done with 1.5- μm distributed feedback lasers, the behavior of near-infrared laser diodes used for blue-green light generation is qualitatively similar. For the feedback levels used in the experiments just described, operation is in Regime III, in which stable oscillation with a narrow linewidth is achieved. However, this stability is obtained only over a rather narrow range of feedback conditions; both Regime II with lower feedback and Regime IV with higher feedback do not provide stable operation of the laser. However, in Regime V, where the feedback is very strong, highly stable operation can be achieved. Operation in this regime generally requires applying a coating to the diode laser facet in order to greatly reduce its reflectivity. With this configuration, the diode chip acts as a short gain element in a longer “extended”

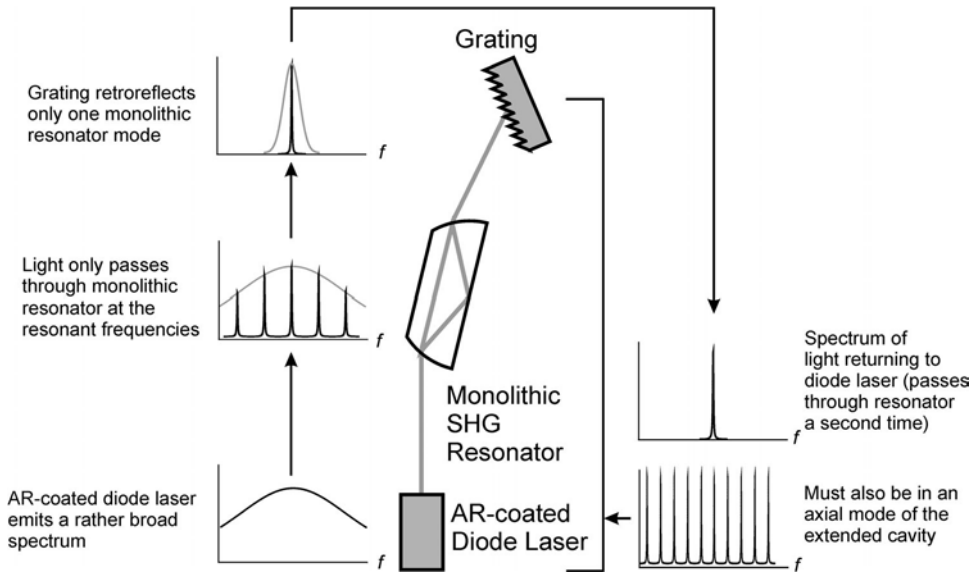


Figure 4.18: Principle behind strong optical locking approach used by Kozlovsky *et al.* (1994).

cavity defined by a reflector external to the diode laser chip; thus, such an arrangement is called an “extended cavity” or “external cavity” laser.

Kozlovsky *et al.* (1994) used strong feedback to lock the emission wavelength of such an extended cavity laser to a resonant frequency of a monolithic KN ring resonator. The operation of this device is shown in simplified form in Figure 4.18. The anti-reflection-coated diode emits light with a relatively broad spectral distribution. The resonator acts as a narrowband transmission filter, so that only those portions of the spectrum corresponding to resonant frequencies can reach the grating. The grating is used in the Littrow mode, so that light in a narrow wavelength range is directly retroreflected back along the same path. Thus, when this grating is properly adjusted, light at only one of the cavity resonant frequencies is returned to the resonator. Since this light is at a resonant frequency, it is transmitted efficiently through the resonator in the reverse direction and returns to the diode laser. The combination of the resonator and the grating thus appears to the diode laser as a very narrowband reflector, and lasing occurs at the wavelength defined by this reflector. However, the lasing frequency must also be an axial mode of the extended cavity, which is given by $f_{ax} = mc/l_{opt}$, where m is an integer; thus, it is necessary to control the length of the extended cavity in order to ensure that an axial mode coincides with a resonant frequency of the monolithic resonator.

The complete configuration used in the experiment is shown in Figure 4.19. The frequency-doubling element was a 6-mm-long monolithic KN ring resonator, having two polished curved surfaces with mirror coatings applied and one polished flat

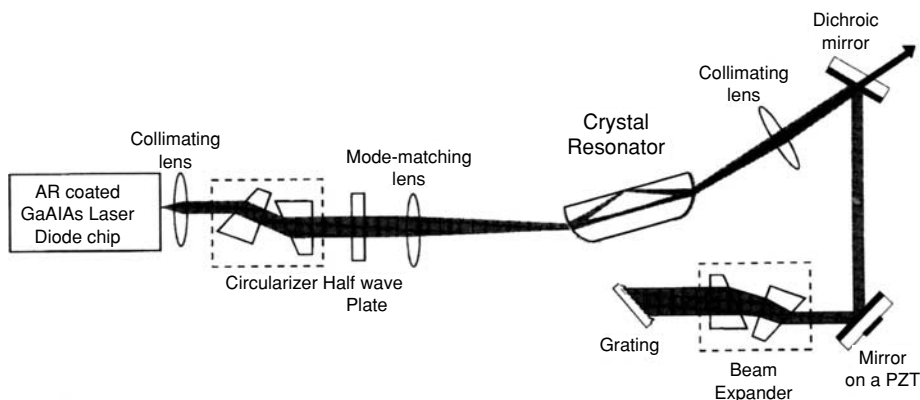


Figure 4.19: Actual experimental arrangement used by Kozlovsky *et al.* (1994) for strong optical locking. (Reprinted with permission from Kozlovsky *et al.* (1994).)

TIR surface. The end of the resonator closest to the diode laser had a radius-of-curvature of 5 mm and a reflectivity of $\sim 92\%$; the other end also had a radius-of-curvature of 5 mm but had a reflectivity of $\sim 99\%$ at 858 nm (the transmission of this coating at 429 nm was $>80\%$). The grating reflected $\sim 65\%$ of the light incident upon it; this reflectivity combined with the transmissivity of other elements in the cavity produced an on-resonance reflectivity as seen by the diode laser of $\sim 3\%$. The spacing between resonant frequencies of the monolithic resonator was ~ 10 GHz and the linewidth of a resonance was ~ 200 MHz. The optical length of the extended laser cavity corresponded to an axial mode spacing of ~ 150 MHz. Dither locking was used to set the extended cavity length so that one of these axial modes coincided with a resonance of the monolithic ring. This device produced 14 mW of 429-nm blue light. For comparison, at the same infrared power level (60 mW), the electronically-locked monolithic ring described earlier (Kozlovsky *et al.*, 1990) produced ~ 20 mW.

Another optical locking approach involving strong feedback is shown in Figure 4.20. In this approach, an anti-reflection-coated laser diode forms the gain element in an extended ring resonator. Inside that ring laser cavity is a second resonator for enhancement of the nonlinear interaction. Optical locking works according to similar principles to those described in connection with Figure 4.18. Wigley *et al.* (1995) used this approach to lock the frequency of the extended-cavity diode laser to a resonance frequency of the cavity containing the nonlinear crystal; however, their goal was not SHG of the diode laser but SFG of the 845-nm diode laser emission with the 1047-nm emission of a diode-pumped Nd:YLF laser (Figure 4.21). They generated 120 mW of blue radiation in a TEM_{00} mode at 467 nm using this approach. The 845-nm laser diode was a tapered amplifier device of the type described in Chapter 3.

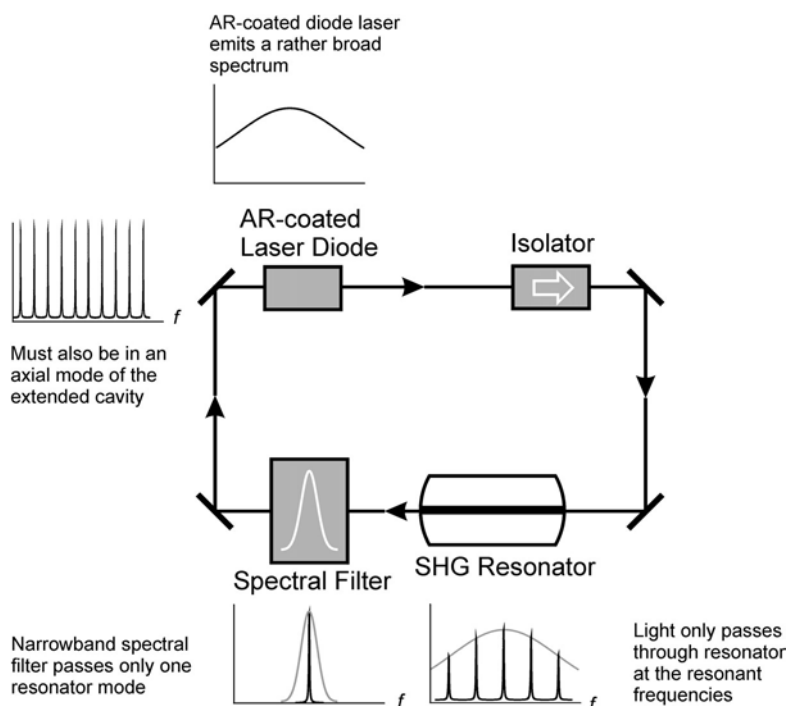


Figure 4.20: Principle behind strong optical locking approach used by Wigley *et al.* (1995).

A new approach to optical locking has been reported by King and Pittaro (1998). Although they did not use it for resonant enhancement of SHG, it deserves mention in this section. We noted earlier that the beam reflected from the resonator is a superposition of a component directly reflected from the input mirror of the cavity and a component that arises from leakage of the circulating field through the input mirror. If the input beam is perfectly mode-matched to the resonator (as described in the next section), these two components have phase fronts with exactly the same radius of curvature, and the cancellation of the beams is uniform over the entire cross-section of the reflected beam. However, if the input beam is not perfectly mode-matched to the resonator, the cancellation is not uniform over the cross-section, and a situation can arise in which the on-axis intensity is higher on-resonance than off-resonance. Thus, if the reflected beam is subjected to appropriate spatial filtering, a signal can be derived that has the required characteristics for optically locking the laser to the resonator.

4.2.5 Mode matching

We have seen in preceding chapters that SHG using gaussian beams is most efficient when the infrared beam is focused to the Boyd–Kleinman optimum. Thus, we would

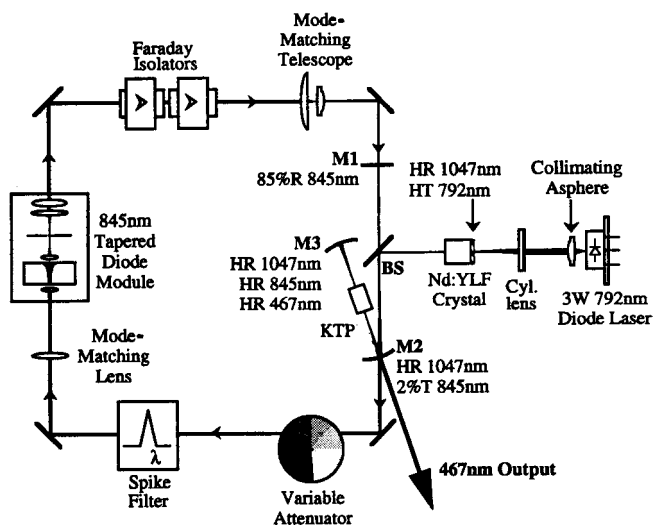


Figure 4.21: Actual experimental configuration used by Wigley and coworkers for strong optical locking. (Reprinted with permission from Wigley *et al.* (1995).)

like to design our resonator so that the beam it confines is focused to such a degree in the nonlinear crystal. If we begin by assuming that the infrared beam is optimally focused, we can see that a resonator constructed of mirrors which match the radius of curvature of the beam at a given point will precisely reflect the beam back on itself, so that a confined gaussian beam with the desired shape will be a transverse mode of the resonator (Figure 4.22).

In order to efficiently excite such a mode, the input beam must be focused so that it matches the spatial variation of this confined mode when it enters the resonator. Such “mode matching” generally requires the design of an optical system that transforms the beam emitted by the infrared laser into a beam having the same spatial variation as the resonator mode (Figure 4.23). If the infrared laser emits a TEM_{00} gaussian mode, this transformation can be accomplished with a simple lens or two; if the infrared laser emits a more complex beam, additional beam-shaping optics may be necessary.

It is beyond the scope of this chapter to present a full discussion of resonator analysis and design, and it is a subject that may be already familiar to many readers. For those unfamiliar with the topic, a number of good introductions are available (for example, Siegman (1986)) and discussions derive from the classic paper by Kogelnik and Li (1966). Two tools introduced in such texts that are useful for mode-matching problems are the “ $ABCD$ matrix” and the “circle diagram”. $ABCD$ matrices provide a mathematical formalism for analyzing gaussian beam propagation through various optical elements. Circle diagrams provide a graphical technique for determining the stability of an optical resonator and locating the

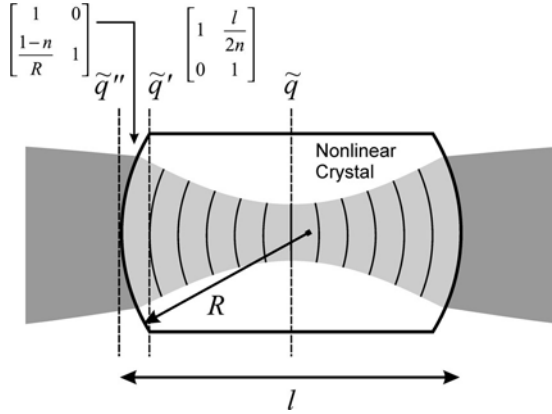


Figure 4.22: Gaussian beam confined to a monolithic resonator.

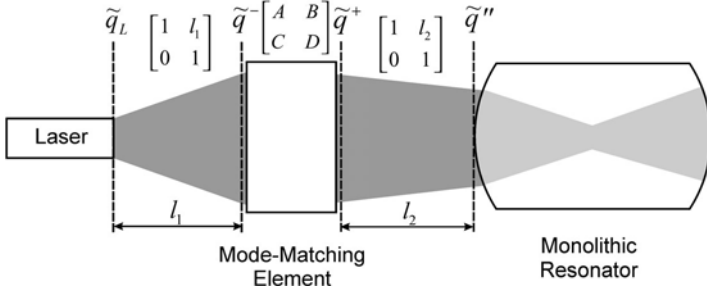


Figure 4.23: Mode matching between the infrared laser and the resonator.

waist of the confined beam. In the specific example that follows, we will use $ABCD$ matrices. Readers interested in the use of circle diagrams are referred to Laures (1967).

We will now consider an example that illustrates some of the physical considerations and mathematical manipulations required for the design of resonators for enhancement of SHG and SFG. Suppose that we wish to form a monolithic standing-wave resonator from a piece of LN with length $l = 10$ mm. In order for the gaussian beam to be optimally focused in the LN, it must have a confocal parameter $b = l/2.84 = 3.52$ mm, and thus, a waist $w_0 = \sqrt{\lambda_1 b / 2\pi n_1} = 16.8 \mu\text{m}$, where $\lambda_1 = 1.064 \mu\text{m}$ and $n_1 = 2.23$. Using the formula given in Chapter 2 for the variation of the radius of curvature of a gaussian beam with distance, we find that at the end of the LN crystal, we have:

$$R(z) = z + \frac{z_r^2}{z} \quad (4.8)$$

$$R(l/2) = \frac{l}{2} + \frac{b^2}{2l} = 5 \text{ mm} + \frac{(3.52 \text{ mm})^2}{2 \cdot 10 \text{ mm}} = 5.6 \text{ mm} \quad (4.9)$$

In order to precisely retroreflect this gaussian beam, and thus trap it in the resonator, the ends of the LN crystal should be polished so that they also have a radius of curvature of 5.6 mm. Using the formula for the width of the gaussian beam as a function of distance from the waist, we find that at the end of the crystal, the spot size is:

$$w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_r}\right)^2} \quad (4.10)$$

$$w(l/2) = w_0 \sqrt{1 + \left(\frac{l}{b}\right)^2} = 16.4 \text{ } \mu\text{m} \sqrt{1 + \left(\frac{10 \text{ mm}}{3.52 \text{ mm}}\right)^2} = 49.4 \text{ } \mu\text{m} \quad (4.11)$$

We will now perform the same calculation using *ABCD* matrices, in order to illustrate their use. The *ABCD* matrices are used to describe the transformation of a gaussian beam as it passes through various optical elements (Figure 4.22). The formalism employs a complex beam parameter $\tilde{q}$ given by:

$$\frac{1}{\tilde{q}} = \frac{1}{R} - j \frac{\lambda}{\pi w^2 n} \quad (4.12)$$

where R is the radius of curvature and w is the spot size at a particular location. The transformation of this gaussian beam by propagation through an optical element produces a different value of this parameter, $\tilde{q}'$, given by:

$$\frac{\tilde{q}'}{n'} = \frac{A \left(\frac{\tilde{q}}{n} \right) + B}{C \left(\frac{\tilde{q}}{n} \right) + D} \quad (4.13)$$

where n is the refractive index at the location described by $\tilde{q}$ and n' is the refractive index at the location described by $\tilde{q}'$.

We can use the *ABCD* formalism to calculate the spot size and radius of curvature at the end of the LN crystal, just as we did above using the standard formulas. We will use $\tilde{q}$ to describe the gaussian beam at the center of the nonlinear crystal and $\tilde{q}'$ to describe the gaussian beam at the end of the nonlinear crystal. The *ABCD* matrix describing propagation over a distance $l/2$ through a uniform medium of refractive index n is:

$$\begin{bmatrix} A & B \\ C & D \end{bmatrix} = \begin{bmatrix} 1 & \frac{l}{2n} \\ 0 & 1 \end{bmatrix} \quad (4.14)$$

thus:

$$\frac{\tilde{q}'}{n} = \frac{\left(\frac{\tilde{q}}{n}\right) + \frac{l}{2n}}{1} \quad (4.15)$$

or

$$\tilde{q}' = \tilde{q} + \frac{l}{2} \quad (4.16)$$

In the center of the crystal, we have $R = \infty$ and $w = w_0$, so that $\tilde{q} = j\pi w_0^2 n / \lambda = jb/2$. We can rewrite Equation (4.15) as:

$$\frac{1}{\tilde{q}'} = \frac{1}{R'} - j \frac{\lambda}{\pi w'^2 n} = \frac{1}{\tilde{q} + \frac{l}{2}} = \frac{1}{j \frac{b}{2} + \frac{l}{2}} = \frac{-j \frac{b}{2} + \frac{l}{2}}{\left(\frac{l}{2}\right)^2 + \left(\frac{b}{2}\right)^2} \quad (4.17)$$

Thus,

$$R' = \frac{\left(\frac{l}{2}\right)^2 + \left(\frac{b}{2}\right)^2}{\frac{l}{2}} = \frac{l}{2} + \frac{b^2}{2l} \quad (4.18)$$

as in Equation (4.8). Similarly, by equating the imaginary parts of Equation (4.17) we can solve for w' to find:

$$-j \frac{\lambda}{\pi w'^2 n} = -j \frac{\frac{b}{2}}{\left(\frac{l}{2}\right)^2 + \left(\frac{b}{2}\right)^2} = -j \frac{2b}{b^2 + l^2} \quad (4.19)$$

$$w'^2 = \frac{\lambda}{\pi n} \frac{b^2 + l^2}{2b} = \frac{\lambda}{2\pi n} b \left[1 + \left(\frac{l}{b}\right)^2 \right] \quad (4.20)$$

or

$$w' = w_0 \sqrt{1 + \left(\frac{l}{b}\right)^2} \quad (4.21)$$

exactly as in Equation (4.10).

Suppose now that the endface of the LN crystal has been polished to have a radius of curvature of 5.6 mm. How do the characteristics of the gaussian beam change as it passes through this curved interface between the LN and air? This question also is easily answered using *ABCD* matrices. The matrix describing the action of an

interface between a medium of index n and air with radius of curvature R is:

$$\begin{bmatrix} A & B \\ C & D \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ \frac{1-n}{R} & 1 \end{bmatrix} \quad (4.22)$$

Thus, we determine that the parameter $\tilde{q}''$ just outside the interface is related to the parameter $\tilde{q}'$ just inside the interface by:

$$\frac{\tilde{q}''}{1} = \frac{A\left(\frac{\tilde{q}'}{n}\right) + B}{C\left(\frac{\tilde{q}'}{n}\right) + D} = \frac{\frac{\tilde{q}'}{n}}{\frac{1-n}{R}\left(\frac{\tilde{q}'}{n}\right) + 1} \quad (4.23)$$

which we can rewrite as:

$$\frac{1}{\tilde{q}''} = \frac{1-n}{R} + \frac{n}{\tilde{q}'} = \frac{1-n}{R} + \frac{n}{R'} - j\frac{\lambda}{\pi w'^2} \quad (4.24)$$

Equating the imaginary parts, we have $w'' = w'$ and equating the real parts, we find

$$\frac{1}{R''} = \frac{1-n}{R} + \frac{n}{R'}$$

Thus, the spot size does not change as the beam passes through the curved interface, but the radius of curvature does, due to refraction at the interface.

The last step in the mode-matching problem is illustrated in Figure 4.23. If we are using an infrared laser whose parameter $\tilde{q}_L$ is known at some location, we need to find a suitable optical system which transforms $\tilde{q}_L$ to $\tilde{q}''$. With reference to Figure 4.23, we see that propagation through the space between the laser and the optical element simply increases $\tilde{q}_L$ by l_1 . Similarly, if we propagate $\tilde{q}''$ back to the element, we find that there we have $\tilde{q}'' + l_2$. Thus, for perfect mode matching we must have:

$$\tilde{q}'' + l_2 = \frac{A(\tilde{q}_L + l_1) + B}{C(\tilde{q}_L + l_1) + D} \quad (4.25)$$

Perhaps the simplest optical system we could imagine is a single thin lens. Under what conditions can such an element provide mode matching? First, let us consider the effect of a thin lens on the $\tilde{q}$ parameter of a gaussian beam. If we use $\tilde{q}^-$ to denote the $\tilde{q}$ parameter just to the left of the lens and $\tilde{q}^+$ to denote the $\tilde{q}$ parameter just to the right of the lens (with light propagating from left to right), using the $ABCD$ matrix for a thin lens gives us:

$$\tilde{q}^+ = \frac{\tilde{q}^-}{-\frac{1}{f}\tilde{q}^- + 1} \quad (4.26)$$

or

$$\frac{1}{\tilde{q}^+} = -\frac{1}{f} + \frac{1}{\tilde{q}^-} \quad (4.27)$$

Comparing real and imaginary parts, we observe that $R^+ = R^- - f$ and $w^+ = w^-$; that is, the lens does not alter the spot size, but it does change the radius of curvature by an amount equal to the focal length. Thus, if we can choose l_1 and l_2 so that

$$\text{Im}\left(\frac{1}{\tilde{q}'' + l_2}\right) = \text{Im}\left(\frac{1}{\tilde{q}_L + l_1}\right)$$

the spot sizes will match at the lens. We must then choose f such that

$$\text{Re}\left(\frac{1}{\tilde{q}'' + l_2}\right) = \text{Re}\left(\frac{1}{\tilde{q}_L + l_1}\right) - \frac{1}{f}$$

in order to make the radii of curvature match. Whether the lengths l_1 and l_2 and the focal length f are practically acceptable will determine whether mode matching can be achieved with a simple lens or whether a more complicated optical system is needed. Even if a more complex arrangement is needed, the same basic *ABCD* analysis can be used to design it.

4.3 OTHER CONSIDERATIONS

4.3.1 Temperature locking

In Section 4.2.4, we examined techniques for matching the frequency of the laser to a resonant frequency of the enhancement cavity. Once the laser is stably frequency-locked to the resonator, the problem still remains of ensuring that this frequency falls within the phasematching bandwidth of the nonlinear process. This problem is usually solved by adjusting the temperature of the nonlinear crystal in order to tune the phasematching wavelength. In a practical system, it is desirable to have some way of automatically adjusting the temperature so that phasematching is optimized. As with frequency locking, a key question is how to generate an error signal that indicates both the degree and direction of departure from phasematching.

A scheme described by Bethune and Kozlovsky (1992) has been used to optimize the temperature of a monolithic KN resonator used for SHG of 860 nm light. Normally, propagation would be along the crystallographic a -axis, in order to obtain the shortest possible wavelength and reap the benefits of noncritical phasematching. However, in their approach, propagation is at an angle of a few degrees with respect to the a -axis, which requires only a slight increase in the phasematching wavelength. When the temperature of the crystal increases, n_c changes by a larger amount than n_{a-b} ; thus, as shown in Figure 4.24, the phasematching direction for the fixed wavelength of operation lies slightly closer to the a -axis. In contrast, when

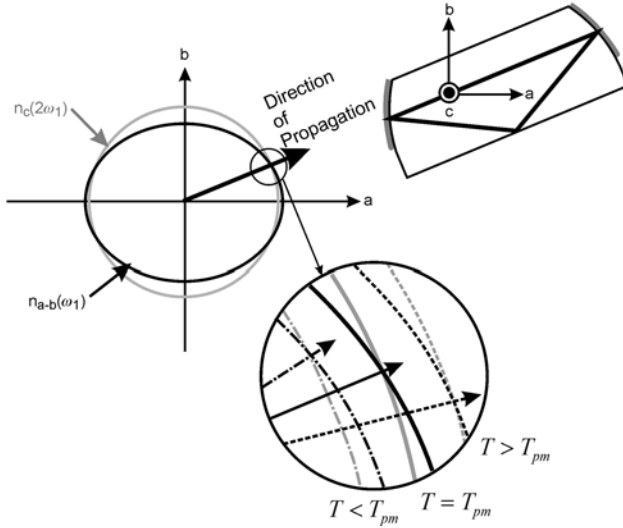


Figure 4.24: Principle behind the temperature locking scheme of Bethune and Kozlovsky (1992).

the temperature of the crystal decreases, the optimum phasematching direction moves slightly farther away from the a -axis. The direction of the focused infrared gaussian beam is fixed, but it contains a range of angular-spectrum components; thus, depending on whether the temperature is above or below that required for optimum phasematching, one side or the other of the infrared gaussian beam will experience more efficient conversion to the second harmonic. The result is a slight deviation of the angle of the blue output beam with temperature. If the position of the blue output beam is monitored with a position-sensitive detector (such as a split photodiode), an error signal can be generated which can then be used in a servo loop to lock the temperature to the optimum value.

4.3.2 Modulation

In many applications, modulation of the blue output beam is essential. Schemes which permit direct modulation of the blue output of a resonant doubler, rather than requiring a separate modulator, are therefore of interest. However, the nature of resonant doubling places some constraints on how this modulation can be produced. In contrast to single-pass SHG (Chapter 3), it is not usually possible to simply modulate the current of the diode laser in order to produce a corresponding modulation of the intensity of the blue output, since such modulation might disrupt whatever technique is used for frequency locking.

However, a few techniques for modulating the blue output have been demonstrated which require essentially only those components already being used for cw

operation. For example, Senoh *et al.* (1992) applied an electric field to the KN crystal in order to detune the phasematching condition. Since r_{33} is about 50 times larger than r_{23} in KN, an electric field applied along the crystallographic c -axis changes the refractive index of the second harmonic (n_c) by a correspondingly greater amount than it changes the refractive index of the fundamental (n_b). Using this approach, they were able to produce nearly 100% intensity modulation, although only at frequencies $\lesssim 1$ kHz.

Kozlovsky and Lenth (1994) demonstrated that they could make pulses of blue light as short as 10 ns, using an approach that is compatible with the frequency-modulated locking technique described in Section 4.2.4. For frequency-modulated locking, the diode laser is weakly frequency-modulated, so that only one significant upper and lower frequency-modulated sideband is produced, at a spacing in frequency that exceeds the width of the cavity resonance. At this weak level of frequency modulation, most of the energy emitted by the laser remains in the carrier frequency. However, if the depth of the frequency modulation is increased, more energy can be taken out of the carrier and placed in the sidebands; in fact, at a certain depth of modulation, the energy at the carrier frequency can be reduced to zero. Since only the carrier frequency is coupled into the cavity, reducing the carrier frequency decreases the blue output power. As long as a small amount of energy remains at the carrier frequency, stable locking can be maintained. They found that they could produce pulses of blue light as short as 10 ns, with an on/off ratio of 10:1 (4 mW off, 40 mW on) by switching the rf modulation applied to the diode for frequency-modulated sideband generation (Figure 4.25).

4.3.3 Bireflection in monolithic ring resonators

In monolithic resonators, the fact that the ring path is contained entirely within the nonlinear crystal introduces some considerations that do not arise in resonators constructed from discrete components. One such consideration is the phenomenon of bireflection, illustrated in Figure 4.26, which shows what happens to light incident upon a monolithic mirror such as M_2 in Figure 4.2(a). As we saw in Chapter 2, most common nonlinear materials are optically anisotropic, so that the refractive index depends on both polarization and direction of travel. Figure 4.26 shows the variation of refractive index with direction of travel for light polarized either parallel or perpendicular to the plane of the ring. Here, the refractive index for light polarized perpendicular to the plane of the ring does not vary with direction of propagation, but the refractive index for light polarized parallel to the plane of the ring does. The result of this behavior is that angle of reflection from the mirror also depends on polarization state. The component of propagation vector tangential to the reflecting surface must be the same for both the incident and

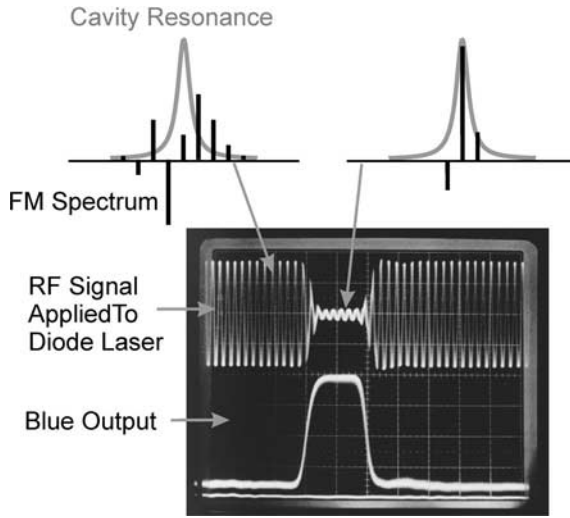


Figure 4.25: Modulation of blue output by varying modulation depth in the frequency-modulated locking scheme. (Reprinted with permission from Kozlovsky and Lenth (1994).)

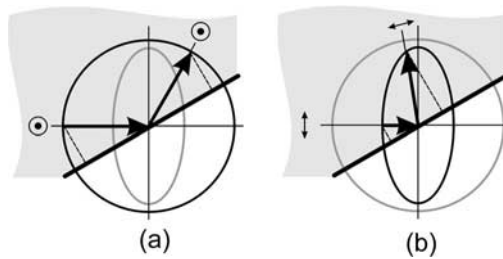


Figure 4.26: The phenomenon of bireflection in an anisotropic medium causes the angle of reflection to depend on polarization. (a) shows light polarized perpendicular to the ring plane incident at an angle on a monolithic reflector, (b) shows light polarized parallel to the ring plane incident upon the reflector.

reflected waves. When the index curve is symmetric about the normal to the reflector, this continuity of the tangential component of momentum produces the familiar result that the angle of incidence equals the angle of reflection. When the refractive index is independent of direction as in Figure 4.26(a), this situation occurs for any angle between the reflector and the symmetry axes. However, in the case shown in Figure 4.26(b), this symmetry does not exist, and the angles of reflection and incidence are different. In this example, a ring design which closes on itself for the perpendicularly-polarized wave would not close for the parallelly-polarized wave. The intrinsically-polarizing nature of monolithic ring resonators can be exploited for implementation of Hänsch–Couillard locking (Kozlovsky *et al.*, 1988).

While bireflection can be circumvented in resonators designed for Type I SHG, it is a serious impediment when using monolithic ring resonators for Type II SHG

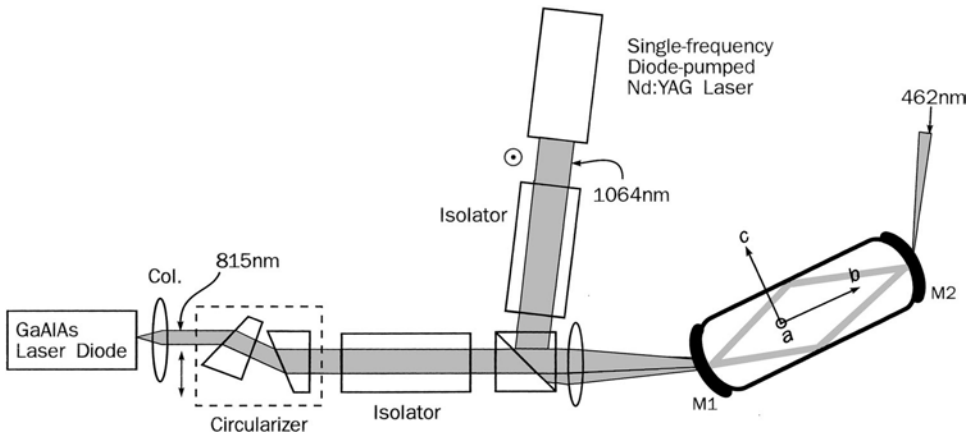


Figure 4.27: Monolithic ring resonator used by Kozlovsky and Risk for generation of blue light by doubly-resonant sum-frequency mixing. (Reprinted with permission from Risk and Kozlovsky (1992).)

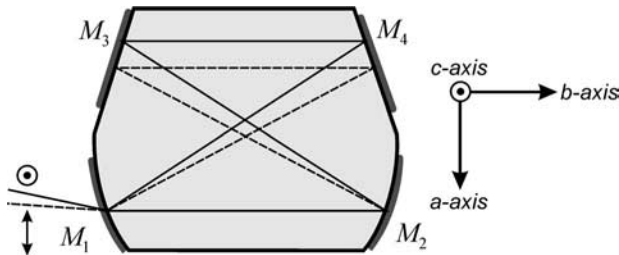


Figure 4.28: Improved monolithic resonator for sum-frequency mixing.

or SFG, in which two orthogonal polarizations must both be resonated. Risk and Kozlovsky (1992) encountered this problem when trying to use a ring resonator to increase the efficiency for mixing of 809-nm and 1064-nm light in KTP. Their solution was a ring resonator having a diamond-shaped path, as shown in Figure 4.27. In this design, bireflection is eliminated by arranging all the reflecting surfaces to be parallel to principal axes of the crystal; thus, a symmetric situation is established for the in-plane polarization (imagine Figure 4.26(b), with the reflector parallel to a principal axis). In this case, the angle of reflection equals the angle of incidence for that polarization also, and the ring closes for both polarizations. However, a disadvantage of the diamond-shaped ring design is that all four legs of the path are phasematched, and sum-frequency is generated along each of them.

An alternative design, shown in Figure 4.28, allows a bow-tie resonator configuration to be implemented in a monolithic ring, which does not eliminate bireflection, but exploits it advantageously (Kozlovsky and Risk, 1995). In this design, mirrors M_1 and M_4 are parallel to each other, as are M_2 and M_3 . When the path of each polarization is traced around the resonator, taking bireflection into account, it is found that the symmetry of the design is such that both polarizations follow a

Table 4.1. Summary of experiments involving resonator-enhanced SHG

Reference	λ_3 (nm)	Blue-green power	Conv. (eff.%)	Material	Resonator configuration	Locking technique	Comments
Kozlovsky <i>et al.</i> (1987)	532	2 mW	13	MgO:LN	M. SW	Dither	SFG of 795 nm & 1064 nm
Kozlovsky <i>et al.</i> (1988)	532	29.7 mW	56	MgO:LN	M. Ring (Tri)	Dither, HC, PDH	
Dixon <i>et al.</i> (1989)	432	0.215 mW	1.7	KN	D. SW	Optical	
Goldberg and Chun (1989)	421	24 mW	45	KN	M. SW	Scanned	
Hemmerlich <i>et al.</i> (1990)	421	6.7 mW	0.57	KN	D. Ring (BT)	Optical	
Goldberg <i>et al.</i> (1990)	455	154 mW	3.2	KN	M. SW	Scanned	Ti:S laser source
Kozlovsky <i>et al.</i> (1990)	428	41	39	KN	M. Ring (Tri)	PDH	
Gerstenberger <i>et al.</i> (1991)	532	200 mW	65	MgO:LN	M. Ring (Tri)	Dither, PDH	
Polzik and Kimble (1991)	430	650 mW	48	KN	D. Ring (Rect)	PDH	
Yang <i>et al.</i> (1991)	532	6.5 W	36	LBO	D. Ring (BT)	PDH	
Jundt <i>et al.</i> (1991)	532	1.7 W	40	PPLN	D. Ring (BT)	PDH	SFG, pump-resonant Nd:YAG
Kean and Dixon (1992)	459	1.2 mW	2.4	KTP	D. SW	Dither	
Senoh <i>et al.</i> (1992)	431	6.6 mW	7.3	KN	D. Ring (Tri)	Optical	
Risk and Kozlovsky (1992)	459	4 mW	6.3	KTP	M. SW	PDH, scanning	
Risk and Kozlovsky (1992)	462	2 mW	2.1	KTP	M. Ring (Dia)	PDH, scanning	
Zimmerman <i>et al.</i> (1992)	486	1.2 mW	4.8	KN	D. Ring (BT)	Optical	Doubly-resonant SFG
Reid (1993)	410	0.36 mW	3.1	KLN	D. SW	None	
Tamm (1993)	370	18 μ W	0.2	LiIO ₃	D. Ring (BT)	Optical	

Fiedler <i>et al.</i> (1993)	532	5 mW	50	MgO:LN	M. Ring (Dia)	PDH	Doubly-resonant SHG
Hemmerlich <i>et al.</i> (1994)	430	22 mW	22	KN	M. Ring (Tri)	Optical	
Alfrey <i>et al.</i> (1994)	532	4.4 W	58.7	LBO	D. Ring (BT)	PDH	
Hayasaki <i>et al.</i> (1994)	397	1.8 mW	1.8	LiIO ₃	D. Ring (BT)	Optical	
Kozlovsky <i>et al.</i> (1994)	429	14 mW	14	KN	M. Ring (Tri)	Optical	
Paschotta <i>et al.</i> (1994)	532	82 mW	82	MgO:LN	M. SW	Dither	First stage of quadrupler
Zimmerman <i>et al.</i> (1994)	486	156 mW	36	KN	D. Ring (BT)	Optical	Second stage of quadrupler
Zimmerman <i>et al.</i> (1994)	243	2.1 mW	1.3	BBO	D. Ring (BT)	HC	Signal-resonant SFG
Wigley <i>et al.</i> (1995)	467	120 mW		KTP	D. SW (F)	Optical	Doubly-resonant SFG
Kaneda and Kubota (1995)	355	186 mW	19.6	BBO	D. Ring (BT)	PDH	
de Angelis <i>et al.</i> (1996)	397	0.85 mW	0.65	LBO	D. Ring (BT)		
Lodahl <i>et al.</i> (1997)	430	8 mW	40	KN	D. Ring (BT)	PDH	
Beier <i>et al.</i> (1997)	403	98 mW	25	LBO	D. Ring (BT)	Optical, HC	
Beier <i>et al.</i> (1997)	403	98 mW	24	BBO	D. Ring (BT)	Optical, HC	
Aire <i>et al.</i> (1998)	532	123.5 mW	55	PPKTP	D. Ring (BT)	PDH	
Woll <i>et al.</i> (1999)	465	1 W	59	LBO	D. Ring (BT)	HC	

Abbreviations

Construction: D. – Discrete Components, M. – Monolithic; Configuration: SW – Standing Wave, Ring – Ring; Ring Path: Tri – Triangular, BT – Bow Tie, Dia – Diamond; Locking: HC – Hänsch–Couillaud, PDH – Pound–Drever–Hall.

bow-tie shaped path, but the path is slightly different for the two orthogonal polarizations. The two paths perfectly coincide only along the segment of the path between mirrors M_1 and M_2 . Propagation is along the phasematching direction (crystallographic b -axis) along two segments (M_1 – M_2 and M_3 – M_4), but the spatial offset of the beams along the M_3 – M_4 segment inhibits frequency conversion. Thus, blue light is generated only along the M_1 – M_2 segment of the path and in only one direction.

4.4 SUMMARY

In this chapter, we have investigated the use of a resonator for enhancement of SHG and SFG. We saw that it is necessary to ensure a number of “matchings” in order for resonator enhancement to work well: impedance matching, mode matching, and frequency matching. We saw that there were primarily two approaches to frequency matching and locking: electronic locking and optical locking. We saw that there were two main resonator configurations – standing-wave and ring – and that these could be constructed either monolithically or from discrete components.

To conclude this chapter, we present Table 4.1, which lists demonstrations of various schemes for resonator enhancement for blue-green light generation. In the next chapter, we consider the enhancement that is possible by placing the nonlinear crystal within the cavity of a solid-state laser, rather than in its own separate resonant cavity.

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5

Intracavity SHG and SFG

5.1 INTRODUCTION

In the last chapter, we examined resonator-enhanced blue-green light generation, in which a nonlinear crystal is placed inside an optical resonator so that the high circulating intensity increases the efficiency of SHG or SFG. We considered some implementations of this approach in which light from a diode-pumped solid-state laser is coupled into such a resonator, and we saw that it becomes necessary to lock the laser frequency to a resonant frequency of the enhancement cavity. Looking at such a system, we might well ask, “Since the solid-state laser itself consists of a cavity which is resonant at the infrared wavelength, why not place the nonlinear crystal inside *that* cavity instead of inside a separate one?” Inclusion of the nonlinear crystal within the resonator of an infrared laser is the basic idea behind intracavity SHG and SFG, which is the subject of this chapter.

Although generation of green light by intracavity frequency doubling of neodymium lasers has been pursued since the mid-1960s (Smith *et al.*, 1965, Geusic *et al.*, 1968), the current wave of interest in this field was ignited in the mid-1980s by the development of high-power, high-brightness diode lasers capable of efficiently pumping solid-state lasers and the demonstration that milliwatt levels of green light could be generated by placing a nonlinear crystal within the cavity of a diode-pumped Nd^{3+} laser (Baer and Keirstead, 1985, Fan *et al.*, 1986). These early demonstrations of compact green lasers based on intracavity SHG spurred efforts to invent and improve laser host and nonlinear materials, to design new resonator configurations that improved efficiency or gained some other advantage (such as very compact size), and to extend the wavelength range that could be generated using this approach to colors other than green.

In this chapter, we will begin with a short consideration of the theory of intracavity SHG. We will then discuss the “green problem”, a term which refers to large amplitude fluctuations observed in the second-harmonic output generated by

intracavity-doubled lasers. In the course of discussing the “green problem” and the various solutions to it that have been investigated, we will review most of the more recent work involving generation of green light using intracavity SHG of diode-pumped Nd lasers. We will then consider some other, related approaches to generation of blue-green light: intracavity SHG of 946-nm Nd:YAG lasers, intracavity SHG of Cr:LiSAF lasers, and intracavity sum-frequency mixing. Finally, we will consider the “self-doubling” approach, in which the same material provides both laser gain and frequency-doubling.

5.2 THEORY OF INTRACAVITY SHG

Before trying to analyze the case of a laser resonator with a nonlinear crystal placed inside, let us first analyze the resonator *without* the nonlinear crystal, in order to gain some familiarity with the procedure, and in order to derive a result which will provide a useful insight into the potential of intracavity SHG. We will consider a solid-state laser with a “semimonolithic” design – that is, one in which mirror M_1 is deposited directly on the polished face of the solid-state material and mirror M_2 is a discrete element (Figure 5.1). If we were to trace the intensity of the infrared light through one round trip in the cavity, we would find the result shown in Figure 5.1. In this simplified conceptual example, we consider all the loss in the cavity as occurring in the region between the gain medium and mirror M_2 . Thus, we initially have some intensity at the output of the gain medium, and this intensity decreases slightly as the wave propagates toward mirror M_2 . When it reaches M_2 , the intensity may decrease abruptly because some of the light is transmitted out of the resonator. The intensity decreases again due to loss on the way back to the gain medium. When it reaches the gain medium, the wave is amplified, and the intensity increases as shown. When the wave encounters mirror M_1 , the intensity will drop abruptly

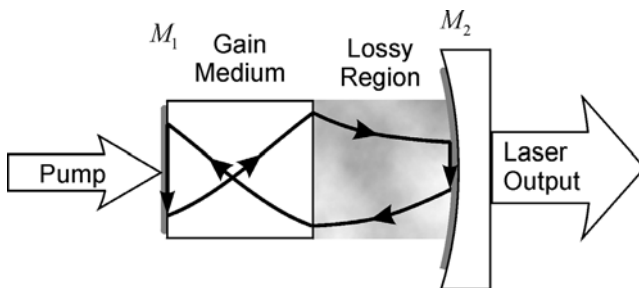


Figure 5.1: Configuration used for analysis of intracavity SHG using a diode-pumped solid-state laser (without a nonlinear crystal). Lines with arrows indicate the evolution of intensity in forward and backward propagation directions.

again, since some portion is transmitted through the mirror. The remaining portion is reflected and undergoes further amplification. In order for the internal field to be self-consistent, the intensity at the output of the gain medium must match what we started out with. Stated differently, in order to maintain a steady-state field inside the laser resonator, we must have the condition that the gain experienced in a round trip is equal to the losses experienced in a round trip.

If we assume that the mirror reflectivities are large so that very little light is transmitted out of the cavity at its ends, and the other intracavity losses are low, then the intensity will not vary much with location in the cavity, and we can consider the behavior of the laser in terms of a constant circulating intensity I_{circ} . We can then write the self-consistency condition as:

$$I_{circ} = I_{circ} R_1 R_2 e^{2gl_1} e^{-2\alpha l_2} \quad (5.1)$$

or

$$1 = R_1 R_2 e^{2gl_1} e^{-2\alpha l_2} \quad (5.2)$$

where g is the gain coefficient, l_1 is the length of the gain medium, α is an absorption coefficient accounting for loss, and l_2 is the length of the lossy section of our resonator. The gain provided by a homogeneously-broadened laser material is intensity-dependent, and can be written as:

$$g(I) = \frac{g_0}{1 + \frac{I}{I_{sat}}} \quad (5.3)$$

where g_0 is the unsaturated gain, and I_{sat} is the saturation intensity. The balance between gain and loss expressed in Equation (5.2) is maintained because as the circulating intensity builds up, the gain is “saturated down” until it matches the loss. In our example, the circulating intensity is present twice in the gain medium, so we must write:

$$g(I_{circ}) = \frac{g_0}{1 + \frac{2I_{circ}}{I_{sat}}} \quad (5.4)$$

If we rearrange Equation (5.2) and take the logarithm of both sides, we obtain:

$$\frac{2g_0 l_1}{1 + \frac{2I_{circ}}{I_{sat}}} = 2\alpha l_2 + \ln \left(\frac{1}{R_1 R_2} \right) \quad (5.5)$$

We can define parameters δ_1 and δ_2 corresponding to M_1 and M_2 , such that $R_1 = e^{-\delta_1}$ and $R_2 = e^{-\delta_2}$. Then we have:

$$\frac{2g_0l_1}{1 + \frac{2I_{circ}}{I_{sat}}} = 2\alpha l_2 + \delta_1 + \delta_2 \quad (5.6)$$

When the reflectivity of the mirrors is large, $R_1 = e^{-\delta_1} \approx 1 - \delta_1 \approx 1 - T_1$, so that $\delta_1 \approx T_1$, and similarly for R_2 . We can also write the absorption factor in terms of a loss L such that $1 - L = e^{-2\alpha l_2}$, so that for low loss, we have $L \approx 2\alpha l_2$.

We can solve Equation (5.6) for I_{circ} to obtain:

$$I_{circ} = \left(\frac{2g_0l_1}{L + T_1 + T_2} - 1 \right) \frac{I_{sat}}{2} \quad (5.7)$$

The total power emitted by the laser is that fraction of circulating intensity transmitted through the mirrors. For convenience, and since it is often the case in practice, let us take $T_1 \approx 0$. Then, the output power is just:

$$I_{out} = T_2 I_{circ} = T_2 \left(\frac{2g_0l_1}{L + T_2} - 1 \right) \frac{I_{sat}}{2} \quad (5.8)$$

We see that if $T_2 \rightarrow 0$, then $I_{out} \rightarrow 0$, because no light is transmitted out of the resonator. Similarly, as $T_2 \rightarrow 2g_0l_1 - L$, $I_{out} \rightarrow 0$, because so much light is being transmitted out of the resonator that even the unsaturated gain is insufficient to compensate for the round-trip loss. Therefore, somewhere in between those two values, there must be an optimum value of T_2 that maximizes the output power. By differentiating Equation (5.8), we can determine that optimum value to be:

$$T_{2,opt} = \sqrt{2g_0l_1L} - L \quad (5.9)$$

and the optimum value of output power is:

$$I_{out,opt} = (\sqrt{2g_0l_1} - \sqrt{L})^2 \frac{I_{sat}}{2} \quad (5.10)$$

The relevance of this result for intracavity SHG will be seen shortly.

Now suppose that we add a nonlinear crystal to the cavity as shown in Figure 5.2. If we now write the self-consistency condition as in Equation (5.2), we obtain:

$$I_{circ} = I_{circ} T_{SH}^2 R_1 R_2 e^{2g l_1} e^{-2\alpha l_2} \quad (5.11)$$

where, as in Chapter 4, $T_{SH} = 1 - \eta_{SH} I_{circ}$ accounts for the loss of the fundamental caused by conversion to the second harmonic. Since, we expect (unfortunately) $\eta_{SH} I_{circ} \ll 1$, we can define another δ parameter such that $T_{SH} = e^{-\delta_{SH}} \approx 1 - \delta_{SH}$.

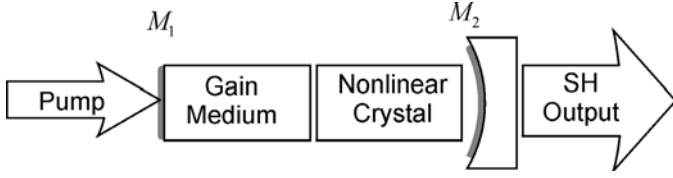


Figure 5.2: Configuration used for analysis of intracavity SHG using a diode-pumped solid-state laser (with a nonlinear crystal).

We can then rewrite Equation (5.11) as:

$$1 = e^{-2\delta_{SH}} e^{-\delta_1} e^{-\delta_2} e^{2g l_1} e^{-2\alpha l_2} \quad (5.12)$$

or, taking the logarithm of both sides:

$$\frac{2g_0 l_1}{1 + \frac{2I_{circ}}{I_{sat}}} = 2\alpha l_2 + \delta_1 + \delta_2 + 2\delta_{SH} \quad (5.13)$$

$$\approx L + T_1 + T_2 + 2\eta_{SH} I_{circ} \quad (5.14)$$

which we can rewrite as:

$$\begin{aligned} 0 = & \left(\frac{L + T_1 + T_2}{2g_0 l_1} \right) - 1 + \frac{I_{circ}}{I_{sat}} \left[\frac{2I_{sat}\eta_{SH}}{2g_0 l_1} + \frac{2(L + T_1 + T_2)}{2g_0 l_1} \right] \\ & + \left(\frac{I_{circ}}{I_{sat}} \right)^2 \frac{4I_{sat}\eta_{SH}}{2g_0 l_1} \end{aligned} \quad (5.15)$$

We can define some normalized parameters to simplify the form of this equation. We define $\gamma = (L + T_1 + T_2)/2g_0 l_1$ as the ratio of total round-trip linear loss to total round-trip unsaturated gain and $\beta = 2\eta_{SH}(I_{sat}/2)/2g_0 l_1$ as the ratio of total round-trip loss due to SHG produced at an intensity equal to half the saturation intensity to total round-trip unsaturated gain. With these definitions, Equation (5.15) becomes the following quadratic in I_{circ}/I_{sat} :

$$4\beta \left(\frac{I_{circ}}{I_{sat}} \right)^2 + 2(\gamma + \beta) \frac{I_{circ}}{I_{sat}} + (\gamma - 1) = 0 \quad (5.16)$$

the solution of which is:

$$\frac{I_{circ}}{I_{sat}} = \frac{-(\gamma + \beta) + \sqrt{(\gamma - \beta)^2 + 4\beta}}{4\beta} \quad (5.17)$$

The generated second-harmonic power is

$$I_{SH} = 2\eta_{SH} I_{circ}^2 = g_0 l_1 I_{sat} \frac{\left[-(\gamma + \beta) + \sqrt{(\gamma - \beta)^2 + 4\beta} \right]^2}{4\beta} \quad (5.18)$$

How does this second-harmonic intensity vary with the normalized parameter β ? If $\beta \rightarrow 0$, that is, if there is no nonlinear conversion, then clearly $I_{SH} \rightarrow 0$. However, $I_{SH} \rightarrow 0$ also if β becomes very large, since the loss due to nonlinear conversion becomes so large that it “pulls down” the circulating intensity. Thus, there must be an optimum value of β that maximizes I_{SH} . By differentiating, Equation (5.18) with respect to β , we find that $\beta_{opt} = \gamma$. Using the definitions for γ and β , we find that:

$$\eta_{SH, opt} = \frac{L + T_1 + T_2}{2I_{sat}} \quad (5.19)$$

that is, the conversion efficiency which optimizes the second-harmonic output power depends on the cavity loss and the saturation intensity, but not on the gain. Under optimum conditions, the maximum second-harmonic output power is thus:

$$I_{SH, max} = g_0 l_1 I_{sat} (1 - \sqrt{\gamma})^2 \quad (5.20)$$

Since we are interested in extracting second-harmonic, not fundamental, power, we should make $T_1 = T_2 \approx 0$. In this case, Equation (5.20) reduces to:

$$I_{SH, max} = \frac{I_{sat}}{2} (\sqrt{2g_0 l_1} - \sqrt{L})^2 \quad (5.21)$$

the right-hand side of which is exactly the same as in Equation (5.10). Thus, we have the following important result: the maximum second-harmonic power that can be generated by intracavity frequency-doubling is the same as the maximum infrared power that can be produced from the same laser with optimized output coupling. Since, for typical solid-state lasers the optimum output coupling is only a few percent, the intracavity conversion efficiency required for optimum SHG is also only a few percent. The other important result that we can obtain from Equation (5.21) is that $I_{SH, max}$ increases as the loss is lowered; thus, the lower we can make the intracavity loss, the better.

In practice, several factors usually prevent us from obtaining the maximum second-harmonic power dictated by theory. As we discussed in Chapter 4, when standing-wave resonators are used, the second-harmonic power may be generated in two oppositely-directed beams, and it may prove difficult to extract the second-harmonic power from both beams in a useful way. The addition of the nonlinear crystal to the cavity introduces additional losses which reduce the amount of second-harmonic power that can be generated. As we will see in the following section, it is sometimes also necessary to insert additional elements into the cavity in order to achieve stable operation, and these also introduce additional losses. In addition, the inherent conversion efficiencies of typical nonlinear crystals and intracavity intensities that can be readily achieved make it difficult to achieve the optimum level of second-harmonic power – most practical intracavity lasers are “undercoupled” relative to the theoretical maximum.

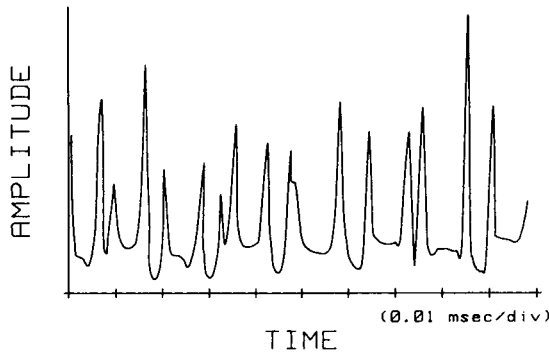


Figure 5.3: Amplitude noise of the 532-nm output from an intracavity-doubled Nd:YAG laser. (Reprinted with permission from Baer (1986).)

5.3 THE “GREEN PROBLEM”

5.3.1 The problem itself

In 1985, Baer and Keirstead reported generating 11 mW of green 532-nm light using KTP for intracavity SHG of a diode-pumped Nd:YAG laser (Baer and Keirstead, 1985). Shortly thereafter, Baer (1986) observed that the green light generated by this technique consisted of a series of randomly occurring large-amplitude spikes superimposed on a small cw level (Figure 5.3), which would render it unsuitable for many applications. Baer further explored the nature of this phenomenon, which became known in the literature as the “green problem”, and explained its origin. He noted that his laser oscillated in several longitudinal modes at 1064 nm. Without the KTP crystal present in the cavity, the amplitude noise present in the infrared emission of the laser was very small. However, when the KTP crystal was included in the cavity, large fluctuations in the amplitude at both 1064 nm and 532 nm occurred. When he placed an étalon within the laser cavity and adjusted it so that only one longitudinal mode could oscillate, the amplitude noise disappeared. When he adjusted the étalon so that two longitudinal modes could oscillate, the amplitude noise became periodic. Using a Fabry–Perot interferometer to frequency filter the output of the laser, he was able to observe the time-dependent behavior of each of these two modes separately. He found that these two modes were turning on and off in a periodic pattern, precisely out of phase with each other (Figure 5.4).

Baer realized that the separation in frequency of the two longitudinal modes was much less than the phasematching bandwidth of the KTP crystal. Thus, not only was SHG of each mode phasematched, but so was sum-frequency mixing of the two modes. We saw in Section 5.2 that the conversion of fundamental to second harmonic can be regarded as an intensity-dependent loss for the fundamental. The same is true in this case, so that if I_1 is the circulating intensity of longitudinal mode 1 and I_2 is the circulating intensity of longitudinal mode 2, mode 1 experiences

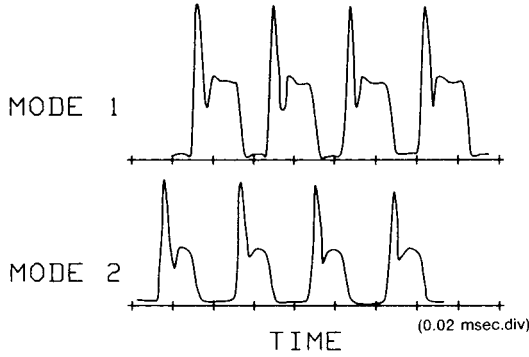


Figure 5.4: Amplitude fluctuations at 1064 nm of each of the longitudinal modes in a two-mode laser with an intracavity SHG crystal. (Reprinted with permission from Baer (1986).)

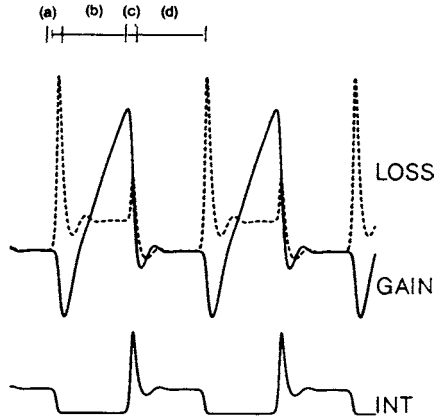


Figure 5.5: Gain and loss of one mode in a two-mode laser with an intracavity doubling crystal as a function of time. (Reprinted with permission from Baer (1986).)

a loss resulting from frequency-doubling of $2\eta_{SH}I_1^2$ and mode 2 experiences a loss of $2\eta_{SH}I_2^2$. However, if both modes are present simultaneously, each mode experiences an additional loss resulting from sum-frequency mixing of the two modes of $4\eta_{SFG}I_1I_2$. Thus, SFG introduces cross-coupling between the two modes such that the behavior of one mode can control the behavior of the other.

Baer explained the switching of the two-mode laser with reference to Figure 5.5. In region (a), mode 2 has just emitted one of the characteristic spikes shown in Figure 5.4. This spike increases the loss seen by mode 1 through SFG. This increased loss causes mode 1 to fall below threshold and turn off. In region (b), with mode 1 off, mode 2 reaches a quasi-steady-state value. The gain for mode 1 begins to build up again, and at some point, exceeds the loss for mode 1 so that it turns on again. In region (c), mode 1 has turned on and emitted a spike, which increases the loss

for mode 2 through sum-frequency generation. Mode 2 falls below threshold and turns off. In region (d), with mode 2 off, mode 1 reaches a quasi-steady-state value. The gain for mode 2 begins to build up again, and at some point, exceeds the loss for mode 2, and it turns on again. The cycle thus repeats. When several modes are present in the cavity, instead of just two, the time-domain behavior is more complex, as shown in Figure 5.4.

5.3.2 Solutions to the “green problem”

Several solutions to the “green problem” (which also, of course, affects lasers of other colors based on intracavity SHG) have been proposed. These solutions can be broadly grouped into three categories: (1) solutions that allow a very large number of modes to oscillate; (2) solutions that allow a few longitudinal modes to oscillate, but somehow preclude sum-frequency generation; and (3) solutions that allow only a single longitudinal mode to oscillate.

5.3.2.1 Extreme multi-mode operation

James *et al.* (1990) analyzed the conditions required for stable operation of a solid-state laser with an intracavity doubling element. Among their findings was the observation that stability should be possible if laser oscillation involves a very large number of modes. Physically, the reason is that if the infrared power is shared by hundreds or thousands of longitudinal modes all weakly coupled together through the nonlinear interaction, no one mode ever achieves sufficient intensity to induce a nonlinear loss large enough to trigger the strong spiking behavior seen when the power is shared by only a few modes.

Magni *et al.* (1993) demonstrated that this approach to solving the green problem works. They used LBO for intracavity SHG of a lamp-pumped Nd:YLF laser with a cavity length of 1.92 m, which oscillated in ~ 250 longitudinal modes. They were able to generate 13.5 W of stable green light using this configuration. Nighan and Cole (1996) obtained similar results, using a diode-pumped Nd:YVO₄ laser with LBO. Their cavity length was ~ 1 m, and the laser oscillated with ~ 100 longitudinal modes. They achieved >6 W of stable cw green output.

5.3.2.2 Few mode operation

Decoupled eigenmode schemes In earlier chapters, we saw that the requirement that a resonator mode accumulate a round-trip phase shift of $2m\pi$, where m is an integer, leads to a spacing in frequency of the longitudinal modes by $\Delta f = c/2l_{opt}$, where l_{opt} is the optical length of the resonator. We also saw that requiring self-consistency of the *amplitude* of a longitudinal mode after a round trip can be used

to derive equations describing the behavior of both passive and active resonators. What we have not previously stated explicitly is that the *polarization* of a longitudinal mode must also reproduce itself after one round trip in the cavity. Understanding these “eigenstates of polarization” is important, since various approaches for achieving stable blue-green operation depend upon manipulating them. These polarization eigenstates can be formally analyzed by applying the Jones matrix description of polarization transformation to resonant cavities (de Lang, 1964, 1969, Bloom, 1974, Nilsson *et al.*, 1989).

When a standing-wave resonator does not contain any birefringent or polarizing elements, the requirement that the polarization state be reproduced after a round trip is a trivial one – since there is nothing in the cavity which can alter the polarization state, *any* polarization state will reproduce itself after one round trip. However, the situation changes when a birefringent element is placed into a resonator. Longitudinal modes must be linearly polarized along either the fast or slow axes of that element; if they are not, the polarization state will be transformed by the element, and will, in general, not return to the original state after a round trip. Thus, there will be two sets of longitudinal modes, one polarized parallel to the fast axis of the birefringent element, and the other polarized parallel to the slow axis. Because the refractive indices are different for the two axes, the frequency spacing will differ for the two sets of modes.

When two birefringent elements are introduced into the resonator, the situation is even more complicated. Obviously, if the principal axes of the two elements are aligned with each other, the longitudinal modes will again be linearly polarized. However, if the principal axes of one element are at some arbitrary angle with respect to the principal axes of the other, a longitudinal mode will undergo a more complicated polarization evolution during a round trip. This evolution can be analyzed using the 2×2 Jones matrix approach mentioned above. The product of the Jones matrices for each individual element encountered in a round trip gives a matrix describing the round-trip polarization change; the two eigenvectors of this matrix describe the two orthogonal polarization states that reproduce themselves after one round trip. Again, there will in general be two sets of longitudinal modes, one set for each eigenpolarization, and the frequency spacing will be different for each set.

In the laser used by Baer for his “green problem” investigation, the only obvious birefringent element is the KTP crystal, and there are two sets of longitudinal modes linearly polarized along the *e* and *o* principal axes of the KTP crystal, which was angle-cut for Type-II SHG of 1064 nm. However, the existence of two sets of orthogonally-polarized modes is also the ideal arrangement for Type-II SFG. As Baer discovered, it is this SFG that leads to the large amplitude fluctuations known as the “green problem”. Oka and Kubota (1988) suggested that the addition

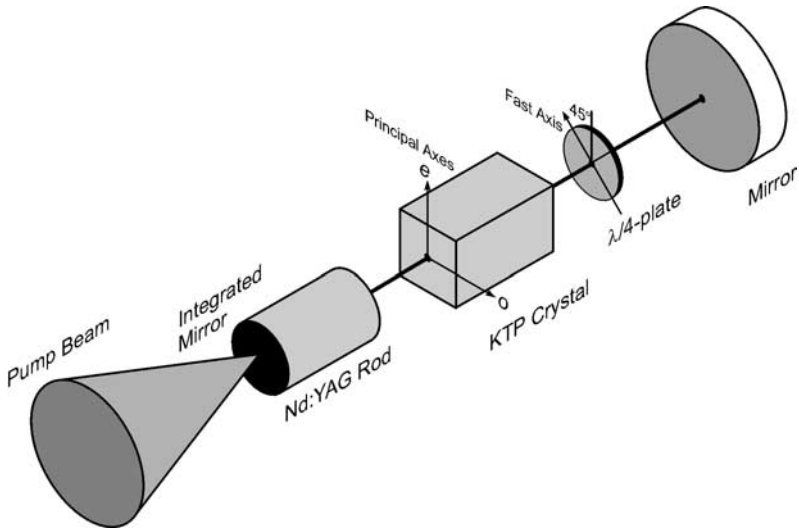


Figure 5.6: Configuration suggested by Oka and Kubota (1988) for solving the green problem.

of a second birefringent element, such as a quarter-wave plate, to the cavity could be used to manipulate the polarization states of the longitudinal modes in such a way that SFG could be suppressed. They used the configuration of Figure 5.6, in which a quarter-wave plate is placed between the KTP crystal and the end mirror, oriented at 45° to the axes of the KTP. Such a configuration produces longitudinal modes that are linearly polarized at $\pm 45^\circ$ at the input to the KTP crystal. Suppose only a $+45^\circ$ mode is present, at frequency ω_1 . Since this component contains both e - and o -components, it can generate the second harmonic at $2\omega_1$ by itself. Similarly, if only a -45° mode is present at frequency ω_2 , it can generate the second harmonic at $2\omega_2$. However, if both modes are present, we find that the sum-frequency contribution created by the mixing of the o -component at ω_1 with the e -component at ω_2 exactly cancels that arising from mixing the e -component at ω_1 with the o -component at ω_2 , so that no net component at $\omega_1 + \omega_2$ is generated. However, although this approach suppresses sum-frequency mixing between longitudinal modes having orthogonal polarizations, it does *not* prevent sum-frequency mixing between longitudinal modes having the same polarization. In principle, spatial hole burning may still allow multiple longitudinal modes of the same polarization to oscillate, and mixing of these modes can cause “green problem” noise (although the mixing efficiency is lower than for mixing of orthogonally-polarized modes).

Oka and Kubota demonstrated that they could achieve green output powers of a few milliwatts, free from “green problem” noise, using this approach. Continued improvement of the system eventually led to generation of 3.5 W of cw, low-noise,

green output power using this technique (Liu *et al.*, 1994). Oka and Kubota (1992) also observed that lasers using this scheme tend to oscillate in two longitudinal modes with equal intensity, one in each eigenpolarization. Oka *et al.* (1995) showed that this condition predominates because it minimizes the overall nonlinear loss. In addition, they demonstrated that their approach is similar to the “twisted mode” technique, in that the modulation of intensity arising from the superposition of these two polarization modes can be made to be small in the gain medium, which greatly reduces spatial hole burning.

James *et al.* (1990) analyzed the configuration of Oka and Kubota in greater detail, examining the behavior of the system from the point-of-view of chaos theory. They also pointed out that this configuration provides stable green output for *any* orientation of the quarter-wave plate when the retardation of the KTP is an odd-integer multiple of $\pi/2$. In subsequent work, James *et al.* (1990a) realized that although Nd:YAG is optically isotropic in principle, in practice, various factors (such as stress) may cause the crystal to acquire a small birefringence. Thus, they extended their previous theory to describe the case where there are two birefringent elements of arbitrary retardation inside the cavity, oriented at an arbitrary angle to each other. This allowed them to consider the case of a cavity like that used by Baer, in which no quarter-wave plate was present, but in which the Nd:YAG crystal may introduce a small additional birefringence. They also considered coupling not only between modes in orthogonal polarizations, but between modes with the same polarization. They found that with any value of birefringence for the Nd:YAG, there is a range of angles for the KTP crystal which produce stable second-harmonic output.

Self-stabilizing schemes Marshall (1997) described a scheme for automatic suppression of the “green problem”. He investigated how the “spiky” time-domain characteristics of the infrared light produced by nonlinear coupling of longitudinal modes change as the length of the laser cavity is changed. He observed that as the cavity length is shortened, the pulses are more closely spaced in time and become shorter, which results in a higher peak power. This higher peak power increases the efficiency of nonlinear conversion, which makes the loss to the fundamental caused by conversion to the second harmonic larger. Thus, the overall loss associated with this “spiky” mode of operation is higher than if the laser were running cw at the same average infrared power. Since the laser will oscillate in the mode of operation with the lowest loss, it will tend to run cw rather than in the pulsed mode. This preference for cw operation is passively enforced – for a cavity of appropriate length, cw oscillation will be automatic. Modelling of such a laser showed that stable cw operation should occur when the cavity length is $\lesssim 20$ mm. Marshall demonstrated this concept in a diode-pumped Nd:YAG laser and achieved 3.1 W of stable cw output.

Tsunekane *et al.* (1997) described another self-stabilizing scheme. They used a cavity that included a Brewster plate and a KTP crystal acting as a birefringent filter, which restricted the number of longitudinal modes in the infrared spectrum to perhaps seven. Assume for a moment that these seven components were equal in amplitude and in-phase with each other. Then, when we perform the convolution required to obtain the spectrum of the green light (as described in Chapter 2), we would expect to find thirteen spectral components with a triangular envelope, the central component being the largest. However, when Tsunekane and colleagues observed the spectrum of the green output from their laser, they found that the central component was missing! The absence of this component suggested that the longitudinal modes *did not* all have the same phase, and that these phase differences led to cancellation of the missing component. Although they did not completely account for how this phase relationship is established, they demonstrated that it could be used to produce low-noise cw green output powers exceeding 6 W, and that this means of stabilization can be robust with respect to pump power fluctuations, temperature fluctuations, turning the laser on and off, and other perturbations. Anthon (1999) interpreted this behavior in terms of “passive FM laser operation.”

5.3.3 Single-mode operation

Short cavities Perhaps the most obvious solution to the “green problem” is to force the laser to oscillate in a single longitudinal mode. One way to do this is to make the frequency separation between adjacent longitudinal modes large in comparison to the gain bandwidth so that only one mode has sufficient gain to oscillate (Figure 5.7). This approach requires a very short cavity, such as that achieved in a “microchip” laser. As originally conceived, microchip lasers consisted of a thin slab of lasing material polished flat and parallel, with dielectric mirrors deposited directly on the polished surfaces (Figure 5.8) (Zayhowski and Mooradian, 1989). Although flat-flat resonators are only marginally stable, a combination of

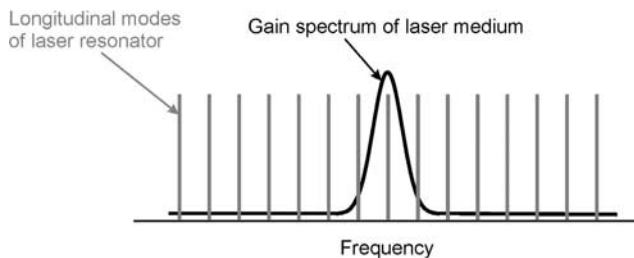


Figure 5.7: Single-mode oscillation can be achieved by making the laser resonator sufficiently short that only one longitudinal mode falls within the bandwidth of the gain spectrum.

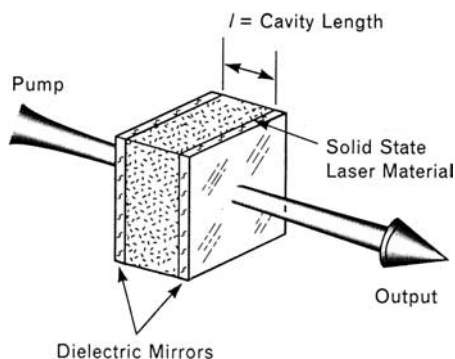


Figure 5.8: Geometry of a microchip laser. (Reprinted with permission from Zayhowski (1990).)

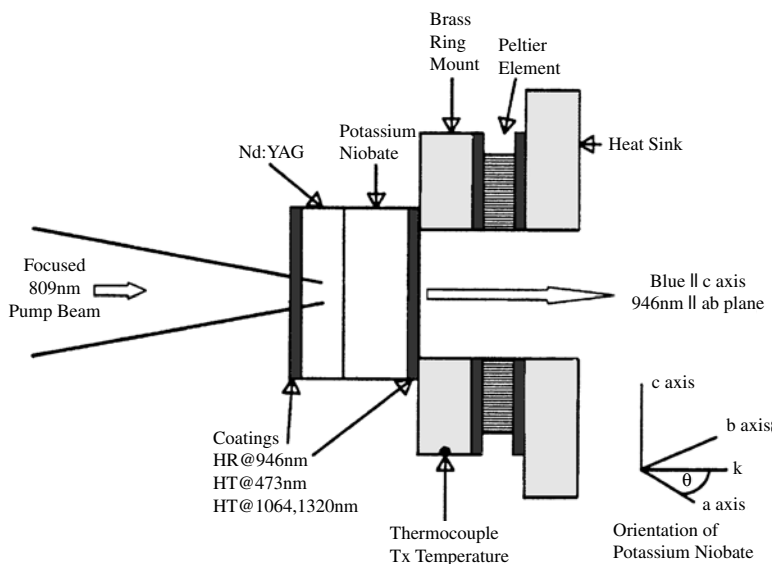


Figure 5.9: Composite-cavity microchip laser used by Matthews and coworkers. (Reprinted with permission from Matthews *et al.* (1996).)

thermal lensing (resulting from heating of the laser medium due to absorption of the pump light) and gain-guiding can bring the cavity into a stable regime (Zayhowski, 1992). The microchip can be close-coupled to a high-power diode laser for pumping, resulting in an extremely compact device (Dixon *et al.*, 1989).

A number of “semimonolithic” or “composite cavity” designs have been explored which attempt to combine the benefits of the microchip laser with intracavity SHG. In most of these designs the resonator consists of a Nd-doped crystal with a mirror deposited on one end in close proximity to a doubling crystal with a mirror deposited on the opposite end (Figure 5.9). MacKinnon *et al.* (1994) and MacKinnon and

Sinclair (1994) used such a composite-cavity laser to generate 132 mW of 532-nm green light. Their configuration employed a 0.5-mm thick crystal of Nd-doped yttrium vanadate (Nd:YVO₄) and a 2-mm thick KTP crystal. The space between the two crystals was filled with an index matching fluid. Meyn and Huber (1994) generated 522 mW of green light in a similar configuration, using Nd-doped lanthanum scandium borate (Nd:LaSc₃(BO₃)₄).

Blue light generators have also been developed using the microcavity approach. Zarrabi *et al.* (1995) used a 1.2-mm thick Nd-doped yttrium orthoaluminate (Nd:YAlO₃) crystal lasing on the $^4F_{3/2} \rightarrow ^4I_{9/2}$ transition at 930 nm and a 1.3-mm thick KN crystal to generate 15 mW of blue power with Ti:S laser pumping. Matthews *et al.* (1996) used Nd:YAG lasing on the same transition at 946 nm to generate 25 mW of 473-nm light with diode laser pumping. Mizell *et al.* (1996) used a similar configuration to produce 23 mW of blue light.

However, the composite-cavity approach has some limitations. The gain spectrum of common laser materials often has many peaks (not a single peak as shown in the simplified depiction given in Figure 5.7), so that it may be difficult to adequately reduce the gain for adjacent longitudinal modes. Furthermore, there is a trade-off between wanting to keep the cavity length short (in order to achieve single-longitudinal-mode operation) and wanting to make both the laser medium long (in order to maximize absorption of the pump energy) and the nonlinear crystal long (in order to maximize blue-green generation). When the cavity is long enough so that more than one longitudinal mode falls with the gain bandwidth, “spatial hole burning” may permit more than one mode to oscillate (Zayhowski, 1990). Spatial hole burning arises because the intensity pattern associated with a particular longitudinal mode in a standing-wave resonator varies spatially along the resonator axis as a result of interference between forward- and backward-propagating waves (hence the term *standing-wave* resonator). Where the intensity is high, the mode efficiently extracts energy from the gain medium, and the gain is correspondingly saturated. However, at the nulls of the standing-wave pattern, the gain is essentially unsaturated (Figure 5.10). Since adjacent longitudinal modes squeeze either one more or one less half-wavelength into the length of the resonator, their nulls do not coincide with those of the original mode, and they may be able to extract sufficient gain from the unsaturated regions to reach threshold. Thus, spatial hole burning must be suppressed in order to support single-longitudinal-mode oscillation, and we will describe a number of approaches for doing so in later sections.

Two approaches to keeping the cavity length short while maintaining high absorption of the pump light have been explored. One involves the use of stoichiometric laser materials, such as NdP₅O₁₄, NdAl₃(BO₃)₄, and LiNdP₄O₁₂ which have higher concentrations of the active ion than materials in which the ion is doped into a host crystal (Chinn *et al.*, 1976, Dixon *et al.*, 1989). Another approach that has been

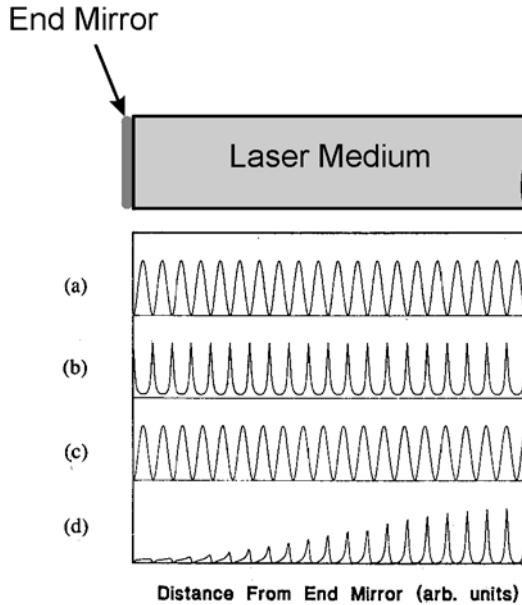


Figure 5.10: Spatial hole burning in a resonator using a dielectric mirror deposited directly on one end of the laser medium: (a) standing-wave intensity pattern of first longitudinal mode; (b) population inversion at ten times above threshold, showing spatial holes burned by first longitudinal mode; (c) standing-wave intensity pattern of second longitudinal mode; (d) differential gain, which is integrated over length of laser medium to give total gain seen by second longitudinal mode. (Reprinted with permission from Kintz and Baer (1990). Copyright: IEEE.)

explored involves placing mirrors around the laser crystal to form a passive resonator at the *pump* frequency. In this way, the pump energy makes many passes through the laser crystal and the overall absorption is increased (Cuthbertson and Dixon, 1991, Kozlovsky and Risk, 1992, Taira *et al.*, 1996).

Intracavity mode selection The limitations of microchip or composite-cavity lasers have fostered a great deal of work aimed at achieving robust single-longitudinal-mode operation in lasers using more conventional resonator designs – that is, assembled from discrete components and having lengths of several centimeters. Suppression of unwanted longitudinal modes requires ensuring that no mode other than the desired one is able to satisfy the condition that round-trip gain equals round-trip loss. There are two ways to accomplish this: reduce the gain available to unwanted modes or increase the loss experienced by unwanted modes.

Decreasing the gain for unwanted modes Kintz and Baer (1990) identified a way in which the gain available to unwanted longitudinal modes can be minimized. Suppose that one of the mirrors which forms the resonator is deposited directly on

the end of the laser medium and the laser is end-pumped through that mirror. This placement of the mirror forces the standing-wave pattern for all longitudinal modes to have a common node at the mirror-end of the laser medium (Figure 5.10). Thus, for a certain distance away from this mirror, the standing-wave patterns corresponding to different longitudinal modes are almost identical. Hence, if one longitudinal mode begins to oscillate and causes spatial holes in the gain, adjacent longitudinal modes will have a poor overlap with the unsaturated gain, and will be not be able to reach threshold. However, since the standing-wave patterns of different longitudinal modes have slightly different wavelengths, if we go far enough away from the mirror, these patterns will become sufficiently out-of-phase that a second longitudinal mode will be able accumulate sufficient gain to reach threshold. By truncating the laser medium before this point is reached, unwanted longitudinal modes can be deprived of gain, and thereby suppressed. In order to meet this condition while still absorbing a large fraction of the incident pump light, it is desirable to use a laser material with inherently strong absorption. Unlike the microchip approach, this scheme does not require a short *cavity* length for single-frequency operation, only a short *absorption* length. Hence, we can use a cavity long enough to conveniently accommodate a doubling crystal of reasonable length, without the need to resort to the more demanding fabrication of a composite-cavity microchip laser.

Sasaki *et al.* (1991) demonstrated the use of this approach in a laser designed for intracavity SHG. They used Nd:YVO₄ as the laser medium and KTP as the doubling crystal, and achieved 16 mW of green output power. Nagai *et al.* (1992) further investigated the noise characteristics of the green output from such lasers, and demonstrated 3 mW of green output power with a relative intensity noise of -140 dB/Hz at frequencies over 1 MHz. Helmfrid and Tatsuno (1994) provided an in-depth theoretical consideration of the characteristics of intracavity-doubled lasers using this approach for suppression of multi-longitudinal mode operation.

Increasing the loss for unwanted modes The loss for unwanted modes can be increased by inserting some sort of filter into the cavity that allows one longitudinal mode to pass with little or no loss, while inflicting higher losses on all other longitudinal modes. One such filter is a simple étalon inserted into the cavity, as Baer used in his original study of the “green problem”.

If a thin metal film is placed inside a laser cavity, modes which do not have a node at the location of the film will experience loss, while a mode with a node at that location will experience relatively little loss – thus, such a film acts as a mode selector (Figure 5.11). This approach was first demonstrated in the late 1960s (Troitskii and Goldina, 1968, Smith *et al.*, 1968), but has experienced a resurgence of interest for use with miniature diode-pumped solid-state lasers (Effenberger and Dixon, 1994, Jabczynski *et al.*, 1997).

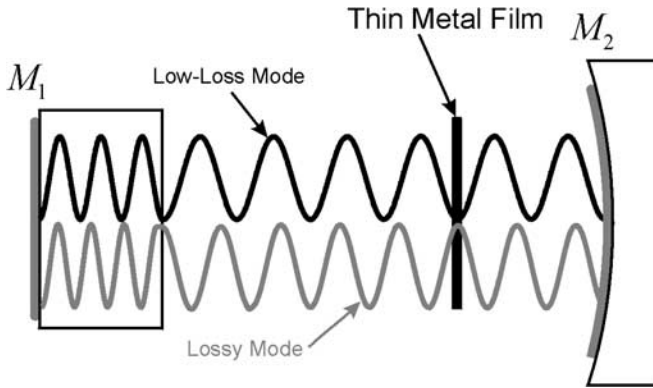


Figure 5.11: Use of a thin metal film to introduce loss for unwanted longitudinal modes.

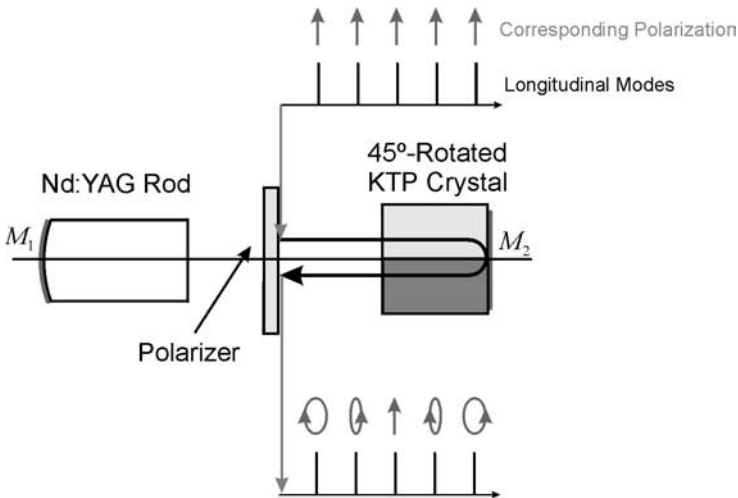


Figure 5.12: Use of a birefringent filter to suppress unwanted longitudinal modes. (Modeled after arrangement used by Nagai *et al.* (1992).)

Another approach is to introduce a “birefringent filter” into the resonator (Figure 5.12). Suppose a perfect polarizer and a birefringent element are placed inside the resonator, with the transmission axis of the polarizer at 45° to the principal axes of the birefringent element. Light that is transmitted through the polarizer must emerge with a linear polarization state aligned to the polarizer axis. After making a round trip through the birefringent element, the light emerges in some elliptically-polarized state, determined by the differential phase shift $\delta = 4\pi \Delta n L / \lambda$ accrued between components polarized along the fast and slow axes of the crystal, where Δn is the birefringence and L is the length of the birefringent element. If this round-trip retardation δ is an integer multiple of 2π , the polarization state will be

the same as what we began with, namely a linearly-polarized state aligned with the polarizer axis. This state will then pass through the polarizer with very little attenuation. Hence, if we can arrange for a particular longitudinal mode to satisfy the condition $\delta = 2m\pi$, it can circulate in the cavity with very little loss. However, since δ varies with λ , adjacent longitudinal modes will emerge from the birefringent element with elliptical polarization states that will experience significant loss, and will stay below threshold.

In practice, partial polarizers – such as Brewster plates or surfaces – are used instead of the perfect polarizer posited in our description. In such cases, the effect of the polarizer on the polarization state present in the laser is less dramatic than what has been described above; however, the principle still applies, since only small differences in round-trip loss are needed to discriminate against unwanted modes, and a partial polarizer within the cavity is sufficient to supply these (Bloom, 1974).

Since Type-II SHG in KTP requires the input fundamental light to be polarized at 45° to the principal axes anyway, KTP can be used as the birefringent element for mode selection and the frequency doubler. Fan *et al.* (1986) demonstrated this principle, using Brewster surfaces on the Nd:YAG crystal to provide the polarization discrimination. Nagai *et al.* (1992) incorporated a separate Brewster plate in the cavity, and achieved 3 mW of green light, limited by the power available from their diode pump laser. Suzuki *et al.* (1994) used spatial walk-off in the Nd:YVO₄ laser crystal to produce the polarization-dependent loss and achieved 20 mW of green output power with noise < -120 dB/Hz.

Elimination of spatial hole burning Even with the use of a short cavity, a gain medium with a short absorption length, or intracavity filters, conventional standing-wave lasers will generally run in multiple longitudinal modes if pumped hard enough, since spatial hole burning has not been eliminated.

Several approaches have been suggested for reducing or eliminating spatial hole burning, including physically moving the laser medium in order to “smear out” the holes (Danielmeyer and Nilsen, 1970) or by electro-optically shifting the standing-wave pattern with respect to the gain medium (Danielmeyer and Turner, 1970). However, two main approaches have been pursued to eliminate spatial hole burning in conjunction with intracavity SHG of diode-pumped solid-state lasers: “twisted mode” resonators and unidirectional ring resonators.

Twisted mode resonators Evtuhov and Siegman (1965) proposed that spatial hole burning could be eliminated in a standing-wave cavity by placing a quarter-wave plate at each end of the resonator, with the laser medium in between (Figure 5.13). It can be shown that in order for the polarization state to be self-consistent after a round trip, it must be linear at each mirror and circularly polarized

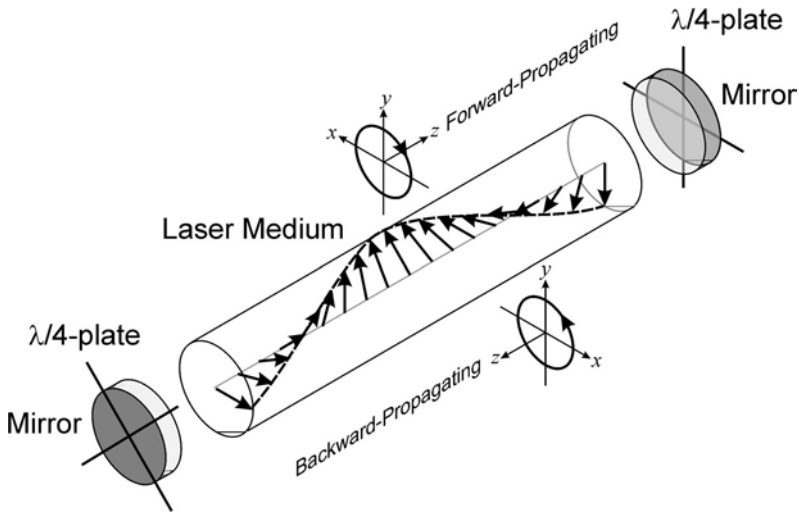


Figure 5.13: Diagram of the twisted mode resonator. Quarter-wave plates with integral mirrors are placed at the ends of the resonator, with their principal axes rotated from each other by some angle (in this case, 45°). The eigenpolarizations are circularly polarized in the laser medium. Superposition of the forward- and backward-propagating light at any point in the gain medium gives a linear polarization state. The orientation of this linearly-polarized electric field vector varies with position, tracing out a helix. The pitch of the helix is equal to the optical wavelength, and is greatly exaggerated here for clarity.

in the laser medium. The interference of two counterpropagating circularly-polarized waves gives a linearly-polarized electric field vector, the angle of which rotates with position in the laser medium, tracing out a helix. The amplitude of the wave is thus the same everywhere in the laser medium, and the gain is equally saturated everywhere, so that spatial hole burning is eliminated. Wallmeroth and Peuser (1988) demonstrated the use of this approach to achieve single-frequency operation of a diode-pumped Nd:YAG laser up to 255 mW in a TEM_{00} spatial mode, and Nakagawa *et al.* (1994) used a slightly modified form of this idea to achieve 950 mW of single-frequency power.

This basic approach can be combined with intracavity SHG to eliminate the “green problem”. Anthon *et al.* (1992) used the cavity design in Figure 5.14 to create the helical electric field in the Nd:YAG, yet still present the correct polarization components to the KTP crystal for Type-II SHG. They were able to generate up to ~ 100 mW of green output power, but noted that the laser typically ran in two orthogonally-polarized longitudinal modes, and often exhibited bistable behavior.

Unidirectional ring lasers An alternative – and more popular – approach for eliminating spatial hole burning has been to construct a unidirectional ring laser, in which the fundamental energy travels through the gain medium in only one direction, so that standing waves do not form and spatial hole burning cannot

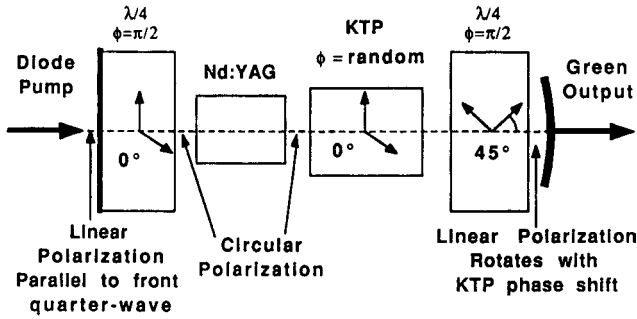


Figure 5.14: Modified twisted-mode resonator for intracavity SHG with KTP used by Anthon and coworkers. (Reprinted with permission from Anthon *et al.* (1992). Copyright: IEEE.)

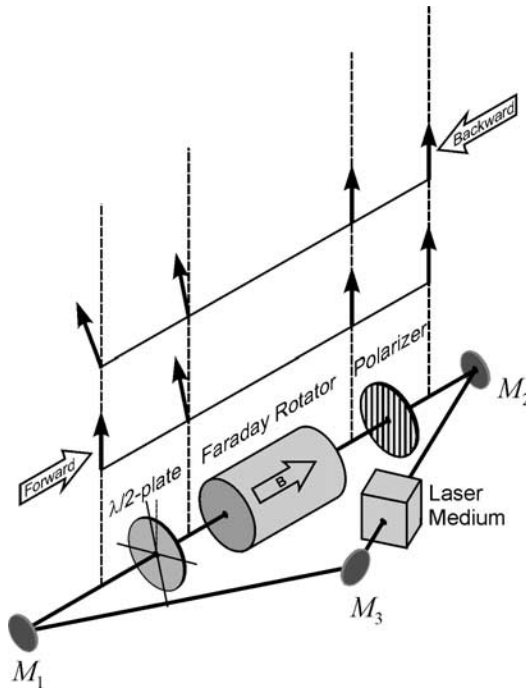


Figure 5.15: A ring resonator using a Faraday rotator to enforce unidirectional operation.

occur. If the loss suffered by a mode propagating in one direction differs sufficiently from the loss encountered by a mode propagating in the other, the laser will oscillate in the direction having the lower loss. Clobes and Brienza (1972) produced this differential loss in a ring resonator using the combination of a half-wave plate and Faraday rotator to produce a direction-dependent polarization rotation (Figure 5.15). A polarizer (in their implementation, Brewster-angled surfaces of the Nd:YAG) establishes a preferred linear polarization state. For propagation in a

clockwise direction around the ring, the Faraday device rotated the linear polarization state of the light by a small angle $+\theta$, and the half-wave plate rotated it by $-\theta$ to restore its original orientation, so that very little loss occurred when the wave passed through the polarizer. However, light propagating in the counterclockwise direction experienced a polarization rotation of $+\theta$ from passage through the Faraday rotator and another $+\theta$ from passage through the half-wave plate. Thus, because the linear polarization state has been rotated by a total of 2θ , light propagating in the counterclockwise direction experiences loss at the polarizer. This difference in loss for light propagating in one sense around the loop compared with the other is sufficient to force oscillation in a single direction, clockwise in this example.

Martin *et al.* (1996) implemented this basic scheme using a bow-tie resonator configuration, a terbium gadolinium garnet (TGG) crystal as the Faraday rotator, a separate Brewster plate polarizer, and a KTP crystal for frequency-doubling. Since frequency-doubling of 1064 nm in KTP requires critical phasematching, spatial separation of the two principal axis polarization states due to Poynting-vector walk-off can be a problem. To combat this problem, they used a special oblique-incidence orientation of KTP which eliminates walk-off, but preserves phasematching (Nightingale, 1993). Pumping with 20 W of diode-laser power, they were able to achieve 3 W of 532-nm output in a single frequency in a TEM₀₀ mode.

In order to reduce the number of lossy intracavity elements present inside the ring, Nightingale and Johnson (1992) developed the resonator configuration shown in Figure 5.16. In this design, only two mirrors were required – the ring was closed by making the Nd:YAG into a prism to refractively redirect the beam. Faraday rotation was obtained by applying a magnetic field to the YAG itself. A Brewster-cut piece of quartz served as both the polarizer and reciprocal polarization rotator. The combination of the KTP and the polarizer also form a birefringent filter, as described above. Using this arrangement with walk-off compensated KTP and pumping with 4 W of diode laser power, they were able to achieve a stable, single-frequency green output power of 0.9 W. In a similar system using LBO as the nonlinear crystal, Selker *et al.* (1996) reported in excess of 8.5 W of single-frequency green output.

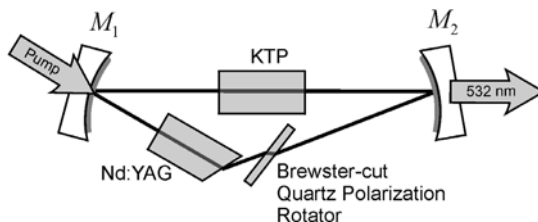


Figure 5.16: Unidirectional ring resonator for intracavity SHG used by Nightingale and Johnson (1992).

5.4 BLUE LASERS BASED ON INTRACAVITY SHG OF 946-nm Nd:YAG LASERS

The most popular Nd^{3+} transition has been the one beginning on the $^4F_{3/2}$ level and terminating on the $^4I_{11/2}$ level. This transition produces lasing at a wavelength around $1\ \mu\text{m}$, the precise wavelength depending on the host into which Nd^{3+} is doped (e.g., $1.064\ \mu\text{m}$ in Nd:YAG). The $^4I_{11/2}$ level has an equilibrium thermal population that is essentially zero at room temperature, so that only a small population must be excited into the $^4F_{3/2}$ upper laser level in order to produce gain. As we have seen, this transition has been the primary “workhorse” for generation of green light by intracavity SHG, and we have surveyed several implementations in our discussion of the “green problem”.

However, the $^4F_{3/2} \rightarrow ^4I_{9/2}$ transition in Nd-doped crystals has also attracted interest, because it provides lasing at a somewhat shorter wavelength (946 nm in Nd:YAG). The main drawbacks of this transition are that the gain cross-section is about an order of magnitude lower than that of the $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition, and the lower laser level is the uppermost Stark component of the ground-state manifold, which has a nonnegligible equilibrium thermal population at room temperature (Figure 5.17). Since there is an equilibrium population in this level, light at 946 nm

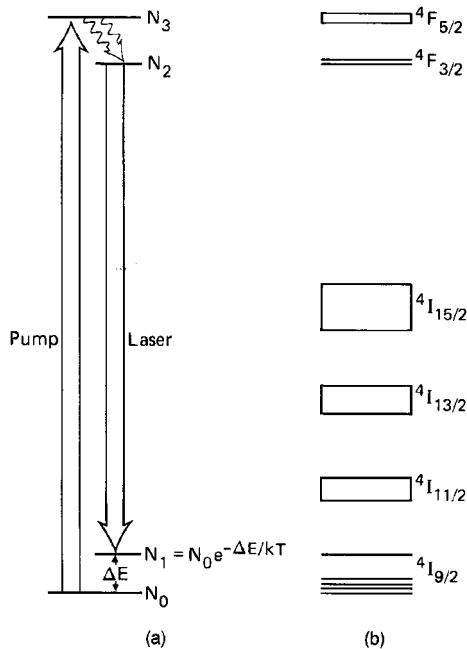


Figure 5.17: (a) Idealized energy-level diagram and (b) energy-level diagram for Nd:YAG. In (b), the actual splittings are shown only for selected levels; for the rest, the range over which level splitting occurs is indicated by a rectangle. (Reprinted with permission from Risk (1988).)

present in the crystal will be absorbed. This “reabsorption loss” is an additional loss which must be overcome to achieve lasing, and can be the largest single component of intracavity loss ($\sim 0.9\%/mm$ in Nd:YAG at room temperature). However, unlike the fixed cavity losses, reabsorption loss is a *saturable* loss – when the intracavity intensity is sufficiently high, the loss is saturated down to a negligible value so that it does not seriously degrade the slope efficiency.

These effects have been explored through mathematical modeling of lasers exhibiting reabsorption losses (Fan and Byer, 1987a, Risk, 1988, 1997, Taira *et al.* 1997). End-pumping of such lasers has been the most common configuration subjected to modeling, since this approach permits concentration of the pump energy within the mode volume of the laser. A brief sketch of the results obtained from this modeling will help to illustrate how reabsorption loss affects laser performance. For an end-pumped laser, the incident pump power required to reach threshold is given by:

$$P_{P,th} = \frac{\pi h \nu_p (w_L^2 + w_p^2) (L + T + 2\sigma N_1^0 l)}{4\sigma \tau \eta_a f} \quad (5.22)$$

where h is Planck’s constant, ν_p is the pump laser frequency, w_L is the gaussian beam radius of the laser mode in the gain medium, w_p is the gaussian beam radius of the focused pump light in the gain medium, L represents cavity losses other than the output coupling T , σ is the gain cross-section, N_1^0 is the equilibrium thermal population of the lower laser level, l is the length of the gain medium, τ is the lifetime of the upper laser level, $\eta_a = (1 - e^{-\alpha l})$ is the fraction of pump power absorbed in the gain medium (where α is the absorption coefficient), and $f = f_1 + f_2$, where f_1 is the fraction of the $^4I_{9/2}$ population that resides in the Stark component used as the lower laser level and f_2 is the fraction of the $^4F_{3/2}$ population that resides in the Stark component used as the upper laser level. The term $2\sigma N_1^0 l$ gives the reabsorption loss resulting from the nonnegligible equilibrium population in the lower laser level. We can see that reabsorption loss increases the threshold just as does any other loss. We also see that as we increase the length of the crystal, the fraction of pump power which is actually absorbed by the gain medium increases (which is good), but the reabsorption loss also increases (which is bad). Thus, there is an optimum length for the gain medium which minimizes the threshold pump power (Figure 5.18). In addition, if the lowest possible threshold is the objective, it is desirable to make both w_L and w_p small.

However, once above threshold, the circulating 946-nm light saturates the reabsorption loss, so that it becomes negligible at sufficiently high powers. The slope efficiency of the laser above threshold is given by

$$\frac{dP_{out}}{dP_p} = \frac{T}{L + T} \frac{\nu_L}{\nu_p} \eta_a \frac{dS}{SF} \quad (5.23)$$

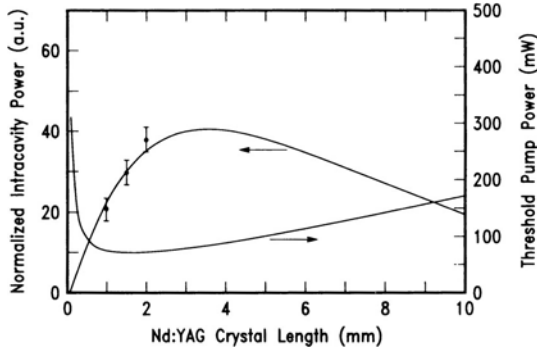


Figure 5.18: Variation of threshold pump power with the length of the laser medium for a 946-nm Nd:YAG laser. The figure also shows the variation of intracavity power. (Reprinted with permission from Risk and Lenth (1989a).)

where $S = 4\sigma\tau P_L/\pi h\nu_L w_L^2$ is a normalized laser power and $F = 4P_P\tau\sigma\eta_a/\pi h\nu_P w_L^2(L+T)$ is a normalized pump power. The relationship between S and F is given by the expression (Risk, 1997)

$$\frac{dS}{dF} = \frac{1 + \frac{B}{fS} \ln(1 + fS)}{f^2 F^2 \int_0^\infty \frac{(e^{-x} - Ba^2/fF)e^{-2a^2x}}{(1 + fSe^{-a^2x})^2} dx} \quad (5.24)$$

where

$$B = \frac{2\sigma N_1^0 l}{L + T}$$

is the ratio of reabsorption loss to the other losses present in the cavity, and $a = w_P/w_L$ is the ratio of the pump waist to the laser waist. In general, this equation must be numerically solved given a particular value of F to find the corresponding value of S (however, it can be solved analytically for integer values of a^{-2} (Fan and Byer, 1987a)). An example of such a numerical solution is given in Figure 5.19, which shows that as the laser intensity increases, dS/dF becomes independent of reabsorption loss and, hence, so does the overall slope efficiency of the laser. The dependence of the dS/dF on the ratio a of pump waist to laser waist becomes complicated when reabsorption loss is present. When there is no reabsorption loss ($B = 0$), as in the case of a 1064-nm laser, dS/dF is maximized by making $a = 0$; that is, by making the pump spot small compared with the laser waist. This condition ensures that a high stimulating intensity is present everywhere there is population inversion, so that gain is efficiently extracted. However, when reabsorption loss is present ($B \neq 0$), there is a nonzero optimum value of a that maximizes dS/dF . This optimum, which depends both on the circulating intensity and the reabsorption loss,

Table 5.1. Summary of experiments involving blue light generation using intracavity SHG of a 946-nm laser

Reference	Laser material	Nonlinear material	Blue output power	Comments
Wallace and Harris (1969)	Nd:YAG	KDP	2 kW	Lamp-pumped, Q-switched
Fan and Byer (1987)	Nd:YAG	N/A	N/A	First report of diode-pumping; SHG not demonstrated, but various nonlinear materials considered
Risk and Lenth (1987)	Nd:YAG	LiIO ₃		KN heated to 185 °C for phasematching
Dixon <i>et al.</i> (1988)	Nd:YAG	KN	0.5 mW	Angle-tuned KN used at room temperature
Risk <i>et al.</i> (1989)	Nd:YAG	KN	3.12 mW	Laser crystal length optimized, Nd:YAG laser emission polarized by walk-off in KN crystal
Risk and Lenth (1989a)	Nd:YAG	KN	9 mW	One-step, segmented guide, Rb/Ba exchange
Hollemann <i>et al.</i> (1994)	Nd:YAG	KN	100 mW	Pulse-pumped operation, used novel diffusion-bonded sandwich of undoped YAG and Nd:YAG
Hanson (1995)	Nd:YAG	KN	366 mW	Noncritical phasematching at room temperature using niobium-doped KTP
Miesak <i>et al.</i> (1995)	Nd:YAG	Nb:KTP	100 μW	Composite-cavity microchip design; pumped by Ti:S
Zarrabi <i>et al.</i> (1995)	Nd:YAlO ₃	KN	15 mW	Composite-cavity microchip design; pumped by diode laser
Matthews <i>et al.</i> (1996)	Nd:YAG	KN	25 mW	Composite-cavity microchip design; pumped by diode laser
Mizell <i>et al.</i> (1996)	Nd:YAG	KN	23 mW	Composite-cavity microchip design; pumped by diode laser
Kellner <i>et al.</i> (1996)	Nd:YAG	BBO	75 mW	
Kellner <i>et al.</i> (1996)	"	LBO	38 mW	
Kellner <i>et al.</i> (1996)	"	LiIO ₃	57 mW	
Kellner <i>et al.</i> (1997)	Nd:YAG	LiIO ₃	550 mW	
Kellner <i>et al.</i> (1997)	"	KN	410 mW	
Kellner <i>et al.</i> (1997a)	Nd:YAG	LiIO ₃	520 mW	Used novel diffusion-bonded sandwich of undoped YAG and Nd:YAG
Kellner <i>et al.</i> (1997a)	"	BBO	550 mW	
Kellner <i>et al.</i> (1997a)	"	LBO	372 mW	
Harada <i>et al.</i> (1997)	Nd:YAG	PPLN	5.9 mW	cw, diode-pumped
Pierrou <i>et al.</i> (1999)	Nd:YAG	PP-KTP	740 mW	Ti:S laser pumped, metal-organic complex nonlinear crystal
Wang <i>et al.</i> (1999)	Nd:YAG	CMTC	1.6 mW	
Zeller and Peuser (2000)	Nd:YAG	LBO	1.5 W	cw, diode-pumped

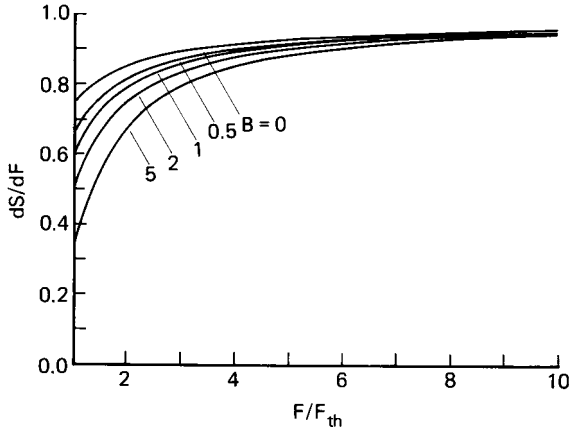


Figure 5.19: Variation of dS/dF with F/F_{th} for various values of B , with $a = 1.0$. (Reprinted with permission from Risk (1988).)

arises from a trade-off between making a small, which ensures a high stimulating intensity over all of the pumped region, and making a large, which minimizes the overlap between the laser field and lossy, unpumped regions which exhibit reabsorption loss.

Blue light at 473 nm can be obtained by intracavity SHG of a 946-nm Nd:YAG laser. Early experiments used dye lasers or Ti:S lasers as the pump sources, since the high spatial quality of the pump beam made it easier to control the value of $a = w_P/w_L$, as dictated by the mathematical modeling. However, several groups have demonstrated and developed efficient diode-laser-pumped 946-nm lasers. Intracavity SHG has been explored with several different materials; historically, KN has probably received the most attention, and both noncritical phasematching at high temperatures and angle-tuned phasematching at room temperature have been investigated. Work on generation of blue light using intracavity SHG of 946-nm lasers is summarized in Table 5.1.

5.5 INTRACAVITY SHG OF Cr:LiSAF LASERS

Another laser that has been investigated for diode-pumped blue-green sources is the $\text{Cr}^{3+}:\text{LiSrAlF}_6$ (or, as it is frequently abbreviated, Cr:LiSAF) laser (Payne *et al.*, 1989). Cr:LiSAF efficiently absorbs red light between 600 and 700 nm, and fluoresces over a broad range between 750 and 1000 nm (Figure 5.20). The advent of high-power red GaAlInP diode lasers has made diode pumping of Cr:LiSAF lasers feasible, and the emission wavelength of these lasers is compatible with a number of frequency-doubling crystals, such as KN.

Balembois *et al.* (1992) demonstrated SHG of a Q-switched Cr:LiSAF laser using a krypton laser for pumping, LiIO_3 as the doubling crystal, and a birefringent

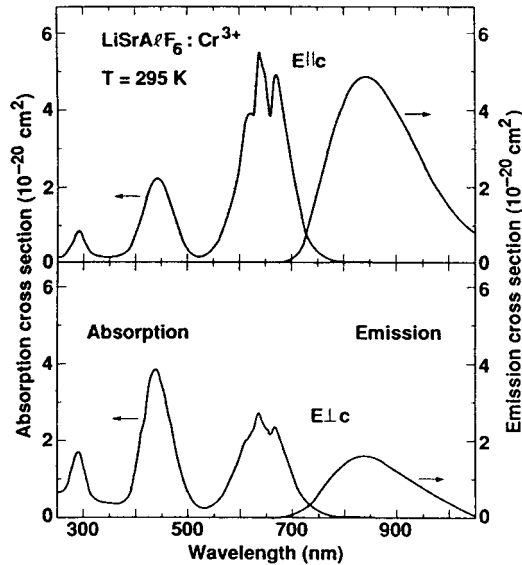


Figure 5.20: Polarized absorption and emission spectra of Cr:LiSAF at room temperature. (Reprinted with permission from Payne *et al.* (1989). Credit must be given to the University of California, Lawrence Livermore National Laboratory, and the Department of Energy under whose auspices the work was performed, when this information or a reproduction of it is used.)

filter to tune the lasing wavelength. They were able to generate light in the 395–435 nm range, and reported an average power of 7 mW at 407 nm. Subsequently, Falcoz *et al.* (1995) used two 500-mW GaAlInP diode lasers as pump sources, and KN as the doubling crystal. They were able to produce blue light in the range 427–443 nm, with a cw output power of 13 mW. Laperle *et al.* (1997) used a set of 19 fiber coupled 665-nm laser diodes to pump Cr:LiSAF, and used LBO as the doubling crystal (Figure 5.21). They achieved a blue output power at 435 nm of 20 mW when the laser was operated cw, and 250 mW when operated with a 25% duty cycle. Eichenholz *et al.* (1998) have reported a composite-cavity Cr:LiSAF/KN microchip laser which produced 0.37 mW of 430-nm blue light. Makio *et al.* (2000) reported 67 mW of cw blue power generated using LBO for frequency-doubling of a diode-pumped Cr:LiSAF laser.

5.6 SELF-FREQUENCY-DOUBLING

As we mentioned in Chapter 2, in some cases it is possible to incorporate active ions directly into a nonlinear material, so that the functions of lasing and frequency-doubling both occur within the same crystal. In principle, having such a material available simplifies the design of blue-green sources based on intracavity SHG.

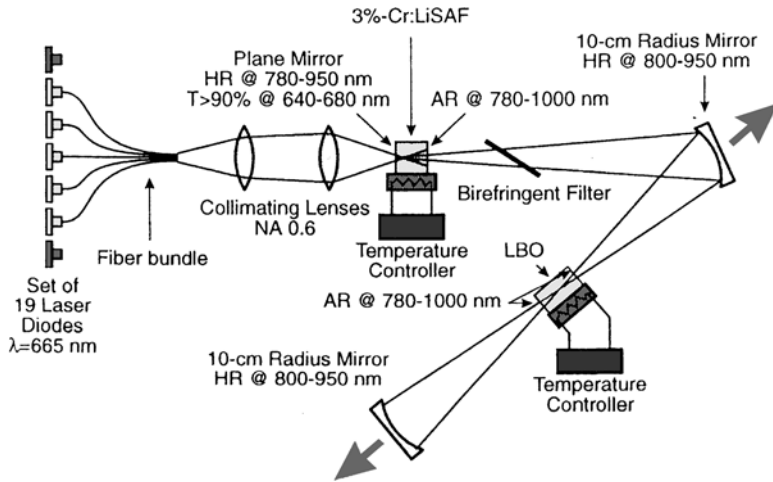


Figure 5.21: Intracavity-doubled, diode-pumped Cr:LiSAF laser of Laperle and co-workers. (Reprinted with permission from Laperle *et al.* (1997).)

In practice, producing materials which have both excellent nonlinear properties and excellent laser characteristics has been difficult. Incorporation of rare-earth dopants into a nonlinear material may complicate crystal growth and degrade the optical quality of the material. Other problems also arise – for example, when Nd^{3+} ions are incorporated into LN, the gain becomes polarization-dependent due to the anisotropy of the LN crystal. Unfortunately, the high-gain π -polarization is the wrong one for Type-I SHG, and the laser must be forced to oscillate in the low-gain σ -polarization. Thus, while self-doubled systems are simpler in principle than systems using separate laser and nonlinear materials, they sometimes have additional constraints and disadvantages which must be overcome in making practical blue-green sources.

5.6.1 Nd:LN

Although several materials have been investigated as self-doubling media (see Chapter 2), most laser experiments have been done with either Nd:MgO:LN or $\text{Nd}_x\text{Y}_{1-x}\text{Al}_3(\text{BO}_3)_4$ (NYAB). Here, we briefly review some of the progress toward making a practical blue-green source based on the use of these self-doubling materials.

Self-frequency-doubling in Tm-doped LN was reported in 1969 (Johnson and Ballman, 1969) and self-frequency-doubling in Nd-doped LN was reported in 1979 (Dmitriev *et al.* 1979). In 1986, Fan and coworkers reported the spectroscopic properties of Nd:MgO:LN and demonstrated a self-doubling laser that produced ~ 1 mW of green output power when pumped with a dye laser and when the crystal

was heated to $\sim 150^\circ\text{C}$ to achieve noncritical phasematching (Fan *et al.*, 1986a). Cordova-Plaza *et al.* (1988) demonstrated diode-pumping of a Nd:MgO:LN laser, but simultaneous frequency-doubling in a diode-pumped laser was not reported until 1994 by Ishibashi and coworkers, who achieved 0.27 mW of green output power using an angle-tuned room-temperature crystal (Ishibashi *et al.*, 1994). Yamamoto *et al.* (1994) investigated Sc_2O_3 (rather than MgO) doping to reduce photorefractivity. They were able to achieve room-temperature noncritical phasematching and produced 0.14 mW of 546 nm green light, pumping with a Ti:S laser. Ishibashi *et al.* (1996) reported a monolithic self-doubling Nd:MgO:LN laser, emitting about 0.2 mW of green power. Improvements in the power available from diode laser pump sources made it possible for Zhang *et al.* (1996) to achieve 8 mW at 547 nm from a Nd:MgO:LiNbO₃ laser. They used an angle-cut crystal to achieve phasematching, and spatial walk-off in the crystal acted as a polarizer to suppress oscillation in the unwanted high-gain π -polarization.

5.6.2 NYAB

Self-doubling in NYAB was reported in 1981 by Dorozhkin *et al.* (1981). Lu *et al.* (1989) studied the effect of Nd concentration on the self-doubling properties of NYAB, and found the optimum Nd concentration to be $\sim 10\%$ (compare this with the typical neodymium concentration in Nd:YAG of $\sim 1\%$). They constructed a 1.06 μm laser using this material, pumped by a dye laser, and demonstrated self-doubling to 0.53 μm . Amano *et al.* (1989) reported diode-pumped operation of a green NYAB laser. Schütz *et al.* (1990) demonstrated 10 mW of cw green output power from a NYAB laser pumped by a 1-W diode laser array. Hemmati (1992) pumped a NYAB laser with two 1-W diode laser arrays and obtained 50 mW of green output power, and Bartschke *et al.* (1997) have reported 225 mW of cw 531-nm output power from an NYAB laser.

Q-switched operation of NYAG lasers has also been of interest. Stone *et al.* (1990) obtained a peak power of 3 W by Q-switching a NYAB laser pumped with 180 mW from a diode laser (the same laser produced 1 mW in cw operation), and Li *et al.* (1994) reported a peak power of 20 W.

Self-doubling materials are attractive for monolithic, microchip lasers. Grubb *et al.* (1991) demonstrated microchip laser operation of 500- μm thick NYAB chips and obtained 2 mW of green output in a TEM₀₀ mode when pumped by a 200-mW laser diode array. Zarrabi *et al.* (1993) fabricated a NYAB microchip laser by depositing mirror coatings directly on the sides of a thin (~ 0.9 mm) slab of the material which had been polished flat and parallel. Pumping with a 1-W GaAlAs laser diode, they were able to achieve 20 mW of output power at 531 nm in a TEM₀₀ mode.

5.6.3 Periodically-poled materials

Perhaps the ultimate expression of the self-frequency-doubling idea would be doping a rare-earth ion or other active species into a material that can be periodically (or aperiodically) poled, in order to take advantage of the design flexibility afforded by quasi-phasematching. Capmany *et al.* (2000) reported a laser based on Nd-doped aperiodically-poled LN (PPLN), and were able to generate 1.5 mW of green light from self-doubling of the $^4F_{3/2} \rightarrow ^4F_{11/2}$ transition (1084 nm) and 0.5 mW of blue light resulting from mixing of light from this transition with the 744-nm pump light. The same group has also reported generation of 10.5 mW of green light using self-doubling in PPLN doped with Yb (Capmany *et al.*, 2000a).

5.6.4 Other materials

Work continues to identify new materials suitable for use in practical self-doubling devices. Capmany *et al.* (1998) reported generation of $\sim 90 \mu\text{W}$ using Nd:LaBGeO₅. Mougél *et al.* (1998) reported generation of 21 mW of green light using laser diode pumping of Nd:Ca₄GdO(BO₃)₃.

5.7 INTRACAVITY SUM-FREQUENCY MIXING

Another approach for generation of blue-green light that involves placing a nonlinear crystal inside a laser resonator is intracavity mixing. In previous chapters, we also discussed the use of sum-frequency mixing for blue-green light generation. In Chapter 3, we considered mixing of two infrared beams, both of which make a single-pass through the crystal. In Chapter 4, we considered configurations in which the intensity of one or both of the interacting beams is increased using a passive resonator. Here, we consider embodiments in which one of the beams involved in the mixing is the intracavity circulating field of a diode-pumped solid-state laser. This approach does not suffer from the “green problem” since mixing of the longitudinal modes of the solid-state laser with each other is not phasematched.

In 1987, Baumert and colleagues reported that KTP could provide noncritical phasematching at room temperature for sum-frequency mixing of 1064-nm and 809-nm light (Baumert *et al.*, 1987). They found that generation of the sum frequency at 459 nm had remarkably broad tolerances to deviations in angle, wavelength, and temperature from optimum phasematching conditions. In particular, the temperature tolerance was remarkably broad – the temperature of the KTP crystal could be varied over a 350 °C range before seriously degrading the efficiency of SFG.

An especially interesting aspect of this phasematching condition was the particular combination of infrared wavelengths. A Nd:YAG laser generating 1064 nm light can be pumped at 809 nm, which suggests the possibility that a KTP crystal

Table 5.2. Summary of experiments involving intracavity SFG

Reference	λ_3 (nm)	λ_2 (nm)	λ_1 (nm)	Material	Blue output power	Comments
Baumert <i>et al.</i> (1987)	459	1064	809	KTP	0.96 mW	Demonstrated broad tolerances for SFG in KTP
Risk <i>et al.</i> (1988)	459	1064	809	KTP	1 mW	Used same 809-nm dye/diode laser for pumping and mixing; demonstrated rapid modulation
Anthon <i>et al.</i> (1988)	459	1064	808	KTP	1 mW	Pointed out advantages of using separate diode lasers for pumping/mixing
Risk and Lenth (1989)	459	1064	809	KTP	14 μ W	Used separate low-power (12 mW) single-mode diode laser for mixing; generated 5 ns pulses
Kean and Dixon (1992)	459	1064	809	KTP	1.2 mW	Resonantly-pumped Nd:YAG laser
Kean <i>et al.</i> (1993)	459	1064	809	KTP	20 mW	1-W broad area diode laser pump, 100-mW single-mode diode laser for mixing
Shichijyo <i>et al.</i> (1994)	490	1064	910	KN	20 mW	Mixed Nd:YVO ₄ laser with Ti:S
Scheps and Myers (1994)	459	1064	809	KTP	109 mW	Used novel dual-wavelength Ti:S laser to provide both 1064 nm and 809 nm
Scheps and Myers (1994a)	459	1064	806	KTP	145 mW	Used novel “branched” laser configuration with two separate Nd:YAG and Ti:S gain elements
Danailov and Apai (1994)	589	1064	1319	KD*P	150 μ J	Dual wavelength Nd:YAG laser
Shichijyo <i>et al.</i> (1996)	418	1064	690	KN	8 mW	
Shichijyo <i>et al.</i> (1996)	450	1064	780		10 mW	
Shichijyo <i>et al.</i> (1996)	475	1064	860		35 mW	
Kretschmann <i>et al.</i> (1997)	618	1080	1444	LBO	212 mW	
Kretschmann <i>et al.</i> (1997)	618	„	1444	KTP	172 mW	
Kretschmann <i>et al.</i> (1999)	618	1080	1444	PPLN	186 mW	
Wang <i>et al.</i> (1999a)	436	946	809	CMTc	310 μ W	Nd:YAG mixed with Ti:S; novel metal-organic complex nonlinear crystal

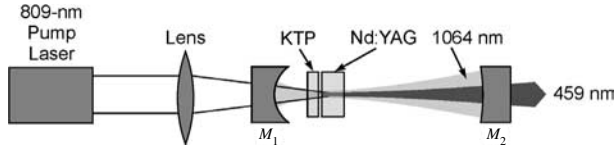


Figure 5.22: Experimental arrangement used by Risk *et al.* (1988) for intracavity SFG of 459-nm light.

could be placed inside the cavity of a diode-pumped Nd:YAG laser to mix the circulating 1064-nm light with the 809-nm pump light. Risk *et al.* (1988) demonstrated this idea using the setup shown in Figure 5.22, and generated ~ 1 mW of 459-nm light using a dye laser pump source and $\sim 10 \mu\text{W}$ with diode laser pumping. They also showed that rapid modulation of the blue output power could be achieved by direct modulation of the 809-nm pump source.

Anthon *et al.* (1988) demonstrated similar results, and pointed out the advantages of using separate diode lasers for pumping and mixing. The demands placed on the spectral and spatial mode quality of the laser are much less severe for the pumping laser than for the mixing laser; thus, a higher-power diode laser with relatively poor spectral and spatial mode quality can be used for pumping the Nd:YAG laser, while a lower-power diode laser with a single spatial and spectral mode can be used for mixing. Risk and Lenth (1989) also explored this approach, and compared the use of a dye laser, a single-mode laser diode, and a broad-area laser diode as the mixing source. Owing to rapid improvements in both kinds of diode laser, Kean *et al.* (1993) were able to generate 12 mW of 459-nm radiation using a 1-W broad-area diode laser as the pump source and a 200-mW single-frequency diode laser as the mixing source.

A variety of other combinations of wavelengths and nonlinear materials have been investigated for intracavity sum-frequency mixing. Some of these results are summarized in Table 5.2.

5.8 SUMMARY

In this chapter, we have considered devices which generate blue-green light by placing a nonlinear crystal inside the cavity of a diode-pumped solid-state laser. In one type of device, intracavity SHG is used to produce the second harmonic of the laser wavelength. We saw that this approach has suffered from the “green problem,” in which sum-frequency mixing of longitudinal modes causes random spiking in the second-harmonic output. We examined several approaches which have been studied as solutions to the green problem, and saw that it can be solved, so that low-noise output can be obtained from intracavity-doubled lasers.

Most green light generation work based on intracavity SHG has been done with 1064-nm Nd lasers, but we also saw that blue light can be generated using intracavity SHG of a 946-nm Nd:YAG laser or Cr:LiSAF lasers. We also considered self-doubling materials, as an alternative approach to intracavity SHG.

Finally, we examined intracavity sum-frequency mixing, in which a nonlinear crystal is placed inside a laser resonator to mix the intracavity field of that resonator with a separate, second laser, which usually makes only a single pass through the nonlinear crystal.

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6

Guided-wave SHG

6.1 INTRODUCTION

In the preceding chapters, we considered implementations of SHG and SFG processes in which an infrared beam is focused into a bulk nonlinear crystal in order to achieve high conversion efficiency. In these approaches the laws of diffraction force a compromise between obtaining two desirable conditions: a small spot size and a long interaction length. Focusing tightly produces a small spot size (and therefore high intensity), but diffraction limits the length over which that spot size can be maintained. Focusing more loosely gives a longer interaction length, but reduces the intensity. The optimum compromise between these competing effects is expressed by the Boyd–Kleinman condition, as introduced in Section 2.3.1. In contrast, a waveguiding structure overcomes the tendency of a tightly-confined beam to diffract, thereby “decoupling” the spot size and the interaction length, so that high intensity can, in principle, be maintained over the entire length of the nonlinear medium.

In Chapter 2, we presented the following equation describing SHG in a waveguide, in the low-conversion efficiency limit and assuming that the nonlinear coefficient is uniform over the waveguide cross-section:

$$P_3 = \frac{8\pi^2 d_{eff}^2 \mathfrak{J}_{ov}^2}{\lambda_1^2 \epsilon_0 c N_{eff}^3} P_1^2(z=0) \left(\frac{e^{-\alpha_1 l} - e^{-\frac{\alpha_3 l}{2}}}{\alpha_1 - \frac{\alpha_3}{2}} \right)^2 \quad (6.1)$$

We saw that, based upon this equation, we can state a number of conditions that must be met for efficient nonlinear upconversion in a waveguide, namely:

- phasematching must be achieved (implicit in this equation);
- the losses incurred by both the fundamental and second harmonic, as defined by the attenuation coefficients α_1 and α_3 , must be low;
- the overlap between the waveguide modes carrying the fundamental and second-harmonic energy must be high, as specified by the overlap integral $\mathfrak{J}_{ov}$;

- as in bulk-optic cases, the inherent efficiency of the nonlinear interaction, as expressed through $d_{\text{eff}}^2/N_{\text{eff}}^3$, must be high;
- the fundamental power coupled into the waveguide, P_1 , must be high;
- the interaction length, l , must be long.

Satisfying these conditions, and doing so in a way that can be consistently repeated, has been the one of the main challenges of waveguide-based blue-green light sources.

Attempts to achieve these mathematical conditions in practice, with real waveguides and real lasers, have met with difficulties that fall into two broad groupings: waveguide fabrication issues and device integration issues. In the first category, the problems are those associated with actually making a waveguide capable of efficient frequency conversion in a nonlinear material. A particular concern in this category is the reproducibility of the process, since it is desirable to be able to make high-efficiency waveguides repeatably. In the second category, the problems are associated with integrating the waveguide doubler with a diode laser. Difficulties encountered in this category include efficient coupling of the laser diode power into the waveguide, stabilization of the laser diode in the presence of feedback from the doubling element, ensuring compatibility of the polarization emitted by the waveguide with that required by the doubler, and achieving and maintaining the phasematching condition.

In this chapter, we will consider some of these issues and how they have been addressed in the quest for a practical blue-green light source based on waveguide frequency-doubling.

6.2 FABRICATION ISSUES

The first step in creating a blue-green light source using a waveguide element for SHG is the fabrication of the waveguide element itself. In Chapter 2, we surveyed some of the techniques that have been used to fabricate waveguides in specific materials. Although earlier work in waveguide SHG explored many different materials and used various phasematching schemes, such as modal phasematching (Uesugi *et al.*, 1979) and Čerenkov phasematching (Taniuchi and Yamamoto, 1987), the most significant results have been obtained using QPM in just three materials: PPLN, PPLT and PPKTP.

In each of these materials, two basic methods for fabrication of periodically-poled waveguide structures have been used. In one approach, the crystal is first periodically poled, then the waveguide is added in a second step (Figure 6.1). The periodic poling can be produced by an applied electric field, an applied electron beam, or a chemical process such as ion-exchange. Waveguide fabrication can then be

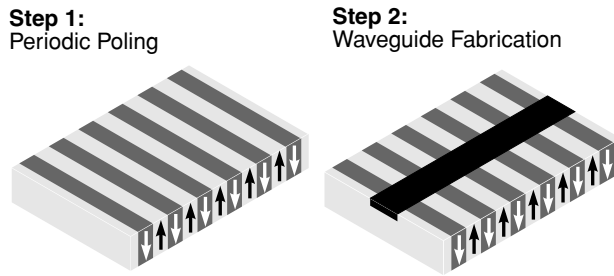


Figure 6.1: Depiction of two-stage process for QPM waveguide fabrication, in which periodic poling is produced in a first step, and waveguide fabrication in a second.

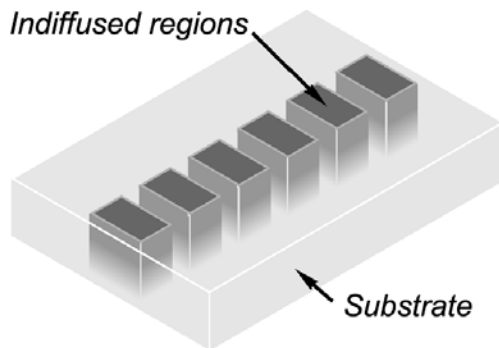


Figure 6.2: Geometry of the segmented waveguide structure used for single-step fabrication of a QPM waveguide.

accomplished by a subsequent ion-exchange or indiffusion step. Since both ion-exchange and indiffusion generally require elevated temperatures, care must be taken that the periodic poling is not undone by the waveguide fabrication process. An additional concern is that the waveguide fabrication process may degrade the nonlinear properties of the material. As we mentioned in Section 2.5.2.3 and illustrated in Figure 2.60, this has been a particular problem with annealed-proton-exchange waveguides in LN.

In the second approach, both periodic poling and formation of the waveguide are accomplished in a single step. This approach has been most often used with KTP, in which both inversion of the ferroelectric domain and formation of the waveguide can be achieved using ion-exchange in a mixed $\text{RbNO}_3/\text{Ba}(\text{NO}_3)_2$ melt, under the right conditions of melt composition, temperature, and exchange time. In order to produce periodic domain inversion, the waveguide must be segmented as shown in Figure 6.2. This segmentation also produces a periodic modulation of the linear properties of the waveguide; however, (perhaps surprisingly) such a segmented guide can still have low loss, as has been observed experimentally

(Bierlein *et al.*, 1990) and justified by several mathematical analyses of such structures (Li and Burke, 1992, Weissman and Hardy, 1993, Stancil, 1996). The extremely anisotropic diffusion properties of *z*-cut KTP lead to sharp, straight boundaries between the segments, which has made KTP a particularly interesting material for segmented waveguide work, although such structures have also been explored in LN and LT (Jongerius *et al.*, 1992, Thyagarajan *et al.*, 1994, Nir *et al.*, 1994, 1995). Since the linear properties of the guide, such as N_{eff} , can be controlled through the duty cycle of the segmentation, these structures have also been explored for purposes other than SHG, including use as Bragg reflectors (Risk and Lau, 1993, Eger *et al.*, 1993, Risk *et al.*, 1994, Roelofs *et al.*, 1994), mode-size transformers (Weissman and Hardy, 1992, Weissman and Hendel, 1995), wavelength demultiplexers (Weissman *et al.*, 1995), and evanescent field sensors (Weissman, 1997).

The fact that this approach requires only one step, rather than two, to produce a guided, periodically-poled structure has both advantages and disadvantages. Requiring only a single step reduces the complexity of the fabrication process, and eliminates the need to align the waveguides to the periodic domain grating. However, it also removes the possibility of optimizing the poling process and waveguide properties independently.

In both these approaches, reproducibility has been a persistent problem. Typically, tens or hundreds of individual waveguides are fabricated on a single substrate, and it has not been unusual for only a few to have very high conversion efficiencies. The best results obtained on different wafers may also vary greatly. Finding recipes that consistently give good results has been one of the main challenges limiting practical development of waveguide-based blue-green light sources.

We may also state some broad differences between the three materials that have been used. LN and LT have a longer history of development than does KTP. LN, in particular, has been extensively developed for surface-acoustic-wave device applications (such as intermediate-frequency filters for television sets), and for use in (non-SHG) integrated-optic devices (such as modulators for telecommunications). That factor, coupled with inherent differences in the ways these materials are produced, has led to the availability of larger wafer sizes for LN and LT than for KTP. Use of 3-in diameter LN wafers has become standard, while KTP substrates are often 1–2 cm squares. The consistency of KTP wafers, both from sample to sample and from place to place within an individual sample, tends to be poorer than in LN. LN is also less expensive than KTP. However, it has proven to be somewhat more difficult to produce the short periods necessary for QPM SHG of blue-green wavelengths in LN than in KTP, and photorefraction has been a serious problem in LN waveguides, while it is usually not significant in KTP waveguides (Kondo *et al.*, 1994, Eger *et al.*, 1997).

Despite these difficulties, and although reproducibility is often unclear, several groups have demonstrated high nonlinear conversion efficiencies in waveguides. A common way of reporting the efficiency is based on the low-loss version of Equation (6.1), which is:

$$P_3 = \frac{8\pi^2 d_{\text{eff}}^2 \mathfrak{J}_{\text{ov}}^2}{\lambda_1^2 \epsilon_0 c N_{\text{eff}}^3} P_1^2 l^2 \quad (6.2)$$

This expression can be written as:

$$\eta_{\text{norm}} = \frac{P_3/P_1}{P_1 l^2} = \frac{8\pi^2 d_{\text{eff}}^2 \mathfrak{J}_{\text{ov}}^2}{\lambda_1^2 \epsilon_0 c N_{\text{eff}}^3} \quad (6.3)$$

The rightmost term contains only parameters that are fixed for a waveguide constructed of particular materials, in a particular way, and with a particular geometry. The middle term contains the usual conversion efficiency P_3/P_1 normalized by two parameters that we might wish to think about changing in order to increase the efficiency obtained from a waveguide whose other properties are fixed: the infrared power in the guide, and the length of the guide. Thus, this quantity is a *normalized* conversion efficiency, expressed as %/W-cm², which allows us to see how the second-harmonic power generated by a guide of fixed characteristics would scale if we could couple in more power or if we could increase the length.

A few brief caveats are in order. Normalized efficiencies reported in the literature (and repeated in Table 6.1) often do not state clearly the characteristics of the laser used to measure the efficiency. As we saw in Section 2.2.6, if the infrared source oscillates in multiple longitudinal modes, the measured conversion efficiency can be as much as a factor of 2 higher than when a single-longitudinal mode source is used. In addition, certain waveguide geometries give an overlap integral that is larger for excitation of a higher-order mode (such as TM₀₁) than it is for the lowest-order TM₀₀ mode. Thus, the highest conversion efficiency reported for a particular guide may occur for excitation of a mode that is not suitable for some applications. Also, reports in the literature do not always clearly indicate whether what is being reported is the *external* conversion efficiency, based on the power emerging from the waveguide, or the *internal* efficiency, in which the power present within the waveguide is estimated by taking into account Fresnel reflections, coupling losses, etc.

As an example of the use of the normalized conversion efficiency, suppose that we wish to generate 10 mW of blue light with no more than 100 mW of infrared present in the waveguide, and that we wish the length of our guide to be 10 mm. In order to accomplish this, we would need a normalized conversion efficiency

Table 6.1. Selected results for single-pass frequency doubling in waveguides

Reference	Material	λ (nm)	Δ (nm)	η_{norm} (%/W-cm ²)	P_3 (mW)	P_1 (mW)	I (mm)	Comments
Yamamoto and Mizuuchi (1992)	PPLT	436.5	4.0	157	23	121	10	
Bierlein (1992)	PPKTP	425	4.0	400				
Jongerius <i>et al.</i> (1992)	PPKTP	460	5	210	6	107	5	
Yamamoto <i>et al.</i> (1993)	PPLT	435	4.0	230	31	145	10	
Yamada <i>et al.</i> (1993)	PPLN	425.9	2.8	600	20.7	195.9	3	Two-step, e-field poling + APE
Mizuuchi and Yamamoto (1994)	PPLT	434	4.0	153	22	120	10	Two-step, PE + heat treatment
Eger <i>et al.</i> (1994)	PPKTP (F)	425	4.0	800	3	51	3.8	One-step, segmented guide, Rb/Ba exchange
Doumuki <i>et al.</i> (1994)	Ta ₂ O ₅ /KTP	415	N/A	210*	28	332	4.5	Low-angle Čerenkov. *Units are therefore %/W-cm
Bortz <i>et al.</i> (1994)	PPLN	488	5.0	204				Excitation of TM ₀₁ mode; two-step, Ti-indiffusion poling
Chen and Risk (1996)	PPKTP (HT)	431	4.0	720	12	146	2.8	Two-step, e-field poling + Rb/Ba exchange
Chen and Risk (1996)	PPKTP (HT)	431	4.0	1100	9	120	2.4	
Risk and Loiacono (1996)	PPRTA	437	3.9	225	0.0087	21.1	0.93	Two-step, e-field poling + Cs exchange
Bierlein <i>et al.</i> (1997)	PPKTP	490		220	107	315	7	One-step, segmented guide
Eger <i>et al.</i> (1997a)	PPKTP (F)	495	4.0	185			9	Two-step, e-field poling + Rb exchange
Webjörn <i>et al.</i> (1997)	PPLN	421.5		170	25	120	10	Two-step, Ti-indiffusion poling + APE
Webjörn <i>et al.</i> (1997)	PPLN	551		300				Two-step, e-field poling + ?
Webjörn <i>et al.</i> (1997)	PPKTP	425	4.0	500	0.8	15.8	8	One-step, Rb/Ba exchange
Brinkmann <i>et al.</i> (1997)	PPLN	484	4.5	208	5	62	7.9	Two-step, e-field poling + APE
Sonoda <i>et al.</i> (1997)	PPLN	475	4.75	323	37	107	10	Polarization-axis-inclined substrate
Mizuuchi <i>et al.</i> (1997a)	PPLN	434	3.4	1200	5.5	42	5	x-cut substrate, coupling to TE ₁₀ mode
Pliska <i>et al.</i> (1998)	KN	438	N/A	22.7	14	340	7.3	Birefringence PM, ridge waveguide
Webjörn <i>et al.</i> (1998)	PPLN	“Green”			590		9	Planar waveguide, unpolarized fiber laser source
Sugita <i>et al.</i> (2000)	PPLN	426	3.2	572	17.3	55	10	Off-axis MgO:LN
Raifailov <i>et al.</i> (2001)	PPKTP	486	N/A	36.2	7.5	144	10	Gain-switched InGaAs/GaAs laser diode source

of $100\%/W\text{-cm}^2$. Table 6.1 summarizes work in fabricating waveguide frequency doublers, and shows that several systems can produce efficiencies in excess of this value. The same table also illustrates that not only high efficiency, but also high absolute blue power has been achieved in some cases.

It should also be mentioned that for a waveguide with a certain refractive index profile and spatial distribution of nonlinearity, there is a particular geometry that will maximize the conversion efficiency. For example, Ramanujam and Burke (1997) have analyzed the optimum structures for PPKTP and PPLT waveguides and Bortz *et al.* (1994) demonstrated the benefits of using a geometry that provides dimensionally-noncritical phasematching. However, modeling solutions for optimum designs are only as good as the information they draw upon, such as refractive index profiles and spatial variation of the nonlinear coefficient. Determination of these parameters with the precision required for useful modeling has really only been done in the case of LN (Bortz *et al.*, 1993). Furthermore, once an optimum design is identified, we must know how to produce that design, which requires a detailed knowledge of how changes in the fabrication process alter the corresponding waveguide parameters. With the exceptions noted above, addressing the difficulties associated with consistently producing frequency-doubling waveguides with reasonably high efficiency has taken precedence over trying to achieve designs that are truly optimum, as has trying to integrate working SHG waveguides with diode lasers in a practical way, which is the subject of the next section.

6.3 INTEGRATION ISSUES

One concept for a waveguide-based blue-green light source that has attracted much interest and inspired much effort is shown in Figure 6.3. This depiction omits a number of important details, but shows the basic concept for such a device. The light from an infrared diode laser is proximity-coupled (or “butt-coupled”, or “close-coupled” (Hunsperger *et al.*, 1977)) into a frequency-doubling waveguide, and the two elements are affixed to a common “submount”, ideally using some sort of micromechanical features to automatically bring the two into alignment (for example, see Wale and Edge (1990)). Such a device would, in principle, be very compact and rugged.

Realization of such a device involves solving a number of technical problems. One that we have previously discussed in another context is that of optical feedback and its destabilizing effect on the diode laser. Another is spatial mode matching between the output beam of the diode laser and the waveguide mode. Another is ensuring that the diode laser is tuned to the phasematching wavelength and held there. Another, perhaps less obvious than the preceding ones, involves polarization

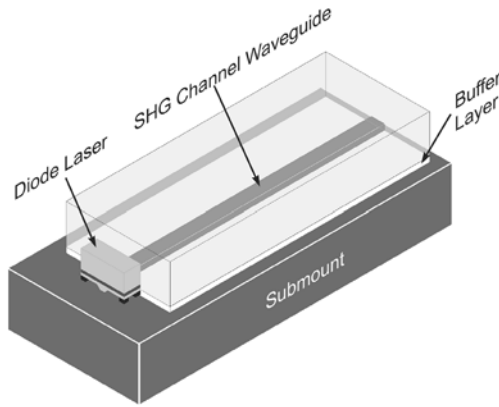


Figure 6.3: Depiction of a blue-green light source based upon a diode laser and SHG waveguide integrated onto a common submount. In this conceptual representation (the details of which should not be taken too seriously), a ridge-type laser is mounted ridge down for more efficient extraction of heat. The SHG waveguide is similarly mounted top down, and a buffer layer is used to protect the surface of the waveguide and reduce loss resulting from mounting.

compatibility – typical diode lasers emit light polarized parallel to the submount in Figure 6.3, whereas typical periodically-poled waveguides would require light polarized perpendicular to the submount.

6.3.1 Feedback and frequency stability

The device depicted in Figure 6.3 uses a diode laser with a facet reflectivity of a few percent in close proximity to the flat, highly polished end of a material with refractive index ~ 2 , and therefore a reflectivity of $\sim 10\%$. Thus, the reflection from the laser diode's own facet will compete for control of the laser with the reflection from the end of the waveguide, which – as we have previously seen – almost guarantees amplitude and/or frequency instability. For example, Sidorin and Howe (1997) showed that the wavelength of a diode laser proximity-coupled to a single-mode fiber could be tuned over ~ 14 nm by changing the spacing between the diode laser facet and the fiber end by only ~ 0.5 μm . The reflection from the endface of the waveguide can be suppressed by anti-reflection coating, or by angling the end of the guide (Kincaid, 1988), although the latter is not generally compatible with the coplanar mounting arrangement of Figure 6.3. Anti-reflection coating can substantially reduce the reflectivity of the waveguide facet; however, even a weak reflection from the endface can lead to instabilities.

An alternative approach involves suppressing the reflection from the waveguide endface as much as possible using an anti-reflection coating, and providing strong,

narrowband frequency-selective feedback to the diode laser from some sort of grating in order to fix its emission wavelength. The stability of this approach can be further improved by also applying an anti-reflection coating to the diode laser facet in order to suppress that reflection as well.

This grating-stabilization approach has been implemented in four ways:

- using a bulk-optic grating;
- using a grating integrated with the waveguide doubling element;
- using a fiber grating inserted between the diode laser and the waveguide doubling element;
- integrating the grating with the diode laser.

6.3.1.1 Bulk-optic grating

A conventional bulk-optic grating can be used to provide the frequency-selective feedback necessary to force stable operation of the laser diode at the phasematching wavelength. Some examples of how this has been implemented are shown in Figure 6.4. Yamamoto *et al.* (1993) used a grating in the “Littrow” mode to retroreflect some of the infrared light emerging from their LT SHG waveguide back to the diode laser (Figure 6.4(a)). Laurell (1993) used a similar configuration with a KTP waveguide, and measured the amplitude fluctuation of the blue output to be $\sim 0.3\%$. In both these cases, a diode laser with a standard facet reflectivity of $\sim 5\%$ was used. Risk *et al.* (1993) used an anti-reflection-coated diode laser with a facet reflectivity of $\sim 0.1\%$ to further improve the stability and tunability of such a device (Figure 6.4(b)). They observed the blue output power to be free of any large amplitude fluctuations and measured the relative intensity noise at 5 MHz to be -110 dBc/Hz, which indicates the efficacy of this “extended cavity” approach in suppressing instabilities due to unwanted optical feedback. In a related approach, Kitaoka *et al.* (1994) used a narrowband transmission filter placed between the laser diode and the waveguide to stabilize the wavelength of the diode laser (Figure 6.4(c)).

6.3.1.2 Integrated grating

Instead of using an external, bulk-optic grating as do the configurations shown in Figure 6.4, the frequency-selective reflection can be produced by a guided-wave distributed Bragg reflector fabricated on the same substrate as the doubling element. The Bragg reflection can be produced by a separate grating structure or, under certain circumstances, can arise from the same periodic structure used for QPM. The first experimental demonstrations of this approach used this latter configuration and exploited the observation that the processes used to produce periodic domain

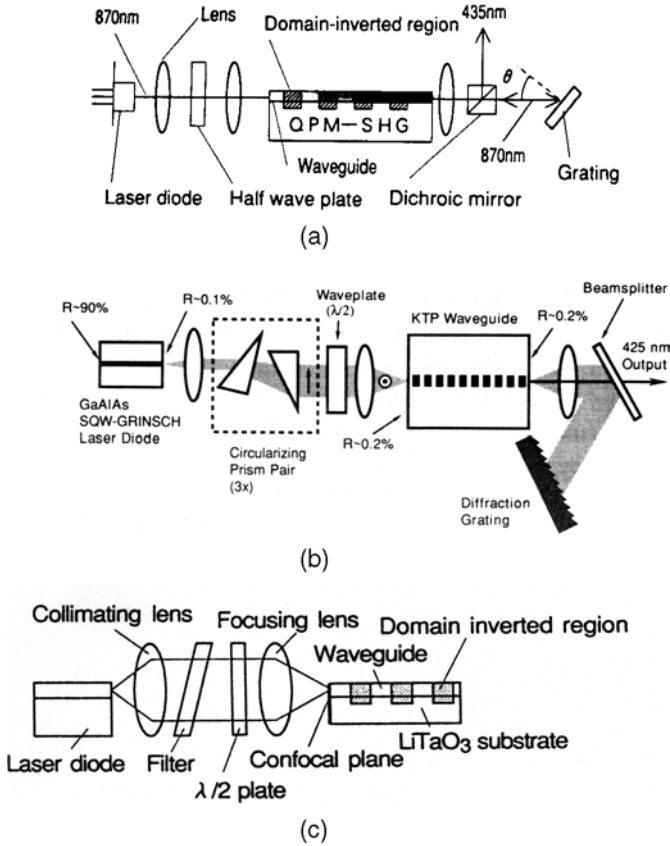


Figure 6.4: Three configurations used for waveguide SHG of diode lasers, using frequency-selective feedback to lock the diode laser wavelength: (a) configuration used by Yamamoto *et al.* (1993); (b) configuration used by Risk *et al.* (1993); (c) configuration used by Kitaoka *et al.* (1994). ((a) Reprinted with permission from Yamamoto *et al.* (1993); (b) reprinted with permission from Risk *et al.* (1993); (c) reprinted with permission from Kitaoka *et al.* (1994).)

inversion in the waveguide also induced a periodic change in the linear properties of the waveguide. Shinozaki *et al.* (1991a) used LN devices, in which domain inversion was produced by titanium indiffusion and the waveguide was formed by subsequent annealed proton exchange. The refractive index was slightly increased in the domain-inverted regions due to incorporation of titanium. Roelofs *et al.* (1992) used segmented KTP waveguides, in which the index difference was somewhat larger.

In both these cases, a single structure provided both the periodic domain inversion necessary for QPM and the periodic modulation of the refractive index necessary to produce distributed Bragg reflection. If the structure is designed so that Bragg reflection and QPM occur at the same wavelength, then feedback from the DBR can

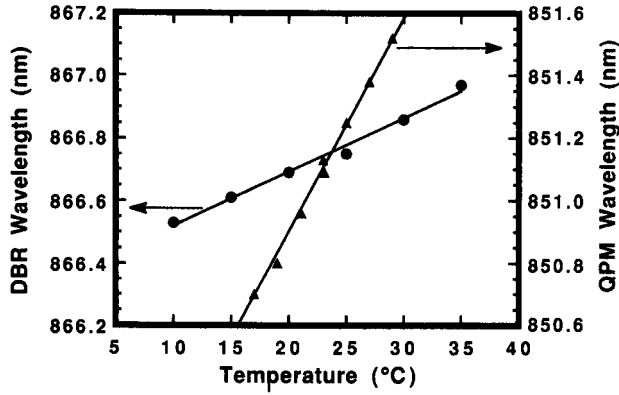


Figure 6.5: Variation of DBR wavelength and QPM wavelength with temperature for a segmented KTP waveguide (4 μm period). (Reprinted with permission from Risk and Lau (1993).)

be used to force the diode laser to operate at the correct wavelength for QPM; hence, this approach has been dubbed “self-QPM” or “automatic QPM”. The wavelength at which both conditions are satisfied is given by:

$$\lambda_{QPM} = 2m\Delta N_{eff}\Lambda = \lambda_{DBR} = \frac{2N_{eff,\lambda_1}\Lambda}{p} \quad (6.4)$$

where Λ is the period of the structure, m is an integer specifying the order of QPM, p is an integer specifying the order of distributed Bragg reflection, N_{eff,λ_1} is the effective index for the guided mode at the fundamental wavelength λ_1 and $\Delta N_{eff} = N_{eff,\lambda_3} - 2N_{eff,\lambda_1}$. In a typical configuration, the period is chosen to give first-order ($m = 1$) QPM, and this period results in a Bragg reflection of high order – for example, the waveguide used by Shinozaki and colleagues employed the 43rd ($p = 43$) Bragg order. Since λ_{QPM} and λ_{DBR} vary with temperature at substantially different rates, the temperature of the waveguide can be adjusted to bring the two into coincidence (Figure 6.5).

One problem with the self-QPM approach is evident from Figure 6.6, where the two constraints given above are displayed graphically for segmented KTP waveguides. This figure shows there are particular combinations of λ_1 and Λ which satisfy both conditions; thus, it is not always possible to satisfy Equation (6.4) for an arbitrary wavelength or period when a perfectly periodic structure is used. We could describe such a structure by writing out the sequence ABABABABABAB... where A might represent an ion-exchanged segment of length 2.0 μm and B might represent an unexchanged segment with length 2.0 μm . Thus, in this structure, the basic pattern AB is repeated with a period of 4.0 μm . In practice, it is often the case that the length of these segments, and hence the period of the structure, can

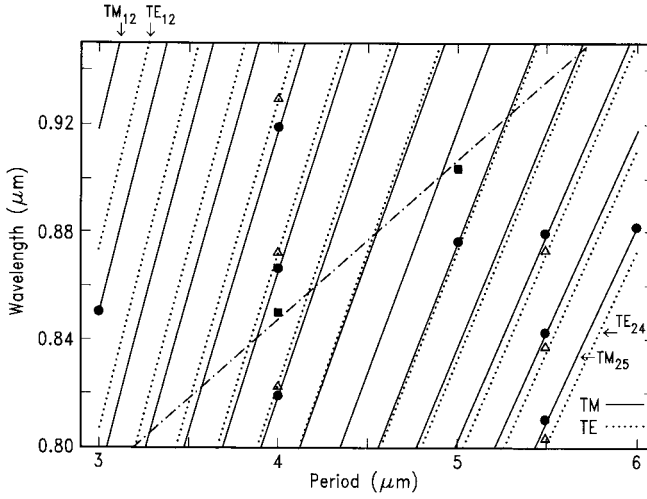


Figure 6.6: Calculated variation of DBR wavelength with period Λ for a segmented KTP waveguide. DBR wavelengths for both TE and TM modes are shown, data points are measured DBR wavelengths. Also shown (by the dot-dash line) is the variation of the QPM wavelength with period. (Reprinted with permission from Risk and Lau (1993).)

be changed only by discrete amounts – $0.1 \mu\text{m}$ is typical – due to limitations of the mask-making apparatus. Roelofs *et al.* (1992) suggested that the design constraints imposed by the perfect periodicity of such patterns could be eased by using a longer basic sequence repeated with a “superperiod” Λ_{sp} . For instance, consider the pattern ABACABACACACABACABACACACABACABAC, where A and B are as before, but C represents an unexchanged segment of length $1.9 \mu\text{m}$. This pattern has a total length of $63 \mu\text{m}$ (hence it would be repeated with $\Lambda_{sp} = 63 \mu\text{m}$) and, 16 pairs of alternating exchanged/unexchanged segments. Thus, the average period is $63/16 \mu\text{m} = 3.9375 \mu\text{m}$. If we replace the last C in this pattern with a B, the total length would be $63.1 \mu\text{m}$, and the average period would be $3.94375 \mu\text{m}$. Thus, the average period, and hence the QPM wavelength, can be tuned in very small increments ($0.00625 \mu\text{m}$) using this approach. From the point of view of Bragg reflections, we can regard this structure as having a periodicity determined by Λ_{sp} , with the reflectivity of each period determined by the particular sequence used. From Equation (6.4), in order to produce a reflection at a given wavelength, a much higher order (p) must be used. The spacing in wavelength between adjacent orders of a distributed Bragg reflector is given by

$$\Delta\lambda_{DBR} = \frac{2N_{eff,\lambda_1}\Lambda}{p(p+1)} \approx \frac{2N_{eff,\lambda_1}\Lambda}{p^2} = \frac{\lambda_{DBR}}{p} \quad (6.5)$$

Thus, when this approach is used, the larger Bragg order corresponding to the superperiod Λ_{sp} ($p \sim 200$) yields Bragg reflections that are much more closely

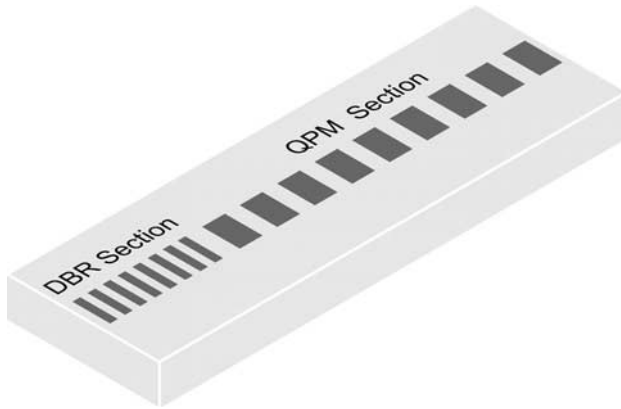


Figure 6.7: Geometry for a device with separate DBR and QPM sections, using segmented KTP waveguides.

spaced in wavelength than when a lower-order Bragg reflection corresponding to the QPM period Λ ($p \sim 20$) is used. The reduced spacing of the Bragg orders makes it easier to design a structure that can simultaneously provide both DBR and QPM functions at a particular wavelength.

However, using the same structure for both QPM and DBR makes optimization of the device difficult. In addition, the reflectivity of the Bragg grating must be limited so that it not “steal” too much power from the nonlinear interaction (Weissman *et al.*, 1995a). Thus, an alternative approach to this problem is to create two physically distinct and separate sections of the waveguide, one which provides quasi-phasematched frequency-doubling and another which acts as a DBR (Figure 6.7). One implementation of this method uses high-order Bragg reflectors having a period comparable to the QPM section (Bierlein *et al.*, 1994, Eger *et al.*, 1995). Eger and coworkers were able to generate 3.6 mW of 429-nm blue light using this approach to lock the frequency of a diode laser.

However, if a separate grating is to be used for Bragg reflection, there are some advantages in making it a short-period, low-order grating. A low-order DBR can achieve a desired reflectivity with a shorter length than can a higher-order one; thus, more of the valuable “real estate” of the waveguide chip can be devoted to frequency-doubling. This can be particularly beneficial if waveguide resonator techniques are used to increase the efficiency of the interaction (Section 6.3.5). In addition, the separation between Bragg reflections for TE and TM waveguide modes is larger with low-order DBRs, and this effect can be used to force the diode laser to oscillate in a TM – rather than TE – mode, which is advantageous for polarization compatibility (Section 6.3.2). Risk *et al.* (1994) used the highly anisotropic diffusion characteristics of KTP to produce third-order Bragg gratings

($\Lambda_{DBR} = 0.7 \mu\text{m}$, $p = 3$) through the same ion-exchange process that is used to produce segmented waveguides for QPM. They found that the reflectivity approached 100% for gratings as short as 0.56 mm, and that the feedback from a grating only 70 μm long was sufficient to produce stable frequency-locking of an anti-reflection-coated diode laser used in an extended-cavity configuration. The short length of this third-order DBR should be compared with the 4-mm length of the sixteenth-order grating used by Eger and colleagues. Raifailov *et al.* (2001) generated 7.5 mW at 486 nm using a PPKTP waveguide with separate SHG and DBR gratings.

6.3.1.3 Fiber gratings

Another variation on the use of a grating for frequency locking the diode laser employs a fiber grating placed between the diode laser and the frequency-doubling waveguide, as suggested by Lim (1996). This approach exploits the commercial development of high-quality DBRs produced directly in single-mode optical fiber (Hill *et al.*, 1993), so that fabrication of a separate Bragg reflector on the SHG waveguide substrate is not necessary. The Bragg reflection from the fiber grating stabilizes the frequency of the diode, as has been demonstrated by Ventrudo *et al.* (1994). Laurell (1997) implemented this approach and achieved 4.8 mW of 489.6-nm light using a KTP waveguide for frequency-doubling.

6.3.1.4 DBR lasers

Another way to use an integrated grating for frequency locking involves putting the grating on the laser diode chip rather than on the SHG waveguide chip. We described such “DBR diode lasers” in Chapter 3. The development of high-power (>270 mW), long-lived ($>20\,000$ h), single-mode DBR lasers operating at ~ 860 nm has made these devices attractive for use in waveguide frequency-doubling (Major *et al.*, 1994, Gulgazov *et al.*, 1997). The emission wavelength of such a DBR laser can be tuned by injecting current into the region of the grating in order to heat it. Gulgazov and coworkers reported that they could tune the wavelength of an AlGaAs DBR laser between 854 nm and 864 nm using this approach. This electronic means of tuning the laser wavelength provides a very convenient way of achieving phasematching. Kitaoka *et al.* (1997) used such a laser in conjunction with a LN waveguide to generate a peak power of 17 mW (modulated at 20 MHz with a duty cycle of $\sim 25\%$).

6.3.2 Polarization compatibility

In the configuration depicted in Figure 6.3, there is an incompatibility between the polarization emitted by typical diode lasers and that required by typical QPM

waveguide doublers. Typical diode lasers oscillate with the electric field vector parallel to the plane of the p – n junction; that is, the diode laser prefers to oscillate in a TE mode of its internal waveguide. In contrast, typical periodically-poled waveguides are designed to exploit the large d_{33} nonlinear coefficient, which requires that the input optical field be a TM mode, polarized perpendicular to the thin dimension of the substrate. Three basic approaches have been suggested for overcoming this problem:

- designing a laser diode that intrinsically prefers to oscillate in a TM mode;
- forcing a normally TE-mode laser diode to oscillate in a TM mode;
- creating an efficient doubler that works with a TE mode instead of a TM mode.

6.3.2.1 TM-mode laser diodes

The reason that laser diodes normally tend to oscillate in a TE mode – that is, with polarization parallel to the plane of the junction – is that the reflectivity of the abrupt interface between the cleaved end of the waveguide and air is slightly higher for TE modes than for TM modes (Ikegami, 1972). However, the presence of tensile stress in the active layer can cause a diode laser to oscillate instead in a TM mode (Chong and Fonstad, 1989). Although TM-mode lasing due to tensile stress has been observed in 868-nm GaAlAs lasers grown on Si substrates (Sakai *et al.*, 1987) and in red 637-nm GaInP lasers (Welch *et al.*, 1991), such devices have not yet been further developed to provide the high-power operation in a single spatial and spectral mode required for efficient waveguide frequency-doubling.

6.3.2.2 Forced TM-mode oscillation

Although a standard diode laser tends to oscillate in a TE mode, Mitsuhashi (1982) showed that by placing a polarizer oriented to transmit the TM mode inside an extended-cavity laser, the diode laser element could be forced to oscillate with TM polarization instead. Welch and Waarts (1993) suggested using this approach in conjunction with frequency-doubling waveguides in order to solve the problem of polarization incompatibility. Risk *et al.* (1993) demonstrated this concept using the arrangement in Figure 6.8, in which a polarizing beamsplitter was inserted into the extended-cavity in order to force TM-mode oscillation. The threshold current required in the forced TM-mode configuration was 49 mA, compared with 28 mA for TE mode operation without the polarizer. With forced TM-mode oscillation, they achieved 0.42 mW of blue 425-nm light, compared with 1.2 mW achieved using the standard configuration.

For an integrated device like the one in Figure 6.3, a bulk-optic polarizer cannot be used, and several approaches have been tried to achieve forced TM-mode operation

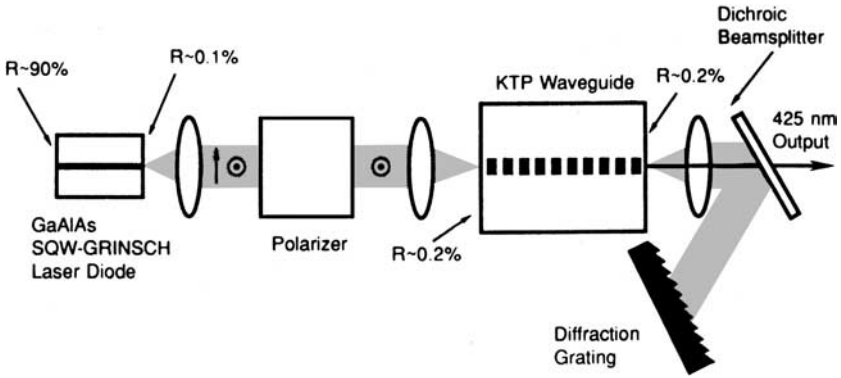


Figure 6.8: Configuration used by Risk and coworkers for forced TM-mode operation of an extended-cavity laser with waveguide SHG element. (Reprinted with permission from Risk *et al.* (1993).)

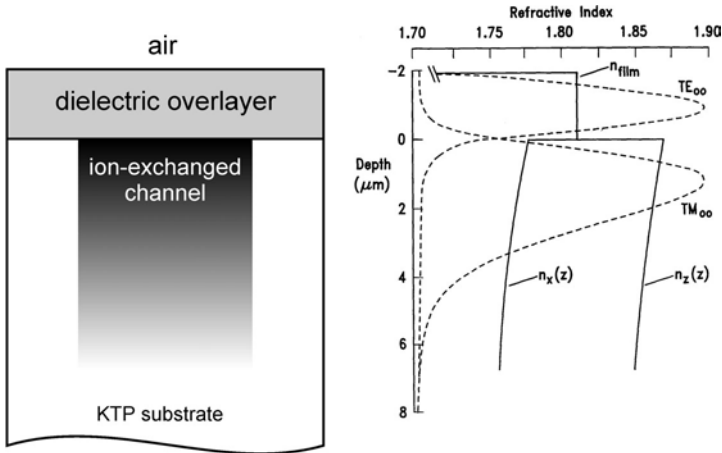


Figure 6.9: Method for producing polarization discrimination in a KTP waveguide using a dielectric overlayer of appropriate refractive index. (Reprinted with permission from Risk *et al.* (1993a).)

using all-integrated components. The most straightforward extension of the bulk-optic implementation involves creating a guided-wave polarizer. Risk *et al.* (1993a) demonstrated a guided-wave implementation of this approach using a dielectric overlayer deposited atop KTP waveguides. Waveguides made by the standard ion-exchange process in mixed melts of RbNO_3 and $\text{Ba}(\text{NO}_3)_2$ guide both TE and TM polarizations, but there is a rather large difference in the effective indices for TE and TM modes, due mostly to the birefringence of the KTP substrate (Figure 6.9). The dielectric overlayer has a refractive index higher than the effective index for TE modes, but lower than the effective index for TM modes. Thus, while the TM

modes remain confined to the channel waveguide, the TE modes are trapped in the overlayer, and experience a weak lateral confinement due to strip-loading by the ion-exchanged channel. When such a composite waveguide structure is placed inside an extended cavity laser, several factors help reduce the amount of TE light that can return to the laser relative to TM:

- Since the TE modes and TM modes are spatially separated (see Figure 6.9), mode matching of the diode laser into the waveguide can preferentially excite TM modes.
- Light that is excited in the TE_{00} mode and subsequently reflected back in the direction of the diode laser is not mode matched to the diode laser beam.
- Higher-order TE modes which overlap the TM_{00} mode may be excited by an input beam that is mode matched to TM_{00} , but these modes are only weakly confined laterally and experience loss through scattering and diffraction.
- The preferential loss for TE modes may be further increased by depositing a metal film atop the overlayer, or by roughening its surface.

Risk and coworkers used such a waveguide in an extended-cavity configuration and found that when they used guides without an overlayer, oscillation was always in a TE mode, while oscillation was always in a TM mode when overlaid guides were used. Thus, this approach to integrating a polarizing element with a doubling waveguide provided sufficient polarization discrimination to force TM-mode oscillation.

In proton-exchanged LN and LT waveguides, only TM modes are guided to begin with, so that the waveguides themselves provide discrimination against the unwanted TE modes without further modification. Chwalek (1995) used this approach to force TM oscillation of a GaAlAs diode laser. He recognized that some amount of TE feedback to the laser is inevitable, as a result of imperfect polarization discrimination and unwanted reflections and scattering, so he also investigated the conditions required to achieve forced TM-mode operation in the presence of various levels of feedback.

In another approach, Risk *et al.* (1994) noted that the separation in wavelength between TM and TE Bragg reflections could be used to force TM-mode oscillation (Figure 6.10). They designed a third-order DBR in a KTP waveguide such that only the TM Bragg reflection fell within the gain bandwidth of the laser; thus, when this device was used as the reflecting element in an extended-cavity laser, the diode was forced to oscillate preferentially in a TM mode.

6.3.2.3 TE-mode doublers

In order to produce inversion of the ferroelectric domains in a material like LN or KTP, it is necessary to apply an electric field along the polar axis, which is

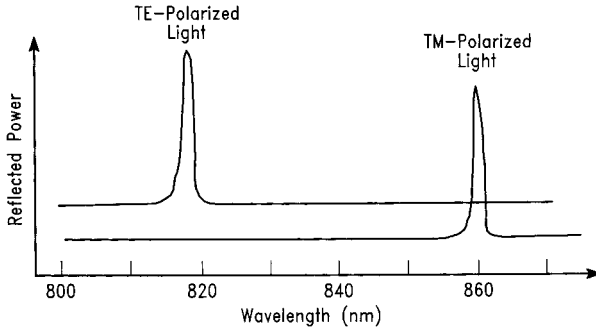


Figure 6.10: Separation in wavelength of TM-mode and TE-mode reflections for the third-order DBR made by Risk and coworkers. (Reprinted with permission from Risk *et al.* (1994).)

usually designated as the z -axis. As we have seen, this electric field can arise from an external voltage applied to the crystal, or it can develop internally through thermoelectric gradients which result from an ion-exchange process. In either case, the most convenient geometry for the crystal is that of a thin plate or wafer, with the z -axis along the thin direction. In particular, when this geometry is used for electric field poling in combination with electrodes applied to the top and bottom surface of the wafer, the electric field is developed across the thickness of the wafer, and thus along the z -axis as required. Making the crystal thin reduces the voltage required to achieve a particular field strength within the crystal.

In using a periodically-poled z -cut wafer for SHG, it has been customary to polarize the infrared light parallel to the z -axis in order to exploit the d_{33} nonlinear coefficient, which is the largest component of the d tensor in LN, LT, and KTP. When this light is confined to a waveguide in the material, this polarization requirement means that a TM mode must be used. However, as we have seen, this geometry leads to a polarization compatibility problem when such a doubling element is used with a standard diode laser, which also has a wafer-like geometry, but emits light having a TE polarization with respect to that wafer.

An alternative to forcing the laser to oscillate in a TM mode is to create a doubling element which uses a TE mode, but still takes advantage of the large d_{33} coefficient made available through QPM. One way to do this is to create periodic poling in an x - or y -cut substrate. Mizuuchi *et al.* (1997) demonstrated one implementation of this approach in using the configuration shown in Figure 6.11 to produce periodic domain inversion in y -cut LN. Voltage V_2 , applied between the two electrodes on the top of the wafer, produces an electric field predominantly along the z -axis, and the crenelated shape of the grounded electrode introduces a periodic modulation

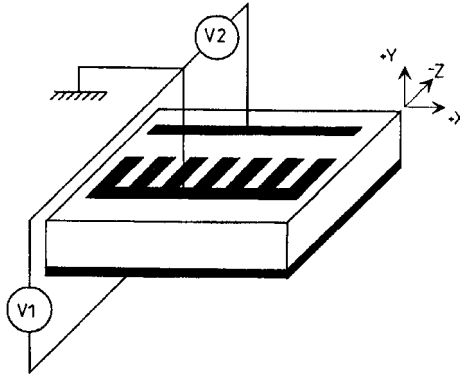


Figure 6.11: Arrangement used by Mizuuchi and coworkers for periodic poling of y-cut MgO:LiNbO₃. (Reprinted with permission from Mizuuchi *et al.* (1997).)

of the electric field in the x -direction, appropriate for producing periodic domain inversion suitable for QPM along that direction. The voltage V1, applied between the crenelated electrode and the uniform electrode on the bottom of the wafer, controls the depth of the domain inversion. Mizuuchi and colleagues were able to produce domain inversion with a $3.4\text{-}\mu\text{m}$ period over a 10-mm length, as deep as $\sim 1.3\text{ }\mu\text{m}$. Using annealed proton exchange, they produced a channel waveguide supporting a single TE mode at the fundamental wavelength. They also applied a thin high-index Nb₂O₅ layer atop the MgO:LN substrate, which they found improved the confinement and overlap between fundamental and second-harmonic modes (although the best overlap is then between the TE₀₀ mode at 868 nm and the TE₁₀ mode at 434 nm (Mizuuchi *et al.*, 1997a)). Using a Ti:S laser for characterization, they obtained 19 mW of 434-nm blue light with 56 mW present in the waveguide. Kitaoka *et al.* (1997) used such a waveguide in combination with a tunable DBR laser to generate blue light with a peak power of 17 mW (modulated at 20 MHz with a duty cycle $\sim 25\%$).

Instead of using a second voltage applied across the thickness of the wafer to increase the depth of domain inversion, as Mizuuchi and coworkers did, it is possible to use a substrate which is slightly angled with respect to the x - or y -axis, as shown in Figure 6.12. In this configuration, as the inverted domain propagates along the z -axis, it also grows deeper with respect to the top surface of the wafer. Sonoda *et al.* (1997) used this approach to demonstrate efficient TE-mode doubling in MgO:LN substrates. Using an inclined substrate with $\theta \sim 3^\circ$, they produced domain inversion to a depth ~ 2.5 times that obtained using a substrate with $\theta = 0^\circ$. Using a tunable extended cavity diode laser (Mehuys *et al.*, 1993) as the infrared source, they produced a blue power at 475 nm of 37 mW .

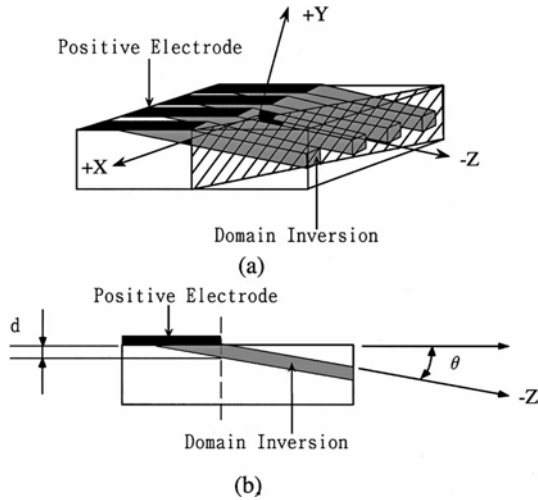


Figure 6.12: Configuration used by Sonoda and colleagues for periodic poling of polarization axis inclined lithium niobate. (Reprinted with permission from Sonoda *et al.* (1997).)

6.3.2.4 Integrated half-wave plate

Still another approach to the polarization incompatibility problem uses an integrated half-wave plate to rotate the polarization of the TE-mode light emitted by the diode laser to the TM-mode orientation required by a SHG waveguide with the standard QPM configuration. One way to achieve a compact half-wave plate is to deposit a thin film of a metal oxide at an oblique angle (Motohiro and Taga, 1989). Kitaoka *et al.* (1996) implemented this idea using a film of Ta_2O_5 deposited obliquely on the end of a LT substrate, which contained waveguides fabricated by proton exchange.

6.3.3 Coupling

In most of the blue-light generation experiments done so far, the light emerging from the diode laser is collimated and then refocused into the SHG waveguide (perhaps with some additional beam-shaping optics in between the two). In order to realize the compact device shown in Figure 6.3, it is desirable to eliminate any intermediate optics between the laser diode and waveguide. Several analyses of proximity coupling between a diode laser and waveguide have appeared. Lee and Wang (1992) developed models that reflected the non-gaussian nature of typical laser diode and waveguide modes. More recently, Karioja and Howe (1996) have considered coupling between a laser diode and an SHG waveguide. Their analysis included the étalon effects and investigated the transverse, longitudinal, and angular alignment tolerances. Sidorin and Howe (1997a) have considered wavelength-pulling effects and optical noise associated with close coupling between a diode laser and waveguide.

Perhaps the best indication of how useful proximity coupling can be in practice is given by the experimental results of Kitaoka *et al.* (1996), who compared lens coupling with proximity coupling. Using one lens to collimate the output of an 820-nm GaAlAs diode laser and a second lens to focus the infrared light into an anti-reflection-coated, proton-exchanged LT waveguide on a *z*-cut substrate, they achieved about 72% coupling efficiency. When the diode laser was proximity coupled to the waveguide, they were able to achieve a maximum coupling of about 60%. Using a waveguide made on an *x*-cut substrate, they were able to achieve 73%, comparable to the best they obtained with lens coupling.

Webjörn *et al.* (1997) reported butt coupling a DBR laser diode to a QPM waveguide and packaging the entire assembly in a compact enclosure, having dimensions of 21 mm × 13 mm × 9 mm. This device generated 4 mW of blue (~425 nm) light with low noise (−140 dB/Hz, dc to MHz), with a TM₀₀ beam having a $M^2 \sim 1.4$ in the direction perpendicular to the waveguide substrate and ~1.2 in the direction parallel. They overcame the polarization compatibility problem by physically rotating the waveguide by 90° relative to the diode laser.

More recently, Kitaoka *et al.* (2000) reported a 2-mW blue laser in a configuration closely resembling Figure 6.3 and in a package measuring only 5 mm × 12 mm × 1.5 mm. They used a DBR laser and QPM chip affixed to the same silicon submount. The QPM chip was fabricated from *x*-cut MgO:LN in order to overcome the polarization problem, as described above. Infrared light emitted by the DBR laser was butt coupled into the waveguide with an efficiency of ~75%.

6.3.4 Control of the phasematching condition

Assuming that we have developed a fabrication process that repeatably gives high-conversion-efficiency waveguides, assuming that we have employed some means for ensuring frequency stability of the diode laser, and assuming that we have settled on some solution to the polarization compatibility problem, there still remains the problem of adjusting the laser frequency to match the phasematching wavelength, or vice-versa. Ideally, this adjustment should take place automatically. One way to adjust the phasematching wavelength is to apply an electric field to the material in order to alter the refractive indices through the electro-optic effect. Uesugi *et al.* (1979) demonstrated this effect using Type-II, birefringently-phasematched SHG in titanium-indiffused LN waveguides, using coplanar strip electrodes to concentrate the electric field in the vicinity of the waveguide. Helmfrid and colleagues (1993) later analyzed a similar approach applied to periodically-poled waveguides. Bortz *et al.* (1996) described the system shown in Figure 6.13, in which the phasematching wavelength is slightly dithered through the electro-optic effect, by applying a small-amplitude (5 V), low-frequency (<100 kHz) electrical signal to electrodes applied

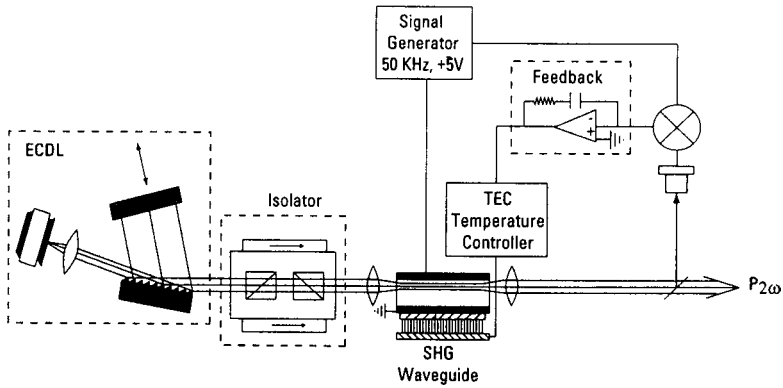


Figure 6.13: Configuration used by Bortz and coworkers for automatic adjustment of the phasematching temperature. (Reprinted with permission from Bortz *et al.* (1996).)

to the crystal. Phase-sensitive detection of a portion of the blue output yields an error signal indicative of the departure from perfect phasematching, which can be used to adjust the temperature of the waveguide substrate accordingly.

Although Bortz and coworkers used a thermoelectric Peltier element to adjust the temperature, a thin-film heater deposited atop the waveguide can be used instead. Mizuuchi *et al.* (1995) deposited a titanium film heater atop PPLT waveguides (with a SiO_2 buffer layer in between to minimize the loss caused by the presence of the Ti film), and showed that they could use it to adjust the phasematching wavelength over a 2–3 nm range.

6.3.5 Extrinsic efficiency enhancement

We have already mentioned various considerations that may lead to an increase in the intrinsic conversion efficiency of a waveguide doubler: for example, increased overlap integral, dimensionally-noncritical geometry, improved quality of the domain inverted grating, longer length. In addition, when a waveguide doubler is integrated with a diode laser, certain extrinsic variations may be explored with the aim of increasing the overall conversion efficiency. We will briefly discuss two of these: pulsed operation of the laser and waveguide resonators.

As mentioned in connection with single-pass SHG (Chapter 3), it is often possible to increase the average second-harmonic power by pulsing the infrared laser, so that a higher peak power is available. In applications which do not require cw blue-green light, this approach can be used very effectively. Azouz *et al.* (1995) demonstrated this approach using the configuration shown in Figure 6.14, in which application strong rf modulation at 650 MHz caused pulsed operation of the external cavity diode laser. They generated blue light at 415 nm by frequency-doubling in a PPLT

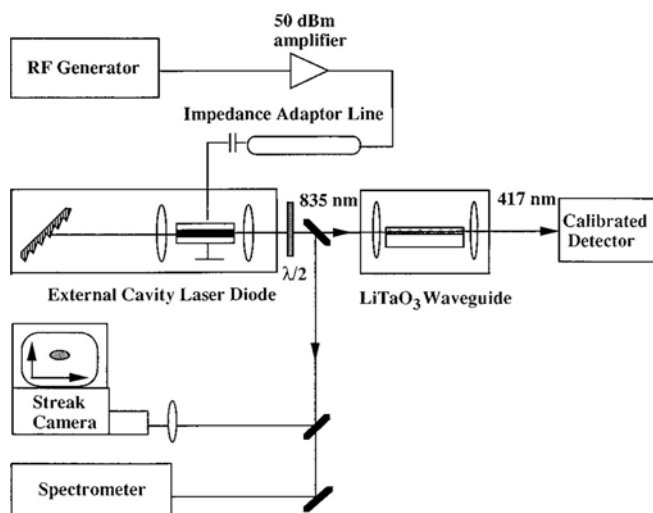


Figure 6.14: Arrangement used by Azouz and coworkers for increasing SHG efficiency by pulsing the laser diode. (Reprinted with permission from Azouz *et al.* (1995). Copyright: American Institute of Physics.)

waveguide, and found that they could achieve a factor of 15 larger conversion efficiency that was obtained when the laser was operated in a cw mode, even though the modulation caused the infrared spectrum to broaden beyond the phasematching bandwidth.

Birkin *et al.* (2001) addressed this problem by using *aperiodically*-poled LN in order to match the frequency acceptance bandwidth for frequency-doubling to the broadened spectrum of a gain-switched InGaAs/GaAs laser (~ 11 nm). With this approach, they generated 3.6 mW of blue (490 nm) light – for comparison, when they ran the laser cw at the same average power and used a PPLN crystal of comparable length, they were able to obtain only ~ 150 μ W of blue output power. Hence, they were able to take full advantage of the higher peak power attained by gain switching without suffering the limitation imposed by the relatively narrow phasematching bandwidth associated with a QPM crystal having a uniform period.

Another approach to externally increasing the conversion efficiency of a waveguide frequency doubler involves placing it within a resonator, similar to the approach used with bulk-crystal SHG discussed in Chapter 4 (Regener and Sohler, 1988). However, compared with its bulk-crystal counterpart, resonant waveguide SHG is generally somewhat less effective in enhancing conversion efficiency, because additional losses associated with the waveguide limit the finesse that can be achieved (Regener and Sohler, 1985).

The mirrors bounding the resonator can be either dielectric coatings applied to the end facets of the waveguide chip or DBRs integrated directly onto the

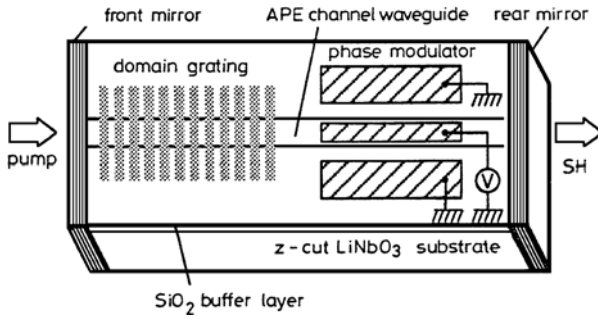


Figure 6.15: Waveguide resonator configuration used by Fujimura and colleagues for enhancement of guided-wave SHG. (Reprinted with permission from Fujimura *et al.* (1996).)

waveguide substrate. Shinozaki *et al.* (1991a) used the latter approach, using a sixteenth-order DBR formed by proton exchange. They were able to achieve a 40% increase in the conversion efficiency obtained on resonance in a QPM LN waveguide compared to a device made without the DBR gratings. Fujimura *et al.* (1996) used multi-layer dielectric mirrors glued to the endfaces of a QPM LN waveguide (Figure 6.15). They also integrated an electro-optic phase modulator to permit convenient tuning of the resonance. On resonance, they achieved a factor of 8 higher conversion efficiency than they obtained without the mirrors attached to the chip.

6.4 SUMMARY

In this chapter, we have examined blue-green light generation based on guided-wave SHG. We have seen that development of sources based on this approach has encountered difficulties that fall roughly into two categories: those associated with developing processing steps that repeatably produce a high yield of waveguides with high conversion efficiencies, and those associated with integrating the waveguide doubler with a semiconductor diode laser in a compact package. We reviewed potential solutions to these problems that have been explored by various groups, and saw that although difficulties remain, it has been possible to construct at least demonstration prototypes of very compact blue light sources based on waveguide doubling.

The next chapter begins a new part, in which we explore a completely different technology for blue-green light generation: one that involves frequency conversion based not on the second-order nonlinear susceptibilities of materials, but on materials having blue-green laser transitions that can be pumped using infrared photons.

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Part 2

Upconversion lasers: Physics and devices

7

Essentials of upconversion laser physics

The organizing idea of the previous chapters is that nonlinear, *nonresonant*, properties of insulating materials can be exploited to convert long-wavelength coherent radiation into short-wavelength radiation. As was discussed at length, this compels the device designer to simultaneously satisfy two demanding operational constraints: (1) Optical intensities at a first-harmonic frequency must be sufficiently high that the electromagnetic response of dielectric media is pushed into the nonlinear regime. (2) A travelling wave thus generated at the second-harmonic frequency must propagate at the same phase velocity as the fundamental wave lest the second harmonic switch roles from receiver to donor of optical power in the device. However, nonlinear frequency generation hardly requires that the operative interactions take place off resonance. Photon adding functions can be accomplished equally well with the aid of *resonant* optical processes in insulating materials, in turn wholly eliminating the two challenging constraints just named. We are, of course, referring to *upconversion lasers*. In this chapter and the next, we present this second approach to the nonlinear generation of short visible wavelengths and discuss what different operational challenges arise in creating a practical upconversion device.

7.1 INTRODUCTION TO UPCONVERSION LASERS AND RARE-EARTH OPTICAL PHYSICS

Upconversion lasers function just as ordinary lasers do, at least insofar as the mechanism by which their output beams are generated: A population inversion is created between two widely separated states thus making possible optical gain and laser oscillation in whatever media play host to the atoms, ions, or molecules possessing those states. The difference comes in the pumping mechanism. Instead of accomplishing upper-state pumping with an electrical discharge as in an argon ion laser, a flashlamp as in a Nd:YAG or ruby laser, or through single-photon laser pumping as with a cw dye laser, upconversion lasers engage pumping mechanisms

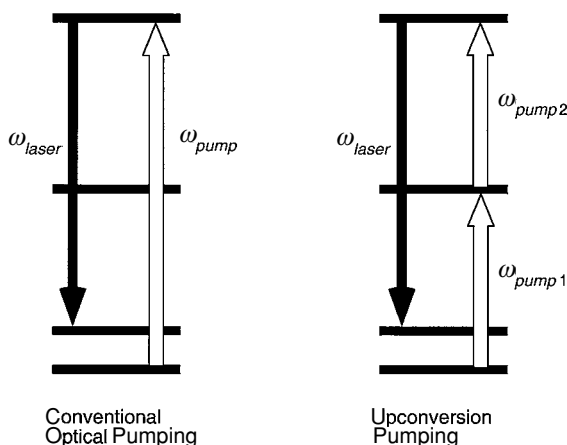


Figure 7.1: Illustration of the upconversion concept as compared with conventional optical pumping.

that combine the energy resident in *two* pump photons in order to create a *single* excited state of the gain-active species.

This idea is depicted in Figure 7.1, which shows a comparison of a conventional one-photon pumping scheme with a two-photon, upconversion scheme. The figure makes obvious why upconversion pumping is a viable approach for constructing compact short-wavelength lasers: For one, thermodynamics requires in the one-photon pumping scheme that the output wavelength be longer than the input wavelength – not of much value because one must already own a source of short-wavelength laser light. If that problem were solved there would be little need for this book. Conversely, the two-photon upconversion scheme clearly makes use of pump radiation at wavelengths longer than the output. If one can find one or two compact, efficient, high-power sources of long-wavelength radiation to serve as pumps plus a gain-active species – most desirably dissolved in a solid-state host – then the combination of these elements offers an attractive means for building a short-wavelength device in a small, durable package.

So let's cut to the chase. *The sources should be near-infrared semiconductor diode lasers and the gain-active species should be trivalent rare-earth ions.* Diode lasers are a clear choice: They offer the greatest advantages in terms of compactness, wide-ranging output wavelengths, high output power (≥ 1 W), modest electrical power requirements, and ready commercial availability at ever decreasing prices. That a solid-state solution of rare-earth or lanthanide ions should serve as the gain medium is a consequence of two important physical properties and one happy coincidence: Rare-earth ions in solids possess high-lying long-lived quantum states that exhibit blue and green fluorescence, suggesting these states' suitability as upper laser levels.

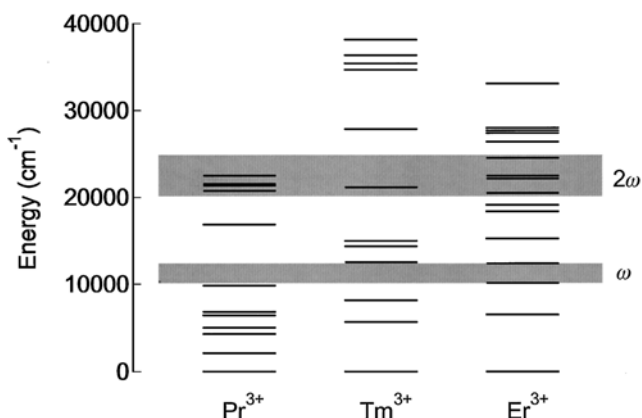


Figure 7.2: Energy-level diagrams for three of the most popular rare-earth ions used in upconversion laser studies. The shaded regions show the states accessible through one- and two-photon absorption with readily available semiconductor diode lasers.

Further, rare-earth species possess long-lived intermediate states able to serve as efficient energy storage levels in the upconversion chain (*viz.* Figure 7.1). As far as these crucial properties go, they are shared by few if any other gain-active species capable of generating visible light in the solid state. The happy coincidence about rare-earth ions is that the needed connecting transitions through the upconversion chain occur at near-infrared wavelengths. The sum of these ideas is captured in Figure 7.2, which shows energy levels for the three most popular rare-earth ions employed in upconversion laser research and their overlaps with the output photon energies of generally available near-infrared laser diodes (say 800–1000 nm). We will later study more subtle mechanisms of upconversion than the straightforward two-photon sequential absorption process discussed so far, still Figure 7.2 illustrates the fundamental strategy behind all of the upconversion mechanisms we will discuss in this book.

We have argued that solid-state upconversion lasers be naturally based on lanthanide ions; thus almost exclusively all upconversion laser research is focused on these species. Our first priority then is to study the optical physics of rare-earth ions in solids as well as other important physics that bears on upconversion laser operation. More specifically, in this chapter we discuss the quantum mechanical basis for the disposition of energy levels of rare-earth ions and the theory of optical transitions between them. Our aim is to provide sufficient detail and completeness on these subjects so that the interested reader can easily pick up threads of development in other books and especially in the often overly terse and specialized literature. These somewhat lengthy developments are followed by a discussion of single-ion nonradiative processes – those of the type that figure in the operation of almost

all lasers – and then by a discussion on the important topic of energy exchange between *pairs* of rare-earth ions. The latter underlie the more subtle mechanisms of upconversion to which we alluded earlier; such energy exchange will ultimately be cast in the form of rate equations that the motivated reader might use to build kinetic models of upconversion devices. Finally, the chapter concludes with a brief presentation of the essential laser physics with which the reader must be fluent in order to grasp the bare essentials of upconversion laser research as reported in the refereed literature. This presentation also emphasizes a rate-equation approach so as to mate smoothly with the rate-equation formulations of upconversion pumping developed just prior. With all the fundamentals in hand, Chapter 8 concludes this part of the book with a thorough account of upconversion laser research appearing in the open literature.

7.1.1 Overview of rare-earth spectroscopy

Much about the optical and physical properties of the triply-charged ions of the 15 rare-earth atoms can be understood with heuristic reasoning and more importantly a minimum of formalism and mathematical clutter. In particular, despite the complications of multi-electron Coulomb interactions, much simplification of the electron many-body problem is possible provided one is willing to be satisfied with calculated energies and wave functions lying within a few percent of the exact results. Even so, such calculations, which largely concern the *radial* electronic coordinates and address the overall energies of the atomic states, need not be explicitly undertaken. The character of the procedure should make clear how the dedicated student of atomic theory would, in principle, accomplish such calculations. (See the books by Cowan (1981), Bethe and Jackiw (1968), and Weissbluth (1978) for fundamental material on the quantum mechanics of multi-electron atoms and the books by Di Bartolo (1968), Dieke (1968), Hübner (1978), Powell (1998), and Wybourne (1965) for detailed theoretical discussions of spectroscopy in the defect solid state.)

More important for our purposes will be establishing how the electronic *angular* degrees of freedom are specifically entangled in both the structural and optical properties of rare-earth ions. This results from the simple fact that it is exclusively states and transitions within a single orbital shell (a notion to be sharpened in Section 7.3) that we need discuss in order to learn the useful lore. We will therefore assume a minimum of foreknowledge concerning the quantum mechanics of angular momentum. The reader should be comfortable with spherical harmonics, the eigenvectors and eigenvalues of the general angular momentum operators $\hat{\mathbf{J}}^2$ and $\hat{J}_z$ (as well as their purely orbital and spin analogs $\hat{\mathbf{L}}^2$, $\hat{L}_z$, $\hat{\mathbf{S}}^2$, and $\hat{S}_z$, respectively) and the angular momentum raising and lowering operators $\hat{J}_{\pm} = \hat{J}_x \pm i\hat{J}_y$. These topics are covered in any undergraduate quantum mechanics text. With only these

elements in hand, we will introduce the vector model of angular momentum and angular momentum coupling schemes, and thereby encounter the Clebsch–Gordon coefficients and the analogous Wigner 3-j symbol.

The goal we are headed toward (accomplished in Section 7.3) is this: For a chosen rare-earth ion in a given host material, the oscillator strength for electric-dipole transitions we will see is given by the simple formula

$$f_{|a\rangle \rightarrow |a'\rangle}^{ed} = K(\omega_0) \sum_{\lambda=2,4,6} \Omega_\lambda Q_\lambda(J, J') \quad (7.1)$$

where the three quantities Ω_2 , Ω_4 , and Ω_6 are the celebrated Judd–Ofelt intensity parameters, which once measured for a given ion in a given host material enable the calculation of the oscillator strength for any upgoing or downgoing transition within the $4f$ electronic shell. On the other hand, the Q_λ are quantities that for any transition from state $|a\rangle$ to $|a'\rangle$ are *independent* of the specific host material within which the ions are embedded and further are readily calculated in terms of the total angular momentum quantum numbers J and J' belonging to the two states. (That is, a great simplification arises in that the Q_λ do not depend on the magnetic quantum numbers m and m' .) Finally, $K(\omega_0)$ is a constant multiplier, dependent only upon the transition frequency ω_0 .

This is a powerful result. Note that a simple absorption measurement covering transitions between the ground state and three or more excited states is sufficient to uniquely determine the three Judd–Ofelt parameters. With this knowledge, the oscillator strength for *any* transition in the system can then be predicted once the simple calculations of the Q_λ are performed. In fact, since the Q_λ are independent of the specific host material at hand, the Q_λ can be computed once and for all for every transition in every rare-earth ion; wave functions computed for the free ion are often used for this purpose and the results published in tabular form. This procedure is useful in assessing the suitability of a given transition for laser applications, especially since many such transitions will occur between a pair of excited states and thus are not readily probed with a simple absorption measurement. That such a simplification is possible is a special property of the rare-earth ions, one that transition metal ions do not share. Here's why.

7.1.2 Qualitative features of rare-earth spectroscopy

The rare-earth or lanthanide series of elements, which begin with lanthanum at atomic number 57 and continue through lutetium at atomic number 71, almost universally assume the trivalent state when present as impurities in transparent insulators. In the $3+$ valence, the electronic structure of the ions is most simply understood as a series of spherically symmetric filled shells, each labelled with both a principal

quantum number n and an orbital angular momentum quantum number l , plus one unfilled shell whose heritage is that of the familiar $4f$ manifold of the hydrogen atom.

We will sharpen the meaning of the term “shell” shortly, but for now we note the most important qualitative feature of rare-earth electronic structure: Although the electrons of highest energy occupy the $4f$ shell, this shell is not located outermost from the atomic nucleus. The filled $5s$ and $5p$ shells, while of lower energy, are spatially localized *outside* of the $4f$ shell, as demonstrated by the relevant electron probability distributions illustrated in Figure 7.3. Assuming only an attractive spherically symmetric potential, this circumstance is a consequence of the more eccentric classical orbits affiliated with small angular momentum: in such orbits, the electron will be found on average at a greater distance from the nucleus than an electron of equal total energy but of larger angular momentum. In the quantum

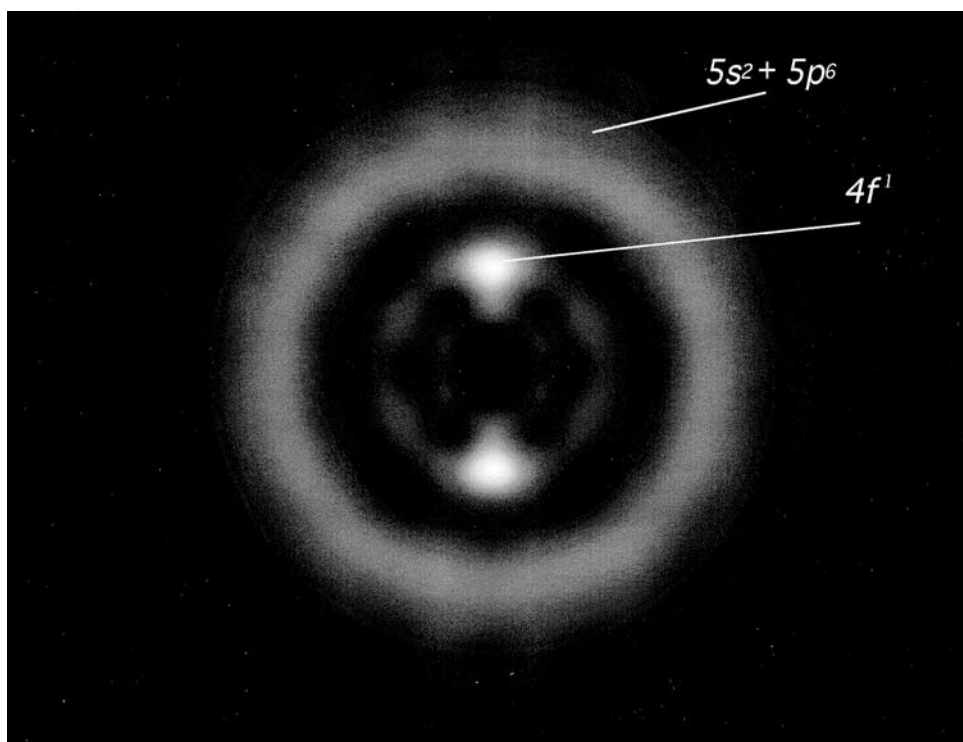


Figure 7.3: Total electron probability density for a single $4f$ electron and the spherically symmetric filled $5s^2$ and $5p^6$ shells. Although the latter two shells comprise electron states lying at lower energy than $4f$ electrons, the probability density affiliated with these electrons is largely concentrated at a greater distance from the ionic nucleus. The shells thus give rise to electrostatic screening in rare-earth doped solids. The plotted density is given by $\rho_{nl} = \sum_{m_l, m_s} |R_{nl}(r)Y_{lm_l}(\theta, \phi)|^2$, where for convenience pure hydrogenic wave functions have been used. The single $4f$ electron occupies the $m = 0$ magnetic sublevel.

mechanical case, the energy difference between the $5s$ and $5p$ states versus the $4f$ states is small enough so as to preserve this qualitative feature of the relative electron probability distributions.¹ The eight electrons comprising the filled $5s$ and $5p$ shells therefore act to shield the $4f$ electrons from external electrostatic fields, most notably those fields arising from a solid or liquid host medium. That is, these outer shells, by virtue of their greater proximity to the charges of the surrounding material, will spatially distort in response to the impressed electrostatic “crystal field” and therefore largely cancel this field in the vicinity of the $4f$ shell. The upshot of shielding is that all energy states of the $4f$ shell will strongly resemble those of the free ion. Such independence from the host material makes possible the plotting of generic energy-level diagrams for the $4f$ states for each rare-earth ion, as shown in Figure 7.4. Especially in crystalline host materials, spectral features are correspondingly sharp, with low-temperature linewidths as narrow as $0.01 \text{ \AA}$ having been observed.

As a point of contrast with the trivalent rare earths, the valence d electron orbitals of the transition metal ions are unshielded by outlying shells and even extend out into the host. As a result, strong coupling between the impurity and the host makes the spectroscopy of the transition metals highly host-dependent. In general, energies of the eigenstates vary widely and absorption lines are broad, the latter reflecting the strong mixing of impurity electronic states with host phonon states. Another consequence of strong impurity–lattice coupling in the transition ions is rapid non-radiative relaxation of the impurity electronic states, thus making transition metals unlikely candidates for laser applications. Nevertheless, a few long-lived excited states in transition metals do exist, most notable among these being the ^2E states of chromium in sapphire, which give rise to the narrow R -line emission and the red laser transitions of ruby.

We have so far spoken rather loosely about the nature of the $4f$ states of rare-earth ions. As an approach to understanding more precisely the physical composition of these states, first consider their origin in the familiar $4f$ hydrogenic wave functions. Including spin, 14 eigenstates exist for the hydrogen atom when $n = 4$ and $l = 3$. This fact entails an electron occupancy ranging from zero (La) to 14 (Lu) in the $4f$ shell of a rare-earth atom. In the hydrogen problem, however, all 14 eigenstates are degenerate, at least as long as spin–orbit coupling is ignored. Recall that the latter effect, as viewed in the rest frame of the orbiting electron, can be understood as arising from the interaction of the electron’s spin magnetic moment with the magnetic field established by the relative motion of the nucleus. In a similar way, and by further excluding the mutual Coulomb repulsion between $4f$ electron pairs, the multi-electron states of the $4f$ shell are also degenerate. For example,

¹ The thoughtful reader may point out that this argument fails to account for the degeneracy of different l shells for the same principal quantum number in the hydrogen atom. This turns out to arise from an additional symmetry of the hydrogenic Hamiltonian that is affiliated with the $1/r$ central potential and peculiar only to such systems.

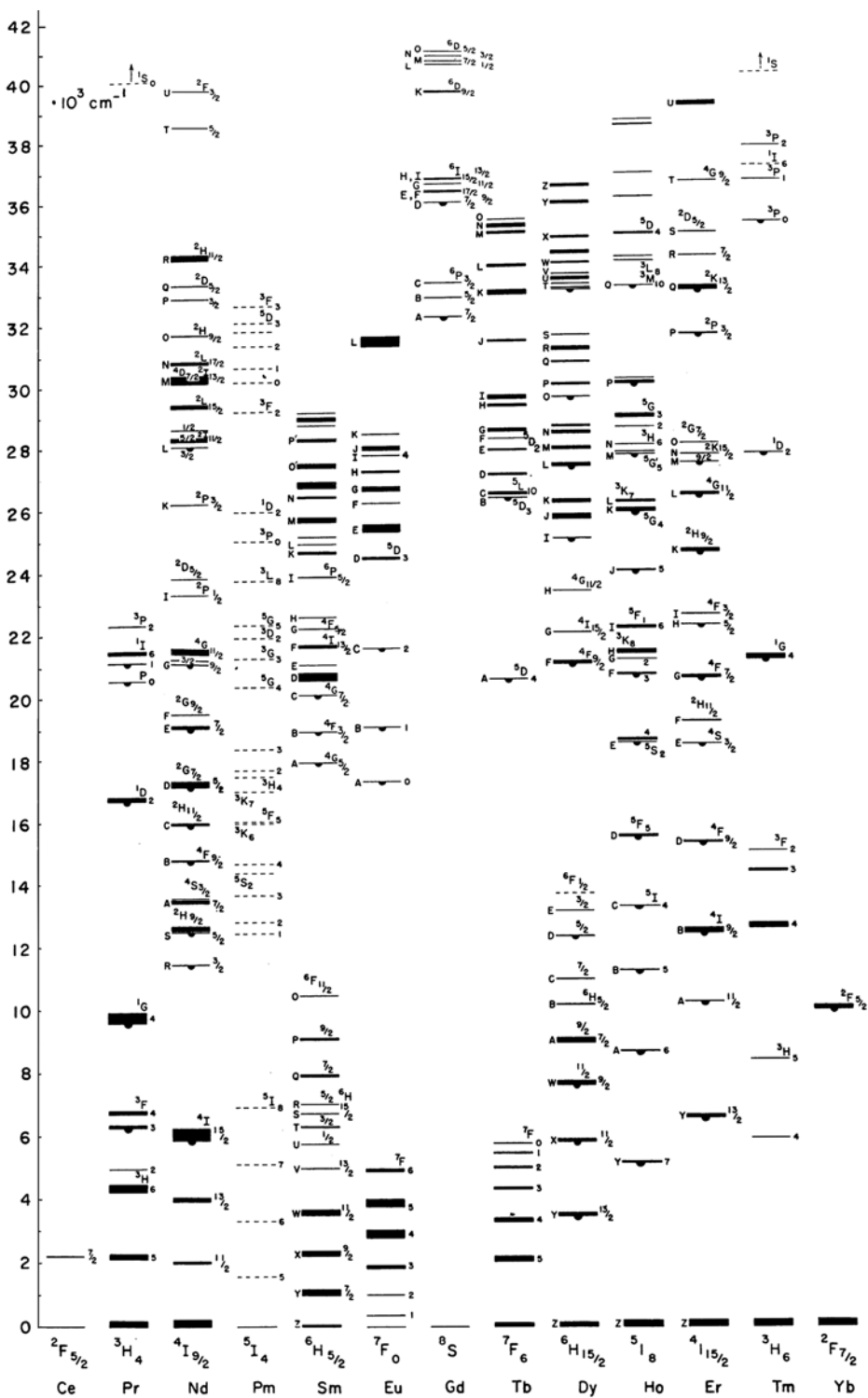


Figure 7.4: Energy levels for trivalent rare-earth ions in a LaF_3 host lattice. (Carnall *et al.*, 1977)

the Pr^{3+} ion possesses two $4f$ electrons which together, consistent with the indistinguishability of identical particles and the Pauli exclusion principle, make for a total of $(14 \times 13)/2 = 91$ degenerate eigenstates. However, when the spin-orbit and repulsive Coulomb interactions are switched on, this degeneracy is partially lifted and a spectrum of states spanning an energy range of about 2.5 eV results (see Figure 7.4). In spite of this splitting, the electron probability distributions derived from the spatial eigenfunctions are still well localized inside the $5s$ and $5p$ shells. Electrostatic shielding therefore prevails and the $4f$ energy states remain atom like when the ion is dissolved in a solid host. Spectral features are sharp, as alluded to previously. However, as will be shown below, the new eigenstates of a Hamiltonian that includes the spin-orbit and Coulomb interactions are just linear combinations of the old degenerate eigenstates. That is, only the original $4f$ eigenstates appear in the expansion of the interaction-modified eigenstates when the former are chosen as basis states for the electronic system – no states outside of the $4f$ shell contribute. This circumstance implies an important feature in the spectroscopy of free rare-earth ions: since all states within the $4f$ shell necessarily possess the same parity, electric-dipole transitions between states within this shell are strictly forbidden.

What then explains the presence of so many features in the absorption and emission spectra of rare-earth-doped solids? A small part of the answer is that a handful of magnetic dipole transitions allowed in the free ions are likewise found in the solid state. But the bulk of the answer is that the crystal field, although strongly shielded by the outer $5s$ and $5p$ shells, still weakly perturbs the $4f$ levels and mixes into their free-ion wave functions opposite-parity states found at energies above the $4f$ shell. In particular, owing to their proximity in energy to the $4f$ levels, it is the states of the $5d$ shell that contribute the lion's share of the required opposite-parity wave functions. This perturbation is much weaker than the spin-orbit or the interelectron Coulomb interaction, so that the $4f$ levels remain largely of free-ion character. In general, however, the result is that eigenstates of a rare-earth ion perturbed by the crystal field no longer possess definite parity and therefore, as far as electric-dipole transitions are concerned, just about anything goes. Transition oscillator strengths are nevertheless quite small (perhaps at most $f \approx 10^{-5}$) compared with ostensibly allowed electric-dipole transitions in free atoms ($f \approx 10^{-3}$ –1). Einstein A coefficients are correspondingly small in rare-earth materials, with excited-state lifetimes on the order of milliseconds commonly observed. Section 7.3 specifically addresses how the crystal field modifies the free-ion eigenstates, how this modification gives rise to nonvanishing electric-dipole oscillator strengths, and therefore how we arrive at the Judd–Ofelt form Equation (7.1). Meanwhile Table 7.1 gives a comparison of spectroscopic parameters for various well-known laser systems including the rare-earth-based Nd:YAG laser.

Table 7.1. *Some spectroscopic parameters for common laser systems.* (From Siegman (1986).)

Transition	Wavelength	Radiative lifetime	Peak stimulated emission cross-section	Oscillator strength
HeNe laser transition $3s_2 \rightarrow 2p_4$	633 nm	0.7 μ s	$\sim 10^{-13}$ cm ²	0.0047
Nd:YAG laser transition ${}^4F_{3/2} \rightarrow {}^4I_{3/2}$	1.064 μ m	1.22 ms	4.6×10^{-19} cm ²	$\sim 8 \times 10^{-6}$
Ruby laser transition ${}^2E \rightarrow {}^4A_2$	694 nm	4.3 ms	2×10^{-20} cm ²	$\sim 10^{-6}$
Rhodamine 6G dye laser transition $S_1 \rightarrow S_0$	620 nm	3.3 ns	2×10^{-16} cm ²	~ 1.1

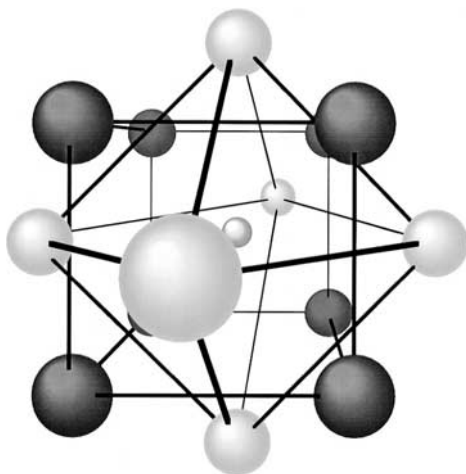


Figure 7.5: Generic picture of a rare-earth ion in an idealized electrostatic crystal field. Atoms of the host lattice are to be viewed as simple point charges fixed in space.

Before proceeding, we offer some final comments relating to the nature of the crystal field. Our picture of a simple electrostatic interaction between the field and the $4f$ shell is something of an idealization. We imagine that well removed from the ion are point charges in place of the host's atomic constituents (Figure 7.5). This so-called crystal field model is best realized for strongly ionic host crystals such as the alkali halides, in which the rare-earth impurity would reside as a cleanly isolated ion, although it is only doubly-charged rare-earths that in practice can be successfully incorporated into such a lattice. At the opposite extreme, *ligand field theory* and *molecular orbital theory* address the possibility of mutually interpenetrating charge

clouds that form conventional chemical bonds, a situation poorly represented by our notion that the $4f$ shell is impressed with a simple electrostatic potential satisfying the Laplace equation $\nabla^2 \Phi(\mathbf{r}) = 0$. Reality lies between these extremes, although in most real-world host materials the case is somewhat tilted towards the more ionic picture, thus allowing us to continue to at least qualitatively view the rare-earth ion as a distinct entity subject to an electrostatic field created by an external point-charge distribution. A more accurate view allows for multi-pole moments of the constituent host atoms to be introduced, but in actuality it is only in a few instances that *a priori* calculation of the ion–host interaction has been attempted. Such approaches have not been wholly successful for reasons still under debate. Far more successful has been the approach that leaves the details of the interaction aside, retains the crystal field model, but deposits the unknown effects of the unspecified exact interaction into adjustable parameters that otherwise arise as well-determined constants in a point-charge picture of the interaction. It is a remarkable success of the theory that these so-called crystal field parameters depend mostly on the host material and otherwise vary slowly and smoothly as one moves through the lanthanide series. In short, the complications of covalency are phenomenologically parametrized within the crystal field model. We will later see how the crystal field parameters arise in the theory and, in particular, how the Judd–Ofelt intensity parameters depend upon them, as they must.

A last point regarding the origin of electric-dipole transitions in matrix-isolated rare-earth ions is that an additional important mechanism exists that is capable of producing the required admixture of mixed-parity free-ion eigenstates within the $4f$ shell. To see how this mechanism appears, first note that it is only odd-parity crystal field components that are responsible for adding opposite-parity states to the $4f$ free-ion eigenstates. While amorphous hosts offer such odd-parity crystal field components in abundance, crystalline materials exhibiting inversion symmetry possess no such components and therefore do not yield allowed electric-dipole transitions. Nevertheless, if one includes the possibility of *dynamical* contributions to the crystal field, namely, those arising from thermal motion of the impurity or more importantly those of the host lattice, then time-dependent, odd-parity, crystal field components readily occur. Aside from the appearance of electric-dipole transitions in high-symmetry crystals, further evidence for the existence of this dynamical contribution is the observation of phonon-related frequency-modulated sidebands and temperature-dependent linewidths in $4f$ spectral features. Thus in low-symmetry hosts the oscillator strengths for electric-dipole transitions derive from both static and dynamic contributions, the former being indistinguishable from the latter at the current state of experimental craft. Despite this additional complication, however, the effects of the dynamic contributions to the oscillator strength are *still* captured phenomenologically by the Judd–Ofelt theory through the three intensity parameters, Ω_λ .

7.2 ELEMENTS OF ATOMIC STRUCTURE

One's naive first impression when faced with the task of finding the eigenstates and eigenvalues of a multi-electron atom is that the many-body interactions part and parcel to this problem make it wholly intractable. The truth is that great simplifications arise as a consequence of spherical symmetry. Nevertheless, it is not our intention here to undertake a full accounting of atomic theory. Instead, we present an outline of the general ideas so that we can speak comfortably of electronic configurations and electronic shells. This will lead to a simple picture of the electronic structure of rare-earth ions: the electrons of the unfilled $4f$ shell will be seen to move in an effective spherically symmetric potential that results from the sum of the nuclear potential and an average Coulomb interaction derived from all the other electrons. In fact, it can be shown that since these latter electrons all occupy filled shells of the atom, this effective central potential is exact so long as one ignores the possibility that the $4f$ electrons might slightly distort the charge distribution in the closed shells and thereby break spherical symmetry. Thus the complexity of computing the energy-level structure of N $4f$ electrons reduces to considering interactions between these N electrons alone subject to the effective central potential. Angular momentum conservation principles therefore figure prominently in this task.

7.2.1 The effective central potential

As originally introduced by Hartree, the essential assumption underlying calculations of the electronic structure of multi-electron atoms is that each electron is described by its own wave function. This amounts to an approximation in which electron–electron correlation effects are ignored. Thus if this single-particle wave function is denoted by $\tilde{\psi}_i(\mathbf{r}_i)$ for the i th electron, the potential energy operator for this electron is given by

$$\hat{U}(\hat{\mathbf{r}}_i) = \frac{-Ze^2}{|\hat{\mathbf{r}}_i|} + \sum_{k \neq i} \int \frac{e^2 |\tilde{\psi}_k(\mathbf{r}_k)|^2}{|\mathbf{r}_k - \hat{\mathbf{r}}_i|} d^3\mathbf{r}_k \quad (7.2)$$

where the sum is over all the other single-particle states with their corresponding spatial probability distributions interpreted as charge densities within the atom. For an N -electron atom, the problem is to find self-consistent solutions to the N coupled time-independent Schrödinger equations

$$\left[\frac{\hat{p}_i^2}{2m_e} + \hat{U}(\hat{\mathbf{r}}_i) \right] |\psi_i\rangle = \varepsilon_i |\psi_i\rangle \quad (7.3)$$

subject to the definition of $\hat{U}(\mathbf{r}_i)$ above. In order to proceed, one then invokes a second approximation whereby Equation (7.2) is averaged over all directions in

order to yield the effective central potential

$$\hat{U}(\hat{r}_i) = \frac{1}{4\pi} \int \hat{U}(\hat{\mathbf{r}}_i) d\hat{\Omega} \quad (7.4)$$

Under the central field approximation, just as for the hydrogen atom, the radial and angular degrees of freedom are separable, thus Equations (7.3) yield solutions of the form

$$\tilde{\psi}_i(r_i, \theta_i, \phi_i) = \frac{R_{i,nl}(r_i)}{r_i} Y_{lm}(\theta_i, \phi_i) \quad (7.5)$$

with quantum numbers n , l , and m , where $R_{i,nl}(r_i)$ is a purely radial function exhibiting $n - l - 1$ nodes in the open interval between $r_i = 0$ and $r_i = \infty$ and the $Y_{lm}(\theta_i, \phi_i)$ are the familiar spherical harmonics.

Owing to the spherical symmetry of the Hamiltonian in the central field approximation, inspection of the wave functions Equation (7.5) reveals that the energy eigenvalues can only depend on the quantum numbers n and l . Based on our knowledge of the hydrogen atom, it further should come as no surprise that the single-electron eigenvalues $\varepsilon_{i,nl}$ increase with increasing principal quantum number n . Since in all generality we are no longer dealing with central potentials of the form $U(r) \sim 1/r$, it can be shown that $\varepsilon_{i,nl}$ likewise increases with increasing l . We are thus led to the concept of the electronic *shell* in the Hartree/central field approximation. By this term, we simply mean the ensemble of single-electron states with common n and l . As further reflection on the form of the wave functions reveals, these states must be degenerate, since for different electrons of the same shell, $R_{i,nl}(r) \equiv R_{j,nl}(r)$. Moreover, consistent with this equivalence of the radial wave functions, combined with the $(2l + 1)$ fold degeneracy of the eigenstates with respect to the angular momentum quantum number l , plus the additional doubling of available states due to electron spin, the Pauli exclusion principle implies that a maximum of $4l + 2$ electrons may be labelled by the same n and l . We may therefore speak of how many electrons out of the allowed maximum of $4l + 2$ occupy a given shell; the shell is called closed if it is fully occupied. In the language of atomic theory, electrons belonging to the same shell are termed “equivalent”; we shall later see that this designation is partly intended to remind us of the special consequences that the Pauli exclusion principle entails for such electrons.

We are now able to demonstrate an important feature of the shell model of the atom, namely, that closed shells exhibit a spherically symmetric charge distribution and therefore contribute an exact spherically symmetric potential to the Hamiltonian. The charge density within a given closed shell is given by

$$\rho(n, l) = 2 \sum_{m=-l}^l |\tilde{\psi}_{nlm}|^2 = 2 \left[\frac{R_{nl}(r)}{r} \right]^2 \sum_{l=-m}^m |Y_{lm}(\theta, \phi)|^2 = 2 \left[\frac{R_{nl}(r)}{r} \right]^2 \frac{2l + 1}{4\pi} \quad (7.6)$$

where we have suppressed the index labels on the different electrons of the shell and exploited the equivalence of the radial portion of the wave function for different electrons. The leading factor of 2 derives from the two-fold spin degeneracy of the wave functions. The last step is the essence of the proof; it is a consequence of the addition theorem of spherical harmonics and may be easily verified by direct substitution for special cases of the quantum number l . What is perhaps even more remarkable is that it is also easily shown that the Coulomb interaction between a chosen electron and the electrons of a separate closed shell that does not include the chosen electron is likewise spherically symmetric. Thus in the spirit of the self-consistent field approach to the solution of the multi-electron atom, a closed shell contributes an *exact* spherically symmetric potential to the Hamiltonian even when Coulomb interactions between the closed shell and an electron outside of the shell are examined explicitly. The central field approximation is a particularly mild one in light of this property. Since for a trivalent rare-earth ion only the $4f$ shell (that is, $n = 4, l = 3$) is unclosed, these electrons experience an exact spherically symmetric potential with respect to the nucleus and the non- $4f$ electrons of the ion.

An important additional element of the theory not originally considered by Hartree was later contributed by Fock and Slater. Specifically, these theorists included at the outset the restriction that the Pauli exclusion principle places on the total wave function of the multi-electron atom, namely, that this wave function be anti-symmetric under exchange of any two electrons. Thus the Hartree–Fock theory proposes as a starting point fully anti-symmetrized multi-electron wave functions constructed from the single-particle wave functions of the type already discussed. These properly symmetrized multi-electron wave functions are given by

$$\Psi_N(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \dots, \mathbf{r}_{N-1}, \mathbf{r}_N) = \frac{1}{\sqrt{N!}} \det \begin{pmatrix} \psi_1(\mathbf{r}_1) & \psi_1(\mathbf{r}_2) & \dots & \psi_1(\mathbf{r}_N) \\ \psi_2(\mathbf{r}_1) & \psi_2(\mathbf{r}_2) & \dots & \psi_2(\mathbf{r}_N) \\ \vdots & \vdots & & \vdots \\ \psi_N(\mathbf{r}_1) & \psi_N(\mathbf{r}_2) & \dots & \psi_N(\mathbf{r}_N) \end{pmatrix} \quad (7.7)$$

Insofar as the effective potential derived from this more appropriate choice of basis states is concerned, many of the same results obtain, except that a so-called “exchange” term appears in the effective electrostatic potential. For closed shells, however, this term likewise contributes an exact effective central potential to the problem, so that unfilled shells aside, an exact spherically symmetric theory results. Many important features of the multi-electron atom may be predicted with the Hartree–Fock model, including the basic structure of the periodic table, ionization potentials, and the filling order of the shells with increasing atomic number. The last leads to the determination of the electronic *configuration* of a multi-electron atom, defined as a sequence of labels $n l^{N_l}$ that indicate the occupancy, N_l , of the various shells. For example, neutral chromium, with 24 electrons, assumes the configuration $1s^2 2s^2 2p^6 3s^2 3p^6 3d^5 4s^1$, where the conventional spectroscopic notation s, p, d, f

is employed for the angular momentum quantum numbers $l = 0, l = 1, l = 2, l = 3$, respectively. Note the competition for occupancy evident between the $3d$ and $4s$ shells, a consequence of the nearly degenerate energies for these shells within the transition metal series of elements.

7.2.2 Electronic structure of the free rare-earth ions

At this point we defer further study of the general electronic structure of multi-electron atoms to authors listed in the references. For our purposes, we now wish to restrict attention to the electronic structure and optical spectroscopy of the $4f$ shell of free trivalent rare-earth ions. We first remind the reader that these ions consist of several closed and therefore spherically symmetric shells plus the unfilled $4f$ shell. This last and highest-energy shell is capable of holding a maximum of 14 electrons; for N $4f$ electrons, the electronic configuration of the N th rare-earth ion is just $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4f^N 5s^2 5p^6$ or $4f^N 5s^2 5p^6$ for short. Our approach will be to initially ignore spin-orbit and Coulomb interactions among the $4f$ electrons and consider only their independent motion in the presence of the central potential due to the nucleus and the remaining electrons. We shall then comment on the explicit effect of these additional interactions on the $4f$ states. Note that as far as the Coulomb interactions are concerned, this procedure departs from the prescription dictated by the pure Hartree-Fock theory in that one $4f$ electron would otherwise be assumed to experience an angle-averaged effective potential arising from the remaining $(N - 1)$ $4f$ electrons of the shell. However, more accurate results are obtained if only the filled shells are constructed according to the Hartree-Fock prescription while the intra- $4f$ Coulomb terms are treated explicitly. The multi-electron problem is thus reduced to an N -electron problem where $0 \leq N \leq 14$ with the free-ion Hamiltonian

$$H = \sum_{i=1}^N \left[\frac{\hat{p}_i^2}{2m_e} + \hat{U}(\hat{r}_i) + \xi(\hat{r}_i) \hat{\mathbf{l}}_i \cdot \hat{\mathbf{s}}_i + \frac{1}{2} \sum_{j \neq i} \frac{e^2}{|\hat{\mathbf{r}}_i - \hat{\mathbf{r}}_j|} \right] \quad (7.8)$$

where $\xi(\hat{r}_i) \hat{\mathbf{l}}_i \cdot \hat{\mathbf{s}}_i$ is the spin-orbit interaction (to be discussed further below) and the Coulomb terms $e^2/|\hat{\mathbf{r}}_i - \hat{\mathbf{r}}_j|$ refer only to the electrons within the $4f$ shell. We illustrate below with several examples how the spectroscopy of the rare earths is interpreted in terms of this Hamiltonian.

7.2.2.1 Lanthanum ($4f^0 5s^2 5p^6$)

With no $4f$ electrons La^{3+} possesses an electronic structure analogous to that of the noble gas xenon. The ion therefore exhibits little in the way of interesting spectroscopy: For one, it can be shown that the spin-orbit interaction is

identically zero within a closed shell; the Hartree–Fock energy of the ground state is unchanged by this interaction. Moreover, since the ground state is singly degenerate (there is only one way $4l + 2$ electrons can occupy a shell consisting of $4l + 2$ single-electron orbitals), no other interactions can yield low-lying excited states even including corrections beyond the Hartree–Fock approximation. The first possible optical transition is therefore the interconfigurational transition $4f^0 5s^2 5p^6 \rightarrow 4f^1 5s^2 5p^5$. However, this transition occurs at an energy of about 17 eV and therefore the ion must be transparent from dc to well into the vacuum ultraviolet. Lanthanum still plays a crucial role in rare-earth materials: Its transparency makes it useful as a constituent of host materials, while due to the similar chemistry of all members of the rare-earth series, the ion is readily replaced with spectroscopically more interesting lanthanide species. Indeed the most complete spectroscopic information on matrix isolated rare-earth ions is for a LaCl_3 host crystal. We can further surmise from these facts that analogous interconfigurational transitions $4f^N 5s^2 5p^6 \rightarrow 4f^{N+1} 5s^2 5p^5$ for the remaining rare earths will likewise occur at multi-electron-volt energies and therefore these transitions do not clutter the visible–ultraviolet spectra of these ions.

7.2.2.2 Cerium ($4f^1 5s^2 5p^6$) and the addition of angular momenta

Trivalent cerium resembles the hydrogen atom insofar as a single electron is bound by a central potential. At the lowest level of approximation, the state of this single electron can thus be denoted by $|\alpha, l m_l s m_s\rangle$, where l and m_l are the quantum numbers affiliated with the orbital angular momentum operators, $\hat{l}^2 = \hat{\mathbf{l}} \cdot \hat{\mathbf{l}}$ and $\hat{l}_z = \hat{\mathbf{l}} \cdot \mathbf{e}_z$, respectively ($\mathbf{e}_z$ being the real-space unit vector pointing in the z -direction), and s and m_s are the corresponding quantities for the spin angular momenta $\hat{s}^2$ and s_z . The symbol α stands for any remaining quantum numbers of the system, in particular the principal quantum number n . The wave function in terms of the real-space coordinates r , θ , and ϕ and the two-component spinor $\chi_s(m_s)$ representing the spin degree of freedom must therefore be

$$\psi_{nlm_l, sm_s} = \frac{R_{nl}(r)}{r} Y_{lm_l}(\theta, \phi) \chi_s(m_s) \quad (7.9)$$

in direct analogy to the hydrogen atom. The radial functions $R_{nl}(r)$, however, are solutions to the more general differential equation in the variable r appropriate to the Hartree–Fock central potential $U(r)$. In the ground state, 14 degenerate states are thus obtained.

Unlike the hydrogen atom, however, the large angular momentum of the ground state plus the larger effective Z of the binding nucleus implies a much more sizable spin–orbit interaction. Indeed the spin–orbit splitting of the ground state of the free

Ce^{3+} ion amounts to 2000 cm^{-1} , about a thousand times larger than in the ground state of hydrogen. A large spin–orbit interaction is a central feature of the lanthanide series so we will invest a few more words on its behalf.

We first recall the argument for the mathematical form of the spin–orbit term as concerns a single electron: Special relativity applied to electromagnetism demonstrates that an object in motion with respect to an electrostatic field will in its rest frame experience a magnetic field according to

$$\mathbf{B} = \frac{1}{c} \mathbf{E} \times \mathbf{v} \quad (7.10)$$

where $\mathbf{v}$ is the velocity of the object and only terms of lowest order in v/c are shown. In short, the fixed charges in the electrostatic field yield a current as viewed in the object's rest frame and therefore through the Biot–Savart law a magnetic field results. If the object is an electron and the electric field arises from a central electrostatic potential $-U(r)/e$ then Equation (7.10) becomes

$$\mathbf{B} = \frac{1}{ec} \frac{dU(r)}{dr} \frac{\mathbf{r}}{r} \times \frac{\mathbf{p}}{m_e} = \frac{1}{em_e c} \left[\frac{1}{r} \frac{dU(r)}{dr} \right] \mathbf{l} \quad (7.11)$$

where $\mathbf{l} \equiv \mathbf{r} \times \mathbf{p}$ is the electron's angular momentum as observed in the rest frame. If the magnetic dipole moment operator of the electron is denoted by $\hat{\mu}_s = -(e/m_e c) \hat{\mathbf{s}}$, where $\hat{\mathbf{s}}$ is the electron spin, then in terms of quantum operators the spin–orbit Hamiltonian becomes

$$\hat{H}_{so} = -\hat{\mathbf{B}} \cdot \hat{\mu}_s = \frac{1}{2m_e^2 c^2} \left[\frac{1}{\hat{r}} \frac{d\hat{U}(\hat{r})}{d\hat{r}} \right] \hat{\mathbf{l}} \cdot \hat{\mathbf{s}} \quad (7.12)$$

where the additional factor of $1/2$ is a relativistic correction arising from the so-called Thomas precession. This form immediately suggests that a mutual torque exists between the two angular momenta and that therefore neither momentum can continue to be a constant of the motion. Rather, our classical expectation is that $\hat{\mathbf{j}} = \hat{\mathbf{l}} + \hat{\mathbf{s}}$ is such a constant. The components of the operator $\hat{\mathbf{j}}$ may be readily shown to exhibit the same commutation relations as those of $\hat{\mathbf{l}}$ or $\hat{\mathbf{s}}$, thus making $\hat{\mathbf{j}}$ in the quantum operator sense a formal angular momentum with eigenvalues and eigenvectors of the type with which we are already familiar. As the reader may recall, since $\hat{\mathbf{j}}$ is a general angular momentum, it admits eigenvalues that may be both integral and half-integral.

In terms of the operator $\hat{\mathbf{j}}$, it is easy to show that $\hat{\mathbf{l}} \cdot \hat{\mathbf{s}} = (\hat{j}^2 - \hat{l}^2 - \hat{s}^2)/2$. Moreover, it is easy to see that the operators $\hat{l}^2$ and $\hat{s}^2$ commute with $\hat{\mathbf{l}} \cdot \hat{\mathbf{s}}$, thus suggesting that we recast the eigenstates of the unperturbed Hamiltonian in terms of eigenstates of the operators $\hat{l}^2$, $\hat{s}^2$, $\hat{j}^2$ and $\hat{j}_z = \hat{l}_z + \hat{s}_z$, the last included in light of our expectation that the total angular momentum $\hat{\mathbf{j}}$ will be conserved. In this way, the spin–orbit Hamiltonian will be automatically diagonalized with respect to the new

basis, at least as far as the angular degrees of freedom are concerned. Of course, the radial factor in the spin-orbit term does not, in general, become diagonal under any simple change of basis and therefore we must still treat the spin-orbit interaction as a perturbation. In particular, with respect to Ce^{3+} , first-order degenerate perturbation theory will tell us how spin-orbit coupling will split the 14-fold degeneracy of the ground state.

Note, however, that in order to carry out this computation in the new basis a crucial step is required: In going from the $\hat{l}^2, \hat{l}_z, \hat{s}^2, s_z$ representation to the $\hat{l}^2, \hat{s}^2, \hat{j}^2, \hat{j}_z$ representation one needs to determine precisely what values of the quantum numbers j and m are to be affiliated with the new basis states $|lsjm\rangle$. We know at the very least that for fixed values of l and s , the total number of new eigenstates should be the same as the number of old ones, namely $(2s + 1) \times (2l + 1)$. We also know that for a given allowed j , m is permitted to range from $-j$ to j , consistent with $\hat{\mathbf{j}}$ being an angular momentum. We might further expect that the largest possible value of m would be $l + s$, given that this sum is the largest possible eigenvalue of $\hat{l}_z + \hat{s}_z$. Hence, the largest possible value of j must be $l + s$. We at this point resort to the classically motivated vector *model* for the addition of two angular momenta to assert that the minimum allowed value of j is just $|l - s|$, as suggested by Figure 7.6, and that this therefore exhausts all the possible j values if $s = \frac{1}{2}$. These expectations are rigorously proved in any advanced quantum mechanics text, but we can verify their self-consistency by noting that for the case $s = \frac{1}{2}$, $l > s$ always, and that therefore the total number of $|lsjm\rangle$ eigenstates must be

$$\sum_{j=l-s}^{l+s} 2j + 1 = [2(l - s) + 1] + [2(l + s) + 1] = 4l + 2 = (2s + 1)(2l + 1) \quad (7.13)$$

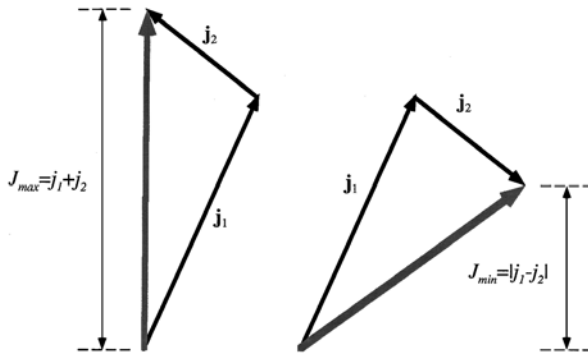


Figure 7.6: Vector model for the addition of two quantum angular momenta, $\hat{\mathbf{J}} = \hat{\mathbf{j}}_1 + \hat{\mathbf{j}}_2$. The model dictates that the allowed values of the total angular momentum quantum number J range from $J_{\max} = j_1 + j_2$ to $J_{\min} = |j_1 - j_2|$, as indicated heuristically in the figure.

as we insisted above. For the single $4f$ electron of Ce^{3+} ($l = 3$, $s = 1/2$), we thus obtain $j = 7/2$ and $j = 5/2$ as the only allowed values of j .

We are now in a position to carry out the prescriptions of degenerate perturbation theory to compute the energies of the low-lying states of Ce^{3+} in the presence of spin-orbit coupling. The procedure is to calculate the eigenvalues of the $(4l + 2) \times (4l + 2)$ array built out of the matrix elements of the interaction within the $(4l + 2)$ -fold degenerate subspace of the nl shell:

$$\begin{aligned} \langle \alpha, lsjm | \hat{H}_{so} | lsj'm' \rangle &= \langle \alpha, lsjm | \hat{\xi}(\hat{r}) \hat{\mathbf{I}} \cdot \hat{\mathbf{S}} | \alpha, lsj'm' \rangle \\ &= \langle \alpha | \hat{\xi}(\hat{r}) | \alpha \rangle \langle lsjm | \frac{\hat{j}^2 - \hat{l}^2 - \hat{s}^2}{2} | lsj'm' \rangle \\ &= \zeta_{nl} \left[\frac{j(j+1) - l(l+1) - s(s+1)}{2} \right] \delta_{jj'} \delta_{mm'} \end{aligned} \quad (7.14)$$

where ζ_{nl} is the *spin-orbit coupling parameter* given by the radial integral

$$\zeta_{nl} = \langle \alpha | \hat{\xi}(\hat{r}) | \alpha \rangle = \int_0^\infty \frac{\hbar^2}{2m_e^2 c^2} \left[\frac{1}{r} \frac{dU(r)}{dr} \right] R_{nl}^2(r) dr \quad (7.15)$$

This last quantity is thus computed exclusively from the radial portion of the Hartree-Fock single-electron wave function, $R_{nl}(r)$, and depends only on the specific shell of interest. It thus will not depend on the number of electrons present, nor their angular variables, and can be computed once and for all for each rare-earth ion once the Hartree-Fock wave functions are known. Tables of the ζ_{nl} are published in the literature (see Table 7.5 for an example); for the $4f$ shell of Ce^{3+} , ζ_{4f} is 625 cm^{-1} .

Returning to the determination of the energy eigenvalues under the spin-orbit interaction, note that from Equation (7.14) the matrix elements are independent of the magnetic quantum numbers and zero unless $j = j'$, the very fact that motivated our change of representation in the first place. For any one electron state of Ce^{3+} , then, the eigenvalue problem simplifies to consideration of the 2×2 array

$$\begin{aligned} H_{so,nl} &= \frac{\zeta_{nl}}{2} \begin{bmatrix} (l - \frac{1}{2})(l + \frac{1}{2}) - l(l+1) - \frac{3}{4} & 0 \\ 0 & (l + \frac{1}{2})(l + \frac{3}{2}) - l(l+1) - \frac{3}{4} \end{bmatrix} \\ &= \frac{\zeta_{nl}}{2} \begin{bmatrix} -(l+1) & 0 \\ 0 & l \end{bmatrix} \end{aligned} \quad (7.16)$$

where the upper-left array element corresponds to $j = l - 1/2$ and the lower-right element corresponds to $j = l + 1/2$. Specializing now to the case of the Ce^{3+} ground state, the eigenvalues with $l = 3$ become for the two allowed j values $\lambda_{5/2} = -2\zeta_{4f}$ and $\lambda_{7/2} = 3\zeta_{4f}/2$. With $\zeta_{4f} = 625 \text{ cm}^{-1}$, the spin-orbit energy splitting is thus $\sim 2190 \text{ cm}^{-1}$. Two new degenerate manifolds are produced, the

lower-energy $j = 5/2$ manifold being six-fold degenerate and the higher-energy $j = 7/2$ being eight-fold degenerate for a total of 14 states. Compare these results with the energy levels shown in Figure 7.4.

As a final comment, we make first acquaintance with a notational scheme that everyone involved with rare-earth materials will encounter and should understand. According to convention, eigenstate manifolds are designated with the spectroscopic notation $^{2S+1}L_J$ so as to remind us of the relevant quantum numbers from which the states are derived. The notation applies to eigenstates of one or more electrons in one or more shells. Thus L designates the quantum number of the total orbital angular momentum $\hat{\mathbf{L}}$; it assumes the form of a capital letter S, P, D, F, G, H, I, K , and alphabetically ordered Latin capitals thereafter for $L = 0, 1, 2, 3, 4, 5, 6, 7$, and so on, respectively. The symbol S in the notation $^{2S+1}L_J$ corresponds to the total spin $\hat{\mathbf{S}}$ and the symbol J corresponds to the sum $\hat{\mathbf{J}} = \hat{\mathbf{L}} + \hat{\mathbf{S}}$. We will say more about how these quantities are defined when we discuss multi-electron ions below.

For now, we apply this spectroscopic notation to Ce^{3+} as follows. Since $l = 3$, we simply have $L \equiv F$, as we are dealing with a single f electron. (A single d electron, for example, would have dictated writing $L = D$.) The subscript J takes on the values $5/2$ or $7/2$ corresponding to the allowed eigenvalues of $\hat{J}^2$ as derived above, while $2s + 1 = 2$ is a quantity called the spin multiplicity. We thus have the two *spectroscopic terms* $^2F_{5/2}$ and $^2F_{7/2}$ for the $4f$ shell of Ce^{3+} . Note the use of these designations in the energy-level diagram of Figure 7.2. These labels continue to be used even as additional interactions are introduced. With the spectroscopic term labels in hand, we are thus able to deduce the total degeneracy (it is just $2J + 1$), the factor by which the degeneracy results from degenerate spin states (the spin multiplicity), and the total orbital angular momentum.

7.2.2.3 Ytterbium and the equivalence of electrons and holes

As mentioned for the case of lanthanum, closed shells are singly degenerate and can be shown to exhibit a vanishing spin-orbit interaction as regards the member electrons of the shell. These two facts lead to the notion of the *hole*, defined in the simplest case as a full shell with one electron removed. We will see that the spectroscopy of a one-hole system is very similar to a one-electron system and therefore all that we have learned about Ce^{3+} applies to the 13-electron ion Yb^{3+} .

The argument goes as follows. One first recognizes that the total angular momentum for a closed shell containing N electrons must vanish. Although we have not yet discussed the theory of angular momentum for a multiply occupied shell, we can easily imagine the existence of a total angular momentum operator for

the entire shell. Such an angular momentum would be conserved insofar as the electrons of this shell experience a purely spherical potential due to the nucleus and the remaining electrons of the atom. Since the filled shell is singly degenerate, the only possible allowed J appropriate to such a state is $J = 0$.

Now consider the result of extracting one electron from the closed shell. Since the remaining $N - 1$ electrons still conserve angular momentum, the angular momentum operators $\hat{\mathbf{I}}$ and $\hat{\mathbf{S}}$ affiliated with the now removed electron may be used to represent the angular momentum of the remaining electrons provided we set

$$m_l = - \sum_{i=1}^{N-1} m_{l_i} \quad (7.17a)$$

and

$$m_s = - \sum_{i=1}^{N-1} m_{s_i} \quad (7.17b)$$

In other words, the total z -component of the orbital and spin angular momenta of the $(N - 1)$ -electron system are just the negatives of the m_l and m_s connected with the unoccupied orbital. The number of possible states for the $N - 1$ electrons is thus the same as for one electron. Ignoring for the moment the possible effect of Coulomb interactions within the one-hole shell, the energy-level structure must therefore be directly analogous to the one-electron states of the same shell. It is possible to show, however, that an electron within a closed shell moves in an exact central potential with respect to the remaining electrons of the shell, in much the same way that this fact holds for such an electron with respect to the other closed shells of the ion. The only effect that the Coulomb interaction can therefore have on the energy states of the one-hole system is an overall shift of the energy levels.

What happens to the spin-orbit interaction for the one-hole shell? This can be determined by noting that the spin-orbit operator for the filled shell is just

$$\hat{H}_{so}(\text{filled}) = \sum_{i=1}^N \hat{\xi}(\hat{r}_i) \hat{\mathbf{l}}_i \cdot \hat{\mathbf{s}}_i \quad (7.18)$$

and thus the spin-orbit operator for the one-hole system may be written

$$\hat{H}_{so}(\text{hole}) = \hat{H}_{so}(\text{filled}) - \hat{\xi}(\hat{r}) \hat{\mathbf{I}} \cdot \hat{\mathbf{S}} = -\hat{\xi}(\hat{r}) \hat{\mathbf{I}} \cdot \hat{\mathbf{S}} \quad (7.19)$$

with the last step justified by our knowledge that the spin-orbit interaction vanishes for a filled shell. Our grand conclusion is that for the Yb^{3+} ion subject to the spin-orbit interaction, the same spectroscopic terms appear in the energy spectrum as do

for Ce^{3+} except that the order of the energies for the permitted spectroscopic terms is reversed (note Figure 7.4). That the energy splitting is larger between the $j = 7/2$ and the $j = 5/2$ degenerate manifolds for Yb^{3+} than for Ce^{3+} is a consequence of the larger value of the spin-orbit coupling parameter for this ion. Specifically, $\zeta = 2870 \text{ cm}^{-1}$ for Yb^{3+} , thus yielding a splitting of $(7/2)\zeta \sim 10050 \text{ cm}^{-1}$. In turn, referring to Equation (7.15), the larger spin-orbit coupling for Yb^{3+} is a result of the larger Z for its nucleus and hence a larger dU/dr .

The notion of the hole is extended in an obvious way to describe shells with more than one removed electron; the spectroscopy of the pairs Pr^{3+} and Tm^{3+} , Nd^{3+} and Er^{3+} , etc. will exhibit the same number of states, the same number of degenerate manifolds, and the same set of term labels.

7.2.2.4 Praseodymium and thulium, intermediate coupling, and the Russell-Saunders eigenstates

Spectroscopic terms The two-electron system Pr^{3+} and its two-hole analog Tm^{3+} display several new features in the spectroscopy of rare-earth ions. These arise from the appearance of Coulomb repulsion in the Hamiltonian and the restrictions that the Pauli exclusion principle imposes on the allowed two-particle eigenstates within the $4f$ shell. Ignoring for the moment the spin-orbit interaction, we should in particular expect that the Coulomb interaction between the $4f$ electrons will break the symmetry of the unperturbed ground state and partially lift the degeneracy of its $14!/(13!2!) = 91$ states. Given, however, that the Coulomb energy depends only on spatial coordinates and not on the electron spin, the total orbital angular momentum $\hat{\mathbf{L}} = \hat{\mathbf{L}}_1 + \hat{\mathbf{L}}_2$ as well as the total spin $\hat{\mathbf{S}} = \hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_2$ will be separate constants of the motion. (Note that with no spin-spin interactions the spin angular momenta $\hat{\mathbf{S}}_1$ and $\hat{\mathbf{S}}_2$ are likewise constants of the motion.) We are thus led to suggest that eigenstates of the operators $\hat{L}^2$, L_z , $\hat{S}^2$, and S_z be used as basis states with which to calculate the matrix elements of the Coulomb interaction since as far as the angular coordinates are concerned this interaction will be diagonal in this representation. That is, in the language of the addition of quantum angular momenta, the procedure is to separately couple the orbital angular momenta and the spin angular momenta of the two electrons in order to produce a useful set of basis states.

This procedure, known as *Russell-Saunders* or *LS coupling*, is thus particularly appropriate in situations where the Coulomb interaction dominates all other perturbations. The good quantum numbers are L , M_L , S , and M_S . On the other hand, the total angular momentum $\hat{\mathbf{J}} = \hat{\mathbf{L}} + \hat{\mathbf{S}}$ is also a constant of the motion. Under the Russell-Saunders coupling scheme, one therefore has the freedom to choose as a complete set of basis states either $|\alpha LSM_L M_S\rangle$ or $|\alpha LSJM_J\rangle$. With regard to these

two representations, yet restricting attention to the angular degrees of freedom, the Coulomb interaction is diagonal in both. (Note that in our notation for the two representations, we have suppressed the good quantum numbers l_1 , l_2 , s_1 , and s_2 since the latter two are fixed under all circumstances and the former two are fixed (and equal) within a given shell.) The choice of basis states may be dictated best by what subsequent perturbations one wishes to consider beyond the Coulomb interaction. For example, if a Zeeman interaction is of interest, then the $|\alpha L S M_L M_S\rangle$ set of basis states would be chosen. If the spin-orbit interaction is the next most significant perturbation, the $|\alpha L S J M_J\rangle$ basis states are most appropriate since for multiple electrons it turns out that this interaction can be written in terms of the operator $\hat{\mathbf{L}} \cdot \hat{\mathbf{S}}$. Spectroscopic term labels in Russell–Saunders coupling thus assume the form ^{2S+1}L in the first basis set (where they are more properly called *multiplets*) or $^{2S+1}L_J$ in the second, where $L = S, P, D, F$, etc. according to the total orbital angular momentum of the state.

Russell–Saunders coupling works well for the unfilled shells of light atoms where the Coulomb interaction dominates. On the other hand, for heavy elements such as the actinides, the spin-orbit interaction dominates the Coulomb interaction and a different coupling scheme is motivated. Specifically, for a two-electron system, one forms the two angular momenta $\hat{\mathbf{J}}_1 = \hat{\mathbf{L}}_1 + \hat{\mathbf{S}}_1$ and $\hat{\mathbf{J}}_2 = \hat{\mathbf{L}}_2 + \hat{\mathbf{S}}_2$, quantities that are separate constants of the motion under the spin-orbit interaction. Again, so also is the total angular momentum $\hat{\mathbf{J}} = \hat{\mathbf{J}}_1 + \hat{\mathbf{J}}_2$. In this *j–j coupling* scheme, basis states assume the form $|j_1 j_2 m_{j_1} m_{j_2}\rangle$ or $|j_1 j_2 J M_J\rangle$, depending upon one's preference.

Now for the bad news: The Coulomb and spin-orbit interactions are of about the same magnitude in the $4f$ shell of the rare-earth ions. Given that neither Russell–Saunders nor *j–j* coupling admits simultaneous diagonalization of these interactions with respect to the angular coordinates, there is no natural set of angular momentum basis states with which to proceed in solving for the eigenvalues and eigenvectors under these perturbations. The only good quantum numbers are J and M_J . Owing to tradition, however, the Russell–Saunders representation $|\alpha L S J M_J\rangle$ has been adopted both as a means for carrying out the joint diagonalization of the combined spin-orbit and Coulomb Hamiltonian and as a means for identifying the resulting set of $(2J + 1)$ -fold degenerate manifolds. The Russell–Saunders spectroscopic term labels $^{2S+1}L_J$ thus appear in energy-level diagrams of the trivalent rare earths as shown in Figure 7.2. It is important to emphasize, however, that these labels cannot be relied upon to deduce the eigenstates. As we shall see, a given term in this *intermediate coupling* scheme will actually correspond to a mixture of exact Russell–Saunders states all with the same J but with generally different values of L and S . Loosely speaking, the larger the spin-orbit coupling parameter, ζ , the larger the mixing.

We hasten to point out that a historical ambiguity still remains in the labeling of rare-earth terms for ions containing from two to twelve $4f$ electrons. Our discussion above implies that a general first-order eigenstate will be an expansion in terms of pure Russell–Saunders states according to

$$|\alpha, {}^{2S+1}L_J\rangle' = A_0|\alpha, {}^{2S+1}L_J\rangle + \sum_{i \neq 0} A_i |\alpha, {}^{2S_i+1}L_J^i\rangle \quad (7.20)$$

where the prime on the left-hand side indicates that this is an intermediate coupling eigenstate and the A s are just the expansion coefficients for the Russell–Saunders basis states. The expansion coefficients are defined so that in the limit $\zeta \rightarrow 0$, $A_0 \rightarrow 1$ and all the other coefficients vanish. This suggests that we simply set ${}^{2S+1}L_J' = {}^{2S+1}L_J$ in order to label the term. On the other hand, A_0 may not be the largest of the coefficients, especially when the spin–orbit mixing is large. Another convention therefore has it that the Russell–Saunders term label be that corresponding to the largest A in the expansion. In most cases, these two conventions yield the same label so that no ambiguity arises. In those cases where it does arise, our preference is for the latter convention and we adopt it here.

We shall now focus our attention on the spectroscopy of the free Pr^{3+} ion in order to illustrate how the machinery of term labeling and the eigenstate calculation operates in the intermediate coupling regime.

Without the Coulomb and spin-orbit interactions, the first task is to deduce which of all the possible Russell–Saunders states $|\alpha, {}^{2S+1}L_J\rangle$ are actually permitted in light of the Pauli exclusion principle. Such a procedure would not be necessary if the two electrons belonged to different shells, that is, if the two electrons were *inequivalent*. In this case, all spin and angular momentum states are available to each electron within their respective shells and thus all possible Russell–Saunders terms appear when the angular momenta are added according to the Russell–Saunders prescription. For two electrons occupying the same shell, that is, for *equivalent* electrons, this is no longer the case and we should only retain those Russell–Saunders terms that are in compliance with the Pauli principle. The result will be a complete set of basis states with which to construct first-order eigenstates when the Coulomb and spin–orbit perturbations are turned on.

One approach to accomplishing this task is to recognize that the only allowed two-electron states are those that are *anti-symmetric* under exchange of the two electrons. Moreover, since the orbital and spin angular momenta operate in orthogonal subspaces, the problem can be reduced by insisting that if the two-electron state is symmetric in the space coordinates then it must be anti-symmetric in the spin coordinates, and *vice-versa*. When undertaking the final vector addition of $\hat{\mathbf{L}} (= \hat{\mathbf{L}}_1 + \hat{\mathbf{L}}_2)$ and $\hat{\mathbf{S}} (= \hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_2)$, a possible way of ensuring that the proper exchange symmetry is obtained is to add (in the sense of quantum vector addition) a two-electron orbital

angular momentum that is anti-symmetric (symmetric) under electron exchange only to a two-electron spin angular momentum that is symmetric (anti-symmetric) under electron exchange. Such a strategy will succeed, of course, provided that for each allowed L and S obtained upon adding the respective one-electron orbital and spin angular momenta a definite exchange symmetry is displayed. This is actually the case as we shall now see.

First we investigate the exchange symmetry for the spin degrees of freedom. The coupled spin angular momentum states are denoted by $|\frac{1}{2}\frac{1}{2}SM_S\rangle$, which we know to be related to the original decoupled spin states by a linear transformation:

$$|\frac{1}{2}\frac{1}{2}SM_S\rangle = \sum_{m_{s_1}, m_{s_2}} C(\frac{1}{2}m_{s_1}\frac{1}{2}m_{s_2}; SM_S) |\frac{1}{2}m_{s_1}\rangle |\frac{1}{2}m_{s_2}\rangle \quad (7.21)$$

where the numbers $C(\frac{1}{2}m_{s_1}\frac{1}{2}m_{s_2}; SM_S)$ are the less-than-pulchritudinous Clebsch–Gordon coefficients. For the addition of two spin- $\frac{1}{2}$ angular momenta, either $S = 0$ or $S = 1$. Taking advantage of published tables for the Clebsch–Gordon coefficients, we see that for the $S = 0$, $M_S = 0$ coupled spin state

$$|\frac{1}{2}\frac{1}{2}00\rangle = \frac{1}{\sqrt{2}}(|\frac{1}{2}\frac{1}{2}\rangle|\frac{1}{2}-\frac{1}{2}\rangle - |-\frac{1}{2}-\frac{1}{2}\rangle|\frac{1}{2}\frac{1}{2}\rangle) \quad (7.22)$$

which is clearly anti-symmetric under exchange of the two electrons. In Russell–Saunders term notation this state is indicated by $^1S_0$. For the $S = 1$ ($^3S_1$) manifold let us first consider the $M_S = 1$ state:

$$|\frac{1}{2}\frac{1}{2}, S = 1, M_S = 1\rangle = |\frac{1}{2}\frac{1}{2}\rangle|\frac{1}{2}\frac{1}{2}\rangle \quad (7.23)$$

which is clearly symmetric under electron exchange. The remaining $S = 1$ states are likewise symmetric since these states may be computed with the help of the lowering operator $\hat{S}_- \equiv \hat{S}_x - i\hat{S}_y$ and this operator is powerless to affect the exchange symmetry.

A similar procedure yields the exchange symmetry for the coupled two-electron orbital states: First note that the allowed L s are 0, 1, 2, 3, 4, 5, and 6. The corresponding term labels are thus $^1S_0$, $^1P_1$, $^1D_2$, $^1F_3$, $^1G_4$, $^1H_5$, and $^1I_6$. Consider first the $^1I_6$ term. The expansion with respect to the decoupled angular momentum states for the $M_L = 6$ is just

$$|^1I_6, M_L = 6\rangle = |3, 3\rangle|3, 3\rangle \quad (7.24)$$

which is symmetric under electron exchange, as therefore are all the states of the $^1I_6$ term by the same argument as we used for the $^3S_1$ spin states.

Consider now the $^1H_5$ term. For $M_L = 5$ we have

$$|^1H_5, M_L = 5\rangle = \frac{1}{\sqrt{2}}(|3, m_{l_1} = 3\rangle|3, m_{l_2} = 2\rangle - |3, m_{l_1} = 2\rangle|3, m_{l_2} = 3\rangle) \quad (7.25)$$

Table 7.2. *Exchange symmetry for orbital and spin Russell–Saunders terms applicable to the two-electron ion Pr^{3+}*

	Symmetric	Antisymmetric
Orbital terms	$^1S_0, ^1D_2, ^1G_4, ^1I_6$	$^1P_1, ^1F_3, ^1H_5$
Spin terms	$^3S_1$	$^1S_0$

which is anti-symmetric under electron exchange. In fact, all the even- L orbital terms are symmetric and all the odd- L orbital terms are anti-symmetric. To summarize, Table 7.2 shows the results of the foregoing procedure for all the orbital and spin terms.

To obtain the final fully coupled Russell–Saunders terms, we just add – in accord with the vector model for the addition of angular momentum – an orbital term to a spin term of the opposite exchange symmetry. For example, adding the $^1I_6$ orbital term to the $^1S_0$ spin term yields just $^1I_6$. Adding the $^1H_5$ orbital term to the $^3S_1$ spin term yields the series of terms $^3H_6$, $^3H_5$, and $^3H_4$. The complete set of allowed terms is $^1S_0, ^1D_2, ^1G_4, ^1I_6, ^3P_2, ^3P_1, ^3P_0, ^3F_4, ^3F_3, ^3F_2, ^3H_6, ^3H_5$, and $^3H_4$, as the reader may wish to confirm and further compare with Figure 7.4.

Eigenvalues and eigenstates With the complete set of Russell–Saunders basis states for the $4f$ shell of Pr^{3+} in hand, it is now possible to calculate the eigenvalues and eigenvectors that result from the simultaneous diagonalization of the Coulomb and spin–orbit Hamiltonians. First-order degenerate perturbation theory is all that is required to accomplish this task. However, we shall not carry out this calculation with full rigor but merely indicate the flavor of how such calculations are accomplished.

The two interaction Hamiltonians are

$$\hat{H}_C = \frac{e^2}{|\hat{\mathbf{r}}_1 - \hat{\mathbf{r}}_2|} \quad (7.26)$$

and

$$\hat{H}_{SO} = \hat{\xi}_{4f}(\hat{r}_1)\hat{\mathbf{l}}_1 \cdot \hat{\mathbf{s}}_1 + \hat{\xi}_{4f}(\hat{r}_2)\hat{\mathbf{l}}_2 \cdot \hat{\mathbf{s}}_2 \quad (7.27)$$

In practice, however, these Hamiltonians are converted to forms more convenient for the computation of matrix elements. In particular, it can be shown that the

Coulomb Hamiltonian can be recast in the form

$$\hat{H}_C = e^2 \sum_{k=0}^{\infty} \frac{\hat{r}_{<}^k}{\hat{r}_{>}^k} \frac{4\pi}{2k+1} \sum_{q=-k}^k \hat{Y}_{kq}^*(\hat{\theta}_1, \hat{\phi}_1) \hat{Y}_{kq}(\hat{\theta}_2, \hat{\phi}_2) \quad (7.28)$$

where $\hat{Y}_{kq}(\hat{\theta}, \hat{\phi})$ is an operator incarnation of the ordinary spherical harmonic function $Y_{kq}(\theta, \phi)$ and $\hat{r}_{<}$ and $\hat{r}_{>}$ are operators that take the values of the smaller and larger of the radial coordinates r_1 and r_2 during double integration over these latter two variables. This result is a straightforward application of the theory of orthogonal functions and is the algebraic equivalent of Equation (7.26). The advantage of the new form is that the radial and angular variables are now separated, which facilitates calculation of matrix elements between eigenstates in which the radial and angular coordinates are separated – precisely the case we wish to address. Moreover, despite the infinite sum on k indicated in Equation (7.28), the symmetry properties of the spherical harmonics dictate that only the $k = 0, 2, 4$, and 6 terms are nonzero with respect to the f -electron Russell–Saunders angular momentum eigenstates. Now as indicated previously the Coulomb interaction must be diagonal with respect to the Russell–Saunders states, so that within the 91-fold degenerate subspace of the two-electron $4f$ shell the Coulomb matrix elements become

$$\langle \alpha(4f^2)LSJM_J | \hat{H}_C | \alpha(4f^2)L'S'J'M_{J'} \rangle = \delta_{LL'} \delta_{SS'} \delta_{JJ'} \delta_{M_J M_{J'}} \sum_{k=0,2,4,6} F_k Q^k(L) \quad (7.29)$$

where the F_k are radial integrals and the $Q^k(L)$ are matrix elements of the Russell–Saunders states and dependent only upon L . Specifically, the diagonal matrix elements $W_C(\alpha(4f^2), {}^{2S+1}L_J)$ are found to be:

$$W_C(\alpha(4f^2), {}^1S_J) = F_0 + 60F_2 + 198F_4 + 1716F_6 = 37475 \text{ cm}^{-1} \quad (7.30a)$$

$$W_C(\alpha(4f^2), {}^1D_J) = F_0 + 19F_2 - 99F_4 + 715F_6 = 4717 \text{ cm}^{-1} \quad (7.30b)$$

$$W_C(\alpha(4f^2), {}^1G_J) = F_0 - 30F_2 + 97F_4 + 78F_6 = -4487 \text{ cm}^{-1} \quad (7.30c)$$

$$W_C(\alpha(4f^2), {}^1I_J) = F_0 + 25F_2 + 9F_4 + F_6 = 8499 \text{ cm}^{-1} \quad (7.30d)$$

$$W_C(\alpha(4f^2), {}^3P_J) = F_0 + 45F_2 + 33F_4 - 1287F_6 = 9840 \text{ cm}^{-1} \quad (7.30e)$$

$$W_C(\alpha(4f^2), {}^3F_J) = F_0 - 10F_2 - 33F_4 - 286F_6 = -6246 \text{ cm}^{-1} \quad (7.30f)$$

$$W_C(\alpha(4f^2), {}^3H_J) = F_0 - 25F_2 - 51F_4 - 13F_6 = -10633 \text{ cm}^{-1} \quad (7.30g)$$

For Pr^{3+} in the free-ion state, $F_2 = 322 \text{ cm}^{-1}$, $F_4 = 49.4 \text{ cm}^{-1}$, $F_6 = 4.88 \text{ cm}^{-1}$, while we can set $F_0 = 0$ since this quantity contributes only an overall shift of the energies.

With regard to the spin-orbit interaction, it may be written in the form

$$\hat{H}_{so} = \hat{\xi}(\hat{r})\lambda(L, S)\hat{\mathbf{L}} \cdot \hat{\mathbf{S}} = \hat{\xi}(\hat{r})\lambda(L, S)\frac{\hat{J}^2 - \hat{L}^2 - \hat{S}^2}{2} \quad (7.31)$$

where $\lambda(L, S)$ is a geometrical factor that depends only on L and S and

$$\hat{\xi}(\hat{r}) = \frac{\hbar^2}{2m_e^2 c^2} \left[\frac{1}{\hat{r}} \frac{d\hat{U}(\hat{r})}{d\hat{r}} \right] \quad (7.32)$$

The result shown in Equation (7.31) is not obvious; its derivation requires some rather fancy techniques in operator algebra beyond the scope of the present treatment. For proof of Equation (7.31) and a description of the means for computing $\lambda(L, S)$, the reader is referred to Weissbluth (1978). For the recklessly curious, the general matrix element of $\hat{H}_{so}$ for two equivalent f electrons or holes is given by

$$\begin{aligned} & \langle \alpha(4f^2)LSJM_J | \hat{H}_{so} | \alpha(4f^2)L'S'J'M_{J'} \rangle \\ &= 6\sqrt{14}\zeta_{4f}(-1)^{J+S'+S+1} \sqrt{(2L+1)(2L'+1)(2S+1)(2S'+1)} \\ & \quad \times \begin{Bmatrix} L' & S' & J \\ S & L & 1 \end{Bmatrix} \begin{Bmatrix} L & 1 & L' \\ 3 & 3 & 3 \end{Bmatrix} \begin{Bmatrix} S & 1 & S' \\ \frac{1}{2} & \frac{1}{2} & \frac{1}{2} \end{Bmatrix} \delta_{JJ'} \end{aligned} \quad (7.33)$$

where the curly braces denote combinatoric quantities known as Wigner 6- j symbols, values for which may be found in published tables (Rotenberg *et al.* 1969).

Equations (7.29) and (7.31) allow the computation of all 91×91 matrix elements $\langle \alpha(4f^2)LSJM_J | \hat{H}_C + \hat{H}_{so} | \alpha(4f^2)L'S'J'M_{J'} \rangle$. However, the problem is considerably simplified in that the matrix elements are both independent of M_J and diagonal in J . A 13×13 matrix results, indexed by the Russell-Saunders terms. If the terms are grouped by common J , then the array of matrix elements becomes block diagonal and the eigenvalues and eigenvectors may be computed within each block. For example, only a single $J = 3$ term exists, namely $^3F_3$. The first-order eigenvalue is immediately

$$E(^3F'_3) = W_C(\alpha(4f^2), ^3F_J) - \frac{1}{2}\zeta_{4f} = -6625 \text{ cm}^{-1} \quad (7.34)$$

where for $\text{Pr}^{3+}\zeta_{4f} = 758 \text{ cm}^{-1}$. The first-order eigenvector in the intermediate coupling regime remains a seven-fold degenerate pure Russell-Saunders term:

$$|\alpha(4f^2)^3F'_3\rangle = |\alpha(4f^2)^3F_3\rangle \quad (7.35)$$

Similarly, the only $J = 5$ term is $^3H_5$ and the eigenvalue is -11012 cm^{-1} .

On the other hand, the $J = 2$ block is a 3×3 array:

$$\begin{pmatrix} W_C(\alpha(4f^2), {}^1D_2) & \frac{3}{\sqrt{2}}\zeta_{4f} & -\sqrt{6}\zeta_{4f} \\ \frac{3}{\sqrt{2}}\zeta_{4f} & W_C(\alpha(4f^2), {}^1P_2) + \frac{\zeta_{4f}}{2} & 0 \\ -\sqrt{6}\zeta_{4f} & 0 & W_C(\alpha(4f^2), {}^1F_2) - 2\zeta_{4f} \end{pmatrix} \quad (7.36)$$

The eigenvectors and eigenvalues of this block are

$$|\alpha(4f^2)^3P_2'\rangle = 0.963|\alpha(4f^2)^3P_2\rangle + 0.269|\alpha(4f^2)^1D_2\rangle - 0.027|\alpha(4f^2)^3F_2\rangle \quad (7.37a)$$

$$|\alpha(4f^2)^3F_2'\rangle = 0.989|\alpha(4f^2)^3F_2\rangle + 0.145|\alpha(4f^2)^1D_2\rangle - 0.013|\alpha(4f^2)^3P_2\rangle \quad (7.37b)$$

and

$$|\alpha(4f^2)^1D_2'\rangle = 0.952|\alpha(4f^2)^1D_2\rangle - 0.270|\alpha(4f^2)^3P_2\rangle - 0.144|\alpha(4f^2)^3F_2\rangle \quad (7.37c)$$

with eigenvalues 10667 cm^{-1} , -8035 cm^{-1} , and 4541 cm^{-1} , respectively.

The complete set of eigenvalues and eigenvectors for free Pr^{3+} are shown in Table 7.3. In the table, the energies have all been shifted by an additive constant so that the lowest-lying term is at the zero of energy. Figure 7.7 shows the energy-level diagram as calculated here compared with ones based on measurements of the free-ion energies and energies measured for a LaF_3 host.

Our discussion of the calculation of energies and eigenstates for Pr^{3+} may have left the reader with the impression that the F_k and ζ_{4f} may be obtained to the required accuracy within the Hartree–Fock approximation. In practice, however, this is not the case, and instead the four radial integrals are considered as adjustable parameters with which the measured spectrum is rationalized. Often, starting values for these parameters are obtained employing simple hydrogenic wave functions; final energies so obtained are typically within 100 cm^{-1} of their experimental values. In instances where even greater accuracy is desired, however, additional adjustable parameters may be introduced by considering the Coulomb and spin–orbit interactions in higher orders of perturbation theory, or lesser additional perturbations such as spin–spin and spin–other-orbit interactions. The most important correction is called the configuration interaction; it results from nonzero Coulomb matrix elements between Russell–Saunders states belonging to the $4f$ shell and a *different* shell. The configuration interaction does not reduce the degeneracy of any of the

Table 7.3. *Energies, term labels, and eigenstates as expansions in Russell–Saunders terms for Pr^{3+} in the intermediate coupling regime*

Energy (cm^{-1})	Term	Eigenstate
0	$^3H_4$	$-0.986 ^3H_4\rangle - 0.163 ^1G_4\rangle + 0.029 ^3F_4\rangle$
2123	$^3H_5$	$ ^3H_5\rangle$
4348	$^3H_6$	$-0.998 ^3H_6\rangle + 0.054 ^1I_6\rangle$
5100	$^3F_2$	$0.989 ^3F_2\rangle + 0.146 ^1D_2\rangle - 0.013 ^3P_2\rangle$
6510	$^3F_3$	$ ^3F_3\rangle$
6958	$^3F_4$	$0.800 ^3F_4\rangle - 0.588 ^1G_4\rangle + 0.121 ^3H_4\rangle$
9941	$^1G_4$	$-0.792 ^1G_4\rangle - 0.599 ^3F_4\rangle + 0.117 ^3H_4\rangle$
17677	$^1D_2$	$0.952 ^1D_2\rangle - 0.270 ^3P_2\rangle - 0.144 ^3F_2\rangle$
21684	$^1I_6$	$0.998 ^1I_6\rangle + 0.054 ^3H_6\rangle$
21976	$^3P_0$	$-0.996 ^3P_0\rangle - 0.091 ^1S_0\rangle$
22596	$^3P_1$	$ ^3P_1\rangle$
23802	$^3P_2$	$0.963 ^3P_2\rangle + 0.269 ^1D_2\rangle - 0.027 ^3F_2\rangle$
50851	$^1S_0$	$0.996 ^1S_0\rangle - 0.091 ^3P_0\rangle$

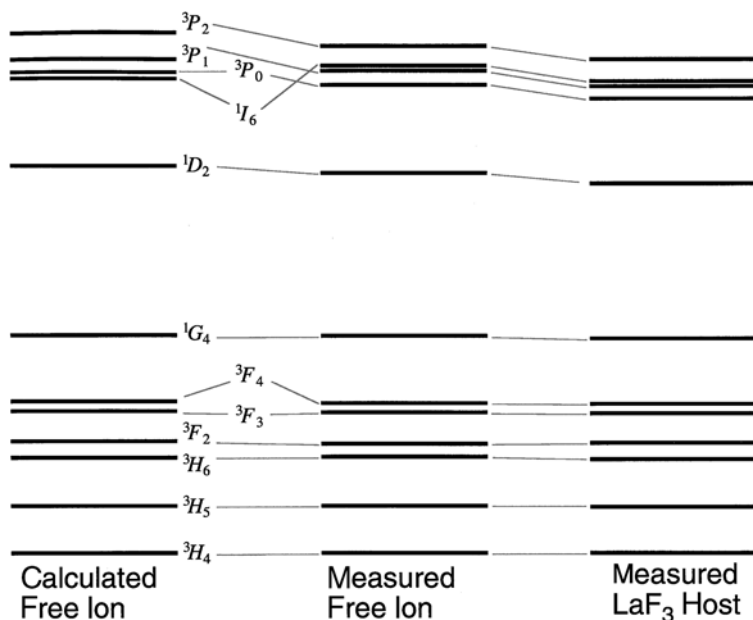


Figure 7.7: Energy-level diagram for a free Pr^{3+} ion as calculated in the text compared with values measured for the free ion (NIST, 1998) and in a LaF_3 host (Carnall *et al.*, 1977). Energies for the latter levels are averages within each crystal field multiplet.

Table 7.4. Permitted Russell–Saunders terms for trivalent rare earth elements. Allowed J s are suppressed, but may be surmised by angular-momentum addition of the listed L and S values. Subscripts indicate the number of times the given Russell–Saunders term label occurs in the full set of those allowed

Configuration	Allowed $2s+1(L)$ terms
$4f, 4f^{13}$	$2F$
$4f^2, 4f^{12}$	$1(SDGI)$
$4f^3, 4f^{11}$	$2(PD_2F_2G_2H_2IKL)$
$4f^4, 4f^{10}$	$1(S_2D_4FG_4H_2I_3KL_2N)$
$4f^5, 4f^9$	$2(P_4D_5F_7G_6H_7I_5K_5L_3M_2NO)$
$4f^6, 4f^8$	$1(S_4PD_6F_4G_8H_4I_7K_3L_4M_2N_2Q)$
$4f^7$	$2(S_2P_5D_7F_{10}G_{10}H_9I_9K_7L_5M_4N_2OQ)$
	$3(PFH)$
	$4(SDFGI)$
	$3(P_3D_2F_4G_3H_4I_2K_2LM)$
	$4(SP_2D_3F_4G_4H_3I_3K_2LM)$
	$3(P_6D_5F_9G_7H_9I_6K_6L_3M_3NO)$
	$4(S_2P_2D_6F_5G_7H_5I_5K_3L_3MN)$
	$5(SDFGI)$
	$6(PFH)$
	$5(SPD_3F_2G_3H_2I_2KL)$
	$6(PDFGHI)$
	$7F$
	$8S$

Table 7.5. *Radial integrals F_k and spin-orbit coupling parameter ζ as calculated in the relativistic Hartree-Fock approximation.*
(After Carnall *et al.* (1977).)

Configuration	RE ³⁺	$F_2(\text{cm}^{-1})$	$F_4(\text{cm}^{-1})$	$F_6(\text{cm}^{-1})$	$\zeta(\text{cm}^{-1})$
$4f^1$	Ce ³⁺				696.41
$4f^2$	Pr ³⁺	438	56.8	6.05	820.22
$4f^3$	Nd ³⁺	456	59.2	6.30	950.51
$4f^4$	Pm ³⁺	473	61.4	6.54	1091.46
$4f^5$	Sm ³⁺	489	63.5	6.76	1243.60
$4f^6$	Eu ³⁺	505	65.5	6.97	1407.71
$4f^7$	Gd ³⁺	520	67.5	7.18	1584.45
$4f^8$	Tb ³⁺	534	69.4	7.38	1774.46
$4f^9$	Dy ³⁺	549	71.2	7.58	1998.44
$4f^{10}$	Ho ³⁺	563	73.0	7.77	2197.06
$4f^{11}$	Er ³⁺	577	74.8	7.96	2431.00
$4f^{12}$	Tm ³⁺	590	76.5	8.15	2680.97
$4f^{13}$	Yb ³⁺				2947.69

Russell-Saunders terms, nor does it mix different Russell-Saunders terms, so our term labels remain the same, but an additional nine adjustable parameters become available with which to fit measured spectra. The practice of treating radial integrals as adjustable parameters is a common theme of rare-earth spectroscopy owing to uncertainties in the radial wave functions, particularly when the ion is in the presence of a crystal field.

7.2.2.5 The other trivalent lanthanides and Hund's rules

By now the reader should have a good feeling for the nature of the lowest-lying quantum states of the trivalent rare earths and the means by which their eigenvalues and eigenvectors are determined. For the ions with three or more electrons or holes, however, straightforward yet tedious techniques (see Weissbluth (1978)) exist for determining the allowed terms so we will not pursue this task here. Suffice to say that with little special knowledge such methods are readily implemented on a computer. At a more sophisticated level, group theoretic methods may be applied to determining the allowed terms for $N > 2$ electrons. A list of the allowed terms for the complete set of f^N configurations is given in Table 7.4. As for the computation of the eigenvalues and eigenvectors, in lowest order the Coulomb interaction yields a different set of the four Coulomb radial integrals $F_{0,2,4,6}$ and a different spin-orbit coupling parameter ζ_{4f} for each rare earth. Table 7.5 lists calculated values of the F coefficients as calculated by Carnall *et al.* (1977). For any number of electrons

or holes, the calculation of the eigenvalues and eigenvectors proceeds identically to the case of Pr^{3+} .

There is, however, an easy way to determine the term label of the ground state. This is by application of Hund's rules, a set of three empirical results broadly applicable to atoms, ions and even nuclei. The rules are:

- (1) Electron spins are aligned so as to produce the largest S consistent with the Pauli principle.
- (2) Of all the terms with maximum S , the one with the largest L will be the lowest in energy.
- (3) For configurations consisting of electrons in a less than half-filled shell, spin-orbit splitting will yield a ground state possessing the smallest J . For a more than half-filled shell, the term with the largest J will be lowest in energy. For the exactly half-filled shell, rules 1 and 2 yield a unique term.

For example, Sm^{3+} , with five f electrons, yields a maximum S of $5/2$ when all five spins are parallel. Since the spins are parallel, the electrons must occupy different orbital states; in this case, a set of m_l values that yields a maximum value for M_L is $\{3, 2, 1, 0, -1\}$. This choice therefore indicates a maximum L of 5. Adding the spin and the orbital angular momenta yields allowed J s of $15/2, 13/2, 11/2, 9/2, 7/2$, and $5/2$, so that the ground state must be ${}^6H_{5/2}$. Moreover, since the spin-orbit splitting is an increasing function of J for fixed L and S , we can predict the order of the higher-lying ${}^6H$ terms above the ground state. In fact this is trivially so for any ${}^{2S+1}L$ multiplet in a less than half-filled shell; a trend readily seen in Figure 7.4. On the other hand, the ground state of the five-hole ion Dy^{3+} must be ${}^6H_{15/2}$, with subsequent excited states decreasing in J .

This concludes our discussion of the static electronic structure of trivalent rare-earth ions in free space. Despite the organization of N -electron states into sums of pure Russell-Saunders terms, it should always be kept in mind that underlying the somewhat abstract symbolism are simple superpositions of single-electron product states all with the same orbital angular momentum $l = 3$. Owing to the hydrogenic selection rule of $\Delta l = \pm 1$, no electric-dipole-allowed transitions therefore exist within the $4f$ shell. In the next chapter, we demonstrate how such transitions nevertheless arise when the ions act as impurities in transparent host media.

7.3 THE JUDD-OFELT EXPRESSION FOR OPTICAL INTENSITIES

The development so far has emphasized the energy-level structures of rare-earth ions as they would be observed for free ions in vacuum. As was qualitatively argued in Section 7.1, the levels so obtained are a good approximation to those observed in the solid state but for the weak perturbative influence of the crystal field. We

now address the affects of the latter perturbation and show how electric-dipole transitions become possible within the $4f$ shell of rare-earth ions.

7.3.1 Basic formulation

The asymmetric charge distributions affiliated with the unfilled $4f$ shell of rare-earth ions imply that a static perturbation lacking spherical symmetry will lift the $(2J + 1)$ -fold degeneracy of each J manifold. Perturbations of this type arise naturally for dilute liquid or solid-state solutions. While an explicit calculation of the new energy levels will reveal the degree to which degeneracies are lifted in the presence of the perturbation, ultimately this is determined by the crystal symmetry and therefore may be deduced through the methods of group theory. For the sake of brevity, however, we will bypass the development of these methods and move directly to a description of how one calculates the electric-dipole transition matrix elements. Nevertheless, the value of group theory in revealing quantum structure and processes cannot be understated; readers wishing to deepen their grasp of condensed-phase rare-earth spectroscopy should therefore make a study of group theoretic techniques their first priority (Di Bartolo, 1968; Wherrett, 1986). As for level degeneracies in amorphous host materials or for crystals of sufficiently low symmetry, terms for ions with an odd number of electrons we now know split into a sequence of *Kramers doublets*, with each doublet corresponding to the $\pm M_J$ states, respectively. In even-electron systems, the $M_J = 0$ state appears as at least one of perhaps several possible singlets along with possible Kramers doublets. In any case, experiment reveals that the crystal field splitting is small compared with the energy spacing between terms. Degenerate perturbation theory therefore applies to the calculation of the crystal field levels and eigenstates. Figure 7.8 shows the relative splitting due to the crystal field compared with the spin–orbit and Coulomb splittings for the lowest two multiplets of Er^{3+} .

However, for our purposes the more important consequence of the crystal field is that it leads to electric-dipole transitions within the $4f$ shell. Recall our model of this perturbation from Section 7.1.2: We idealize the field as purely electrostatic in character and further insist that the electrostatic potential be a solution of the Poisson equation $\nabla^2 \Phi = 0$. A general solution to this equation may be written as an expansion in spherical harmonics:

$$\Phi(\mathbf{r}) = \left(-\frac{1}{e}\right) \sum_{k,q} \bar{B}_{kq} r^k Y_{kq}(\theta, \phi) \quad (7.38)$$

where $r = 0$ is located at the nucleus of the rare-earth impurity ion and the $\bar{B}_{kq}$ are so-called crystal field parameters. This form is motivated by our recognition

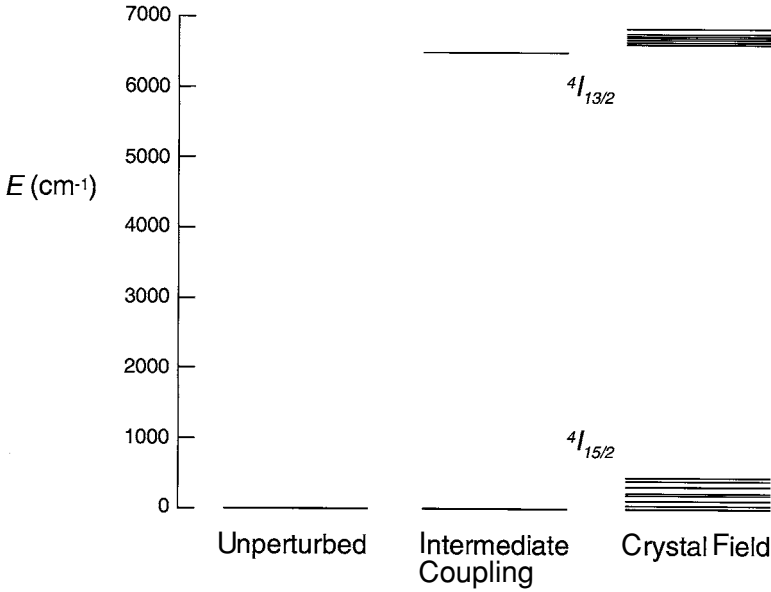


Figure 7.8: The effect of the crystal field for a LaF_3 host in splitting the lowest two J manifolds of Er^{3+} compared with the splittings obtained by virtue of the spin-orbit and Coulomb interactions. In all cases the ground state is referenced to $E = 0$.

that the matrix elements that we will soon need will involve angular momentum eigenstates. Writing the electrostatic potential so that it is separate in the radial and angular coordinates will facilitate this calculation. With the potential in hand, the interaction Hamiltonian for an N -electron ion thus becomes

$$\hat{V} = -e \sum_{i=1}^N \Phi(\mathbf{r}_i) = \sum_{i=1}^N \sum_{k,q} \bar{B}_{kq} \hat{r}_i^k \hat{Y}_{kq}(\hat{\theta}_i, \hat{\phi}_i) \quad (7.39)$$

In addition to splitting the degeneracy of terms lying within the $4f^N$ configuration, this interaction will mix the $4f^N$ eigenstates with states appearing in higher-energy configurations. For example, owing to its proximity in energy to the $4f^N$ configuration, the largest contributions will come from $4f^{N-1}5d^1$. But what is most important about the crystal-field-induced admixture of the $4f^N$ and $4f^{N-1}5d^1$ configurations is that *even-parity* states are combined with the exclusively *odd-parity* states of the $4f$ shell. That is, although the crystal field states are mainly of f character, the small degree of added d character makes parity no longer a good quantum number and in particular makes $\Delta l = \pm 1$ electric-dipole transitions possible within the $4f$ shell. We can therefore immediately state that it is only the *odd-parity* components of the crystal field interaction $\hat{V}$ that introduce into the perturbed eigenfunctions the *even-parity* states required for electric-dipole transitions. Crystalline hosts lacking

inversion symmetry possess these odd-parity components; we therefore expect that low-symmetry hosts will exhibit the strongest absorption and emission lines.

Applying first-order nondegenerate perturbation theory to the states of the $4f^N$ configuration leads to the crystal-field-perturbed eigenstates

$$\begin{aligned} |\alpha(4f^N), SLJM_J\rangle &= |\alpha(4f^N), SLJM_J\rangle \\ &+ \sum_{\alpha', S', L', J', M_{J'}} \frac{\langle \alpha(4f^N), SLJM_J | \hat{V} | \alpha', S' L' J' M_{J'} \rangle}{E_{4f^N}(SLJM_J) - E_{\alpha'}(S' L' J' M_{J'})} |\alpha', S' L' J' M_{J'}\rangle \end{aligned} \quad (7.40)$$

In this expression, the symbol α' denotes the purely radial portion of wave functions belonging to a general configuration of the ion, as well as any other incidental quantum numbers. These include same-parity states from within the $4f^N$ configuration as well as states of both parities belonging to higher-energy configurations such as $4f^{N-1}nl^1$, $4f^{N-2}nl^2$, $4f^{N-2}nl^1n'l'^1$, and so on. We emphasize again that, owing to the odd parity of the electric-dipole-moment operator, it is the opposite-parity states found only in the higher-energy configurations that will give rise to electric-dipole transitions between states of the $4f^N$ configuration. As mentioned above, foremost among these is the $4f^{N-1}5d^1$ configuration. Note that Equation (7.40) shows the expansion for a perturbed Russell–Saunders term and not for the full intermediate-coupling eigenstates constructed in the previous chapter. For now, we will consider only electric-dipole matrix elements between the perturbed Russell–Saunders terms and later perform the necessary sum over those Russell–Saunders basis states appearing in the intermediate-coupling solutions.

We seek electric-dipole matrix elements between an initial crystal field level $|\bar{i}\rangle$ and a final level $|\bar{f}\rangle$ of the form $\langle \bar{i} | \hat{\mu} | \bar{f} \rangle$, where in terms of the position operators $\hat{\mathbf{r}}_i$ for the N $4f$ electrons

$$\hat{\mu} = -e \sum_{i=1}^N \hat{\mathbf{r}}_i \quad (7.41)$$

is the electric-dipole-moment operator.

With the help of the expansion Equation (7.40), the matrix elements then become

$$\begin{aligned} \langle \bar{i} | \hat{\mu} | \bar{f} \rangle &= \langle \alpha(4f^N), S^i L^i J^i M_{Ji} | \hat{\mu} | \alpha(4f^N), S^f L^f J^f M_{Jf} \rangle \\ &= \sum_{\alpha', S', L', J', M_{J'}} \frac{\langle \alpha(4f^N), S^i L^i J^i M_{Ji} | \hat{V} | \alpha', S' L' J' M_{J'} \rangle \langle \alpha(4f^N), S^f L^f J^f M_{Jf} | \hat{\mu} | \alpha', S' L' J' M_{J'} \rangle}{E_{4f^N}(S^i L^i J^i M_{Ji}) - E_{\alpha'}(S' L' J' M_{J'})} \\ &+ \sum_{\alpha', S', L', J', M_{J'}} \frac{\langle \alpha(4f^N), S^f L^f J^f M_{Jf} | \hat{V} | \alpha', S' L' J' M_{J'} \rangle \langle \alpha(4f^N), S^i L^i J^i M_{Ji} | \hat{\mu} | \alpha', S' L' J' M_{J'} \rangle}{E_{4f^N}(S^f L^f J^f M_{Jf}) - E_{\alpha'}(S' L' J' M_{J'})} \end{aligned} \quad (7.42)$$

Unfortunately, the full execution of this calculation requires extensive use of advanced theorems in operator algebra that reach well beyond the scope of our

treatment here. However, the reader should be aware of two apparently drastic approximations leading to the result we soon quote. First, it is assumed that the energies $E_{\alpha'}(S'L'J'M_{J'})$ are degenerate with respect to the quantum numbers S' , L' , J' , and $M_{J'}$ (but not n and l). The second approximation is that the energies of the initial and final states are the same, that is, that $E_{4f^N}(S^iL^iJ^iM_{J^i}) = E_{4f^N}(S^fL^fJ^fM_{J^f})$. Given that the nearest configuration ($4f^{N-1}5d^1$) that will contribute to the dipole matrix element ranges in energy from $50\,000\text{ cm}^{-1}$ to $100\,000\text{ cm}^{-1}$ above the $4f^N$ configuration, and, moreover, that the transitions of interest will be between states separated by about $25\,000\text{ cm}^{-1}$, we see that these approximations exert considerable stress on the final result. The benefit of this simplification, however, is that the two sums in Equation (7.42) can be combined into one; in this sum, symmetry happens to dictate that only three terms are nonzero.

Since we will be most concerned with laser transitions between two different crystal-field-split manifolds, rather than between individual crystal field states within these manifolds, the results of the dipole-moment calculation must be summed over M_J and $M_{J'}$. To see exactly how this goes, let us write the oscillator strength for transitions between a degenerate initial manifold $|I\rangle$ and a degenerate final manifold $|F\rangle$, where capital letters are used to distinguish these manifolds from individual crystal field member states $|i\rangle$ and $|f\rangle$. In particular, the oscillator strength is given by

$$f_{I \rightarrow F} = \frac{2m_e\omega_0 g_F \langle |\tilde{\mu}_{IF}|^2 \rangle}{3\hbar e^2} \quad (7.43)$$

where ω_0 is the frequency of the transition, g_F is the degeneracy of the final-state manifold, and the angle brackets indicate the *average* squared dipole-moment matrix element for all transitions $i \rightarrow f$ between member crystal field levels of the two manifolds, respectively. Note that the populations in the individual sublevels of the initial manifold are assumed equal, and further that a disordered system is assumed, so we specialize to this case. We then have

$$\langle |\tilde{\mu}_{IF}|^2 \rangle = \frac{1}{g_I g_F} \sum_i \sum_f |\tilde{\mu}_{if}|^2 = \frac{1}{(2J+1)(2J'+1)} \sum_{M_J=-J}^J \sum_{M_{J'}=-J'}^{J'} |\langle i | \hat{\mu} | f \rangle|^2 \quad (7.44)$$

so that the oscillator strength becomes

$$f_{I \rightarrow F} = \frac{2m_e\omega_0}{3\hbar e^2(2J+1)} \sum_{M_J=-J}^J \sum_{M_{J'}=-J'}^{J'} |\langle i | \hat{\mu} | f \rangle|^2 \quad (7.45)$$

7.3.2 The Judd–Ofelt expression for the oscillator strength

B. R. Judd (1962) and G. S. Ofelt (1962) independently evaluated the double sum that appears in Equation (7.45) by using Equation (7.42) for the matrix elements between individual crystal field sublevels; they obtained the result

$$f_{I \rightarrow F} = \frac{2m_e \omega_0}{3\hbar e^2 (2J + 1)} \sum_{\lambda=2,4,6} \Omega_\lambda |\langle \alpha(4f^N), S^I L^I J^I \| \hat{\mathbf{U}}_\lambda \| \alpha(4f^N), S^F L^F J^F \rangle|^2 \quad (7.46)$$

where Ω_λ is a so-called Judd–Ofelt intensity parameter and the squared quantity is known as a reduced matrix element. We now explain the meaning of these new quantities.

First, the Judd–Ofelt parameters are given by an expression of the form

$$\Omega_\lambda \propto \sum_{kq} [\text{crystal field radial integral}]_{kq} \left[\sum_{\alpha'} \frac{\text{Hartree–Fock radial} \times \text{angular integrals}}{E_{4f^N}(S^I L^I J^I) - E_{\alpha'}} \right]_{k\lambda} \quad (7.47)$$

That is, the first term is a (squared) matrix element between the radial wave functions $|\alpha(4f^N)\rangle$ and $|\alpha'\rangle$ of the purely radial portions of the crystal field potential operator $\hat{V}$, namely

$$|\langle \alpha(4f^N) | \bar{B}_{kq} \hat{r}^k | \alpha' \rangle|^2 \quad (7.48)$$

The second term involves Hartree–Fock single-electron orbitals lying both within and above the $4f^N$ configuration. In particular, products of radial factors of the form

$$\langle \alpha(4f^N) | \hat{r} | \alpha' \rangle \quad (7.49)$$

and single-orbital angular factors of the form

$$\langle f | \hat{Y}_{km'} | l \rangle \quad (7.50)$$

appear. (The quantum number m' is a dummy index that has no impact on the final result.) The radial portion owes its heritage to our expression for the dipole-moment operator Equation (7.41), while the angular integrals arise from both the angular portion of the dipole-moment operator and that of the crystal field potential. These angular integrals can be calculated exactly depending as they do on only angular momentum eigenstates. The radial integrals are much more uncertain, especially if multiplied by the crystal field coefficients $\bar{B}_{kq}$. The latter uncertainties are what require us to simply take the Ω_λ s as adjustable parameters. That only three such parameters occur is a consequence of the symmetries of the angular momentum eigenstates and the spherical harmonics with which we chose to express the operators for the electric-dipole moment and the crystal field potential. Note that in the

second factor of Equation (7.47), the energy denominator is smallest for configurations nearest in energy to $4f^N$ and thus these configurations contribute the most to Ω_λ .

Now for the punch line: Nothing in our discussion of the factors contributing to Ω_λ depended at all on the exact Russell–Saunders term serving as either an initial or a final state. Only factors involving single-orbital radial and angular matrix elements appear in the expression for Ω_λ . Hence, these quantities are fixed once and for all for a given rare-earth ion within a given host, at least subject to the approximations discussed above. The dependence of the Ω_λ s on the host material enters through the values of the crystal field coefficients $\bar{B}_{kq}$, while the dependence on the particular rare-earth ion of interest results from the Z -dependence of the Hartree–Fock radial wave functions as one moves across the lanthanide series. It might be said that the Ω_λ s only express the nature of the crystal field plus that of the pristine $4f$ shell unperturbed by Coulomb or spin-orbit interactions.

Spectroscopic measurements reveal that for a given rare-earth ion the Ω_2 parameter varies by as much as a factor of several hundred as one goes from host to host while the Ω_4 and Ω_6 parameters vary by perhaps a factor of 2. Physical interpretations of the Judd–Ofelt parameters have been offered as explanations for these trends: The Ω_2 parameter is believed to express the degree of covalency in the ion-lattice interaction, while the Ω_4 and Ω_6 parameters have been linked to the host material's viscosity (Reisfeld and Jørgensen, 1987).

As for the reduced matrix element $\langle \alpha(4f^N), S^I L^I J^I \| \hat{U}_\lambda \| \alpha(4f^N), S^F L^F J^F \rangle$, rather more explanation is required. The most important fact about this term is that it can be calculated exactly, at least for pure Russell–Saunders terms acting as initial and final states. In practice, however, we must keep in mind that the intermediate-coupling eigenstates of the rare-earth ions are superpositions of such pure Russell–Saunders terms, albeit all of the same J , and that therefore the reduced matrix element should in general be replaced by the double sum (suppressing the quantum numbers $\alpha(4f^N)$)

$$\begin{aligned} & \langle S^{I'} L^{I'} J^{I'} \| \hat{U}_\lambda \| S^{F'} L^{F'} J^{F'} \rangle \\ &= \sum_{S^I, L^I} \sum_{S^F, L^F} A_I^*(S^I, L^I) A_F(S^F, L^F) \langle S^I L^I J^I \| \hat{U}_\lambda \| S^F L^F J^F \rangle \quad (7.51) \end{aligned}$$

In this expression, $A(S, L)$ is just the expansion coefficient for the $^{2S+1}L_J$ Russell–Saunders basis term as it appears in the intermediate-coupling eigenstate. Note that this incorporation of the more correct eigenstates has no effect on the Ω_λ s because as just discussed, these quantities are independent of the specific Russell–Saunders terms at hand. Despite this added complication, the good news is that insofar as the $A(S, L)$ s are determined almost exclusively by the Coulomb and spin-orbit

interactions of the free ion, and are only weakly affected by the crystal field, free-ion values for these coefficients are often adequate for calculation of the reduced matrix element. To a better approximation, the expansion coefficients may be taken as those determined for a given material and then applied to related materials.

We can now see the real value of our result for the oscillator strength, Equation (7.46): We have managed to completely separate the effects of the crystal field from the specific intermediate-coupling eigenstates between which we seek the oscillator strength. The reduced matrix elements are only weakly dependent on the host material and may be looked up in published tables (Nielson and Koster, 1964, Carnall *et al.*, 1968, Carnall *et al.*, 1977, Cowan, 1981). All of the host dependence appears in the Judd–Ofelt parameters, Ω_λ . One can therefore perform a room-temperature absorption measurement from which one then calculates the absorption oscillator strengths for transitions from the ground manifold to at least three excited J manifolds. With reduced matrix elements obtained from the literature (or by direct calculation), Equation (7.46) may be applied to fit the measured oscillator strengths with the Ω_λ s taken as adjustable parameters. With possession of the Judd–Ofelt parameters plus the reduced matrix elements, Equation (7.46) can then be used to estimate the oscillator strength for a transition between *any* pair of the ion’s J manifolds. A simple absorption measurement is thus of great value in identifying or comparing possible laser transitions, despite the fairly serious approximations involved in arriving at the Judd–Ofelt result. Prospective host materials may be readily surveyed, while for a given host and impurity ion those transitions most suitable for laser applications can be readily identified without the need for optical pumping experiments. As will be seen later, such measurements are also useful in quantifying radiationless energy transfer processes between both similar and dissimilar rare-earth ions.

But what exactly is a reduced matrix element and what exactly is $\hat{\mathbf{U}}_\lambda$? The former comes about through the famous Wigner–Eckart theorem, which concerns a broad class of quantities $\hat{T}_{kq}$ known as irreducible tensor operators. The number k is a nonnegative integer called the rank of the tensor, while q is an integer that ranges in value from $-k$ to k . Good examples of such operators are the familiar spherical harmonics $\hat{Y}_{kq}(\hat{\theta}, \hat{\phi})$; another example is simply the position operator $\hat{\mathbf{r}}$ for a single electron, a rank-1 irreducible tensor provided that we make the identification $\hat{T}_{1,-1} = (\hat{x} - i\hat{y})/\sqrt{2}$, $\hat{T}_{1,0} = \hat{z}$, and $\hat{T}_{1,1} = (\hat{x} + i\hat{y})/\sqrt{2}$. The Wigner–Eckart theorem then states that a general matrix element for an irreducible tensor operator is given by

$$\langle \alpha J M | \hat{T}_{kq} | \alpha' J' M' \rangle = (-1)^{J-M} \begin{pmatrix} J & k & J' \\ -M & q & M' \end{pmatrix} \langle \alpha J || \hat{\mathbf{T}}^{(k)} || \alpha' J' \rangle \quad (7.52)$$

where the matrix like construction enclosed in parentheses, called the Wigner 3- j

symbol, is simply related to the Clebsch–Gordan coefficients according to

$$\langle j_1 j_2 m_1 m_2 | j_1 j_2 J M \rangle = (-1)^{j_1 - j_2 + M} \begin{pmatrix} j_1 & j_2 & J \\ -m_1 & m_2 & M \end{pmatrix} \quad (7.53)$$

The 3- j symbol vanishes unless the bottom row of numbers sums to zero and the top row of numbers satisfy the triangle inequality $|j_1 - j_2| \leq J \leq j_1 + j_2$, as we would expect from what we know about the addition of angular momenta. The meaning of the theorem is that provided one knows the reduced matrix element $\langle \alpha J \| \hat{\mathbf{T}}^{(k)} \| \alpha' J' \rangle$, which depends on only α, α', J, J' , and k , then any matrix element may be readily calculated by evaluating the 3- j symbol (plus the $(-1)^{J-M}$ phase factor). However, it is important to recognize that through the 3- j symbol the dependence of the matrix element on q, M , and M' is purely geometrical: if one were to rotate the coordinate axes in some arbitrary way, the 3- j symbol would embody the full effect of this transformation, at least as long as the reduced matrix element itself is unchanged by such a coordinate transformation. This is certainly the case as regards J and J' ; coordinate rotations do not change the spin of a particle nor do they change a p orbital into a d orbital. It is also the case that k is unchanged by rotations; it is part of what *defines* irreducibility for the operator $\hat{T}_{kq}$. For these reasons, it is often said that the reduced matrix element carries exclusively the physical content of the matrix element while its pre-factors in Equation (7.53) carry exclusively the geometrical content. The actual value of the reduced matrix element is found by writing

$$\langle \alpha J \| \hat{\mathbf{T}}^{(k)} \| \alpha' J' \rangle = \frac{\langle \alpha J M | \hat{T}_{kq} | \alpha' J' M' \rangle}{(-1)^{J-M} \begin{pmatrix} J & k & J' \\ -M & q & M' \end{pmatrix}} \quad (7.54)$$

and – provided the 3- j symbol is nonzero – evaluating the right-hand side with any convenient choice of q, M , and M' .

A few more comments should help make the concept of an irreducible tensor operator more comfortable. We suggested above that the spherical harmonics $\hat{Y}_{kq}(\hat{\theta}, \hat{\phi})$ comprise a set of such operators; for each rank k , there thus exist $2q + 1$ tensor components. What makes the spherical harmonics “irreducible” is that under a coordinate rotation that takes $\{x, y, z\} \rightarrow \{x', y', z'\}$ and therefore $\{\hat{\theta}, \hat{\phi}\} \rightarrow \{\hat{\theta}', \hat{\phi}'\}$, the spherical harmonics in the new coordinates can be related to those in the old coordinates by the linear transformation

$$\hat{Y}_{kq'}(\hat{\theta}', \hat{\phi}') = \sum_{q=-k}^k D_{q'q}^{(k)} \hat{Y}_{kq}(\hat{\theta}, \hat{\phi}) \quad (7.55)$$

That is, the coordinate rotation only mixes spherical harmonics of the same k . A general irreducible tensor operator is therefore one that obeys the above transformation

law under rotation of the coordinate system. We see that the position operator $\hat{\mathbf{r}}$ is trivially such an operator, as are $\hat{\mathbf{L}}$, $\hat{\mathbf{S}}$, and $\hat{\mathbf{J}}$, or indeed any vector operator $\hat{\mathbf{V}}$ provided that the tensor components are defined in terms of the cartesian components according to

$$\hat{\mathbf{V}} = (\hat{V}_{-1}, \hat{V}_0, \hat{V}_1) \equiv \left(\frac{\hat{V}_x - i\hat{V}_y}{\sqrt{2}}, \hat{V}_z, \frac{\hat{V}_x + i\hat{V}_y}{\sqrt{2}} \right) \quad (7.56)$$

Similar identifications can be made for cartesian tensors of higher rank. For example, while the most general second-rank cartesian tensor in three dimensions is not irreducible, it can be broken down into a sum of three irreducible pieces: a constant tensor with all elements equal (irreducible tensor of rank 0, a single independent component), an anti-symmetric tensor with zeros along the diagonal and otherwise $T_{ij} = -T_{ji}$ (irreducible tensor of rank 1, three independent components), and a traceless symmetric tensor with the diagonal elements summing to zero and otherwise $T_{ij} = T_{ji}$ (irreducible tensor of rank 2, five independent components). Be careful to note that the meaning of the term “rank” is different in different contexts: the rank of a cartesian tensor must, in general, be distinguished from the ranks of its irreducible parts.

We are now in a position to state the meaning of $\hat{\mathbf{U}}_\lambda$ as it appears in Equation (7.46) for the oscillator strength. First of all, it is an irreducible tensor operator of rank λ given by a sum over all the electrons in an N -electron configuration:

$$\hat{\mathbf{U}}_\lambda = \sum_{i=1}^N \hat{\mathbf{u}}_\lambda^i \quad (7.57)$$

where $\hat{\mathbf{u}}_\lambda^i$ is called the unit tensor; it is also an irreducible tensor of rank λ , but one which acts only on a single-electron orbital. The unit tensor is *defined* by the simple property

$$\langle \alpha l \| \hat{\mathbf{u}}_\lambda \| \alpha' l' \rangle \equiv \delta_{\alpha\alpha'} \delta_{ll'} \quad (7.58)$$

A good example of an explicit form for a unit tensor, at least when $l + l' + \lambda$ is even is

$$\hat{u}_{\lambda q} = \frac{\hat{Y}_{\lambda q}(\hat{\theta}, \hat{\phi})}{\langle l \| \hat{\mathbf{Y}}_\lambda \| l' \rangle} \quad (7.59)$$

For brevity, we shall not explain in detail how $\hat{\mathbf{U}}_\lambda$ arises in the derivation of Equation (7.46) for the oscillator strength. Suffice to say that it is a consequence of the sum over all electrons that appears in Equation (7.39) for the crystal field interaction $\hat{V}$ and the example just given for a unit tensor operator. Note how $\hat{V}$ depends on the irreducible single-electron operators $\hat{Y}_{kq}(\hat{\theta}_i, \hat{\phi}_i)$. More important

for the present purposes is to gain some understanding of how the reduced matrix element is evaluated for a pair of pure Russell–Saunders states $|\alpha(4f^N), SLJ\rangle$ and $|\alpha(4f^N), S'L'J'\rangle$.

The first step in the calculation is to recognize that $\hat{\mathbf{U}}_\lambda$ acts only on the spatial coordinates and not on the spin coordinates. This is a direct consequence of the electrostatic character of the crystal field interaction; the electron spin does not couple to purely electric forces. We can therefore immediately state that the reduced matrix element vanishes unless $S = S'$, a simple example of our assertion that the reduced matrix element embodies the physical content of the problem at hand. Of greater consequence, however, is that ultimately we must decompose the Russell–Saunders states into something that will permit use of Equation (7.58) for the reduced matrix element of the unit tensor. In other words, $\langle\alpha(4f^N), SLJ\|\hat{\mathbf{U}}_\lambda\|\alpha(4f^N), S'L'J'\rangle$ must be transformed in such a way as to explicitly show its dependence on at least one single-electron orbital. The next step is thus to decouple $\hat{\mathbf{L}}$ and $\hat{\mathbf{S}}$ with the help of the theorem

$$\begin{aligned} \langle\alpha(4f^N), SLJ\|\hat{\mathbf{U}}_\lambda\|\alpha(4f^N), S'L'J'\rangle &= (-1)^{L+S+J'+k} \delta_{SS'} \\ &\times \sqrt{(2J+1)(2J'+1)} \left\{ \begin{matrix} J & \lambda & J' \\ L' & S & L \end{matrix} \right\} \langle\alpha(4f^N), LS\|\hat{\mathbf{U}}_\lambda\|\alpha(4f^N), L'S'\rangle \end{aligned} \quad (7.60)$$

where the reduced matrix element now refers to the basis set $|\alpha(4f^N), LSM_L M_S\rangle \equiv |\alpha(4f^N), LM_L\rangle |SM_S\rangle$. Next, note that

$$\begin{aligned} \langle\alpha(4f^N), LS\|\hat{\mathbf{U}}_\lambda\|\alpha(4f^N), L'S'\rangle &= \sum_{i=1}^N \langle\alpha(4f^N), LS\|\hat{\mathbf{u}}_\lambda^i\|\alpha(4f^N), L'S'\rangle \\ &= N \langle\alpha(4f^N), LS\|\hat{\mathbf{u}}_\lambda^n\|\alpha(4f^N), L'S'\rangle \end{aligned} \quad (7.61)$$

where for these fully anti-symmetrized basis states advantage is taken of the independence of the unit tensor reduced matrix element on the identity of the electron (here taken to be the n th).

The next step is to break down the N -electron eigenstates $|\alpha(4f^N), LS\rangle$ into a superposition of the form

$$|\alpha(4f^N), \beta LS\rangle = \sum_{\bar{\beta}\bar{L}\bar{S}} G_{\bar{\beta}\bar{L}\bar{S}}^{\beta LS} |\alpha(4f^{N-1})\bar{\beta}\bar{L}\bar{S}, l|LS\rangle \quad (7.62)$$

This expression is a little complicated; here is what it means. First, the introduction of the quantum number β is meant to distinguish different terms of the same L and S . This point is one we have not made emphatic up to now, but is alluded to in our presentation of allowed Russell–Saunders terms for rare-earth ions as given in Table 7.4. Beginning with Nd^{3+} and continuing through Er^{3+} , examples exist of different eigenstates possessing different energies but which share the same L and S (and hence J). The quantum number β is used to give a unique label to these

different terms. Second, the meaning of the symbol $|\alpha(4f^{N-1})\bar{\beta}\bar{L}\bar{S}, l|LS\rangle$ is: “this is a state one obtains by angular momentum coupling of a totally antisymmetrized $(N-1)$ -electron state of total orbital angular momentum $\bar{L}$ and total spin $\bar{S}$ to the one-electron state of orbital angular momentum l (and spin s) so as to yield the N -electron state of total orbital angular momentum L and total spin S .” That is, this state is the (nonantisymmetrized) state that results from straightforward angular momentum coupling of the states $|\alpha(4f^{N-1})\bar{\beta}\bar{L}\bar{S}M_{\bar{L}}M_{\bar{S}}\rangle$ and $|lsm_l m_s\rangle$ by use of the Clebsch–Gordan coefficients. Equation (7.62) thus gives the correct fully antisymmetrized N -electron state $|\alpha(4f^N), \beta LS\rangle$ in terms of basis states constructed by simple angular momentum addition of fully-anti-symmetrized $N-1$ -electron states to the single-electron state $|LS\rangle$. The quantities $G_{\beta LS}^{\beta LS}$ are therefore the expansion coefficients in terms of the chosen basis; they guarantee that the resulting wave function is anti-symmetric under exchange of any two electrons and are called *coefficients of fractional parentage*. These coefficients may be looked up in tables and are often denoted by the symbol $\langle l^{N-1}(\bar{\beta}\bar{L}\bar{S})l|LS\rangle |l^N \beta LS\rangle$ (Cowan, 1981).

The final step then is to calculate the reduced matrix element

$$\begin{aligned}
 & \langle \alpha(4f^N), LS \| \hat{\mathbf{u}}_\lambda^n \| \alpha(4f^N), L'S' \rangle \\
 &= \sum_{\beta \bar{L} \bar{S}} \sum_{\bar{\beta} \bar{L} \bar{S}} G_{\bar{\beta} \bar{L} \bar{S}}^{\beta' L' S'} G_{\bar{\beta} \bar{L} \bar{S}}^{\beta LS} \langle \alpha(4f^{N-1}) \bar{\beta} \bar{L} \bar{S}, l | LS \| \hat{\mathbf{u}}_\lambda^n \| \alpha(4f^{N-1}) \bar{\beta} \bar{L} \bar{S}, l | L'S' \rangle \\
 &= \sum_{\beta \bar{L} \bar{S}} \sum_{\bar{\beta} \bar{L} \bar{S}} G_{\bar{\beta} \bar{L} \bar{S}}^{\beta' L' S'} G_{\bar{\beta} \bar{L} \bar{S}}^{\beta LS} (-1)^{\bar{L}+l+L'+\lambda} \delta_{\bar{L}\bar{L}} \sqrt{(2L+1)(2L'+1)} \begin{Bmatrix} L & \lambda & L' \\ l & \bar{L} & l \end{Bmatrix} \langle l | \hat{\mathbf{u}}_\lambda^n | l \rangle \\
 &= \sum_{\beta \bar{L} \bar{S}} G_{\bar{\beta} \bar{L} \bar{S}}^{\beta' L' S'} G_{\bar{\beta} \bar{L} \bar{S}}^{\beta LS} (-1)^{\bar{L}+l+L'+\lambda} \sqrt{(2L+1)(2L'+1)} \begin{Bmatrix} L & \lambda & L' \\ l & \bar{L} & l \end{Bmatrix} \quad (7.63)
 \end{aligned}$$

where in the next to last step a decoupling formula analogous to Equation (7.60) has been used and in the last step the definition for the reduced matrix element of the unit tensor Equation (7.58) has been used.

Combining Equations (7.60), (7.61) and (7.63), the final result is

$$\begin{aligned}
 & \langle \alpha(4f^N), SLJ \| \hat{\mathbf{U}}_\lambda \| \alpha(4f^N), S'L'J' \rangle = \delta_{SS'} N \sum_{\beta \bar{L} \bar{S}} G_{\bar{\beta} \bar{L} \bar{S}}^{\beta' L' S'} G_{\bar{\beta} \bar{L} \bar{S}}^{\beta LS} (-1)^{L+S+J'\bar{L}+l+L'+2\lambda} \\
 & \times \sqrt{(2J+1)(2J'+1)} \sqrt{(2L+1)(2L'+1)} \begin{Bmatrix} J & \lambda & J' \\ L' & S & L \end{Bmatrix} \begin{Bmatrix} L & \lambda & L' \\ l & \bar{L} & l \end{Bmatrix} \quad (7.64)
 \end{aligned}$$

In practice, however, one can simply look up the values of the reduced matrix elements and plug them directly into Equation (7.46) in order to calculate the desired oscillator strengths (Nielson and Koster, 1964, Carnall *et al.*, 1977, Cowan, 1981). An example is given in Table 7.6, which lists values for pure Russell–Saunders terms relevant to Pr^{3+} . Note in reference to use of such tables that there exists a

Table 7.6. Nonzero reduced matrix elements for Russell–Saunders terms relevant to Pr^{3+}

	$\ U_2\ $	$\ U_4\ $	$\ U_6\ $
$\langle {}^1S_0 \ \hat{U}_\lambda \ {}^1S_0 \rangle$	0	0	0
$\langle {}^1S_0 \ \hat{U}_\lambda \ {}^1D_2 \rangle$	0.755929	0	0
$\langle {}^1S_0 \ \hat{U}_\lambda \ {}^1G_4 \rangle$	0	0.755929	0
$\langle {}^1S_0 \ \hat{U}_\lambda \ {}^1I_6 \rangle$	0	0	0.755929
$\langle {}^3P_{0,1,2} \ \hat{U}_\lambda \ {}^3P_{0,1,2} \rangle$	−0.801784	0	0
$\langle {}^3P_{0,1,2} \ \hat{U}_\lambda \ {}^3F_{0,1,2} \rangle$	0.92582	0.723747	0
$\langle {}^3P_{0,1,2} \ \hat{U}_\lambda \ {}^3H_{4,5,6} \rangle$	0	0.690066	−0.462910
$\langle {}^1D_2 \ \hat{U}_\lambda \ {}^1D_2 \rangle$	−0.641533	0.706305	0
$\langle {}^1D_2 \ \hat{U}_\lambda \ {}^1G_4 \rangle$	0.947607	0.166821	0.360375
$\langle {}^1D_2 \ \hat{U}_\lambda \ {}^1I_6 \rangle$	0	0.433117	1.37620
$\langle {}^3F_{0,1,2} \ \hat{U}_\lambda \ {}^3F_{0,1,2} \rangle$	−0.333333	−0.333333	−0.333333
$\langle {}^3F_{0,1,2} \ \hat{U}_\lambda \ {}^3H_{4,5,6} \rangle$	0.835711	1.01575	−1.24722
$\langle {}^1G_4 \ \hat{U}_\lambda \ {}^1G_4 \rangle$	0.223814	−1.07698	1.19770
$\langle {}^1G_4 \ \hat{U}_\lambda \ {}^1I_6 \rangle$	0.581087	1.43945	1.06017
$\langle {}^1I_6 \ \hat{U}_\lambda \ {}^1I_6 \rangle$	2.21906	1.27417	0.356532

“handedness” to the reduced matrix elements; that is,

$$\langle \alpha(4f^N), SLJ \| \hat{U}_\lambda \| \alpha(4f^N), S' L' J' \rangle = -\langle \alpha(4f^N), S' L' J' \| \hat{U}_\lambda \| \alpha(4f^N), SLJ \rangle \quad (7.65)$$

An excellent example of the use of reduced matrix elements and the calculation of oscillator strengths with the Judd–Ofelt formula may be found in Weber *et al.* (1972).

Readers interested in pursuing the details of this formally complex section are referred to Wybourne (1965) and Hüfner (1978) for discussions of line strength calculations and the derivation of the Judd–Ofelt expression, and to Weissbluth (1978) and Cowan (1981) for discussions of operator algebra, the Wigner–Eckart theorem, and the calculation of reduced matrix elements.

7.3.3 Selection rules for electric dipole transitions

Embedded within Equation (7.64) are selection rules for electric-dipole transitions between pure Russell–Saunders terms of rare-earth ions in a condensed host. These

are

$$\Delta S = 0, \quad |\Delta L| \leq 6, \quad |\Delta J| \leq 6$$

The spin selection rule derives simply from the Kronecker delta pre-factor in Equation (7.64), the origin of the latter being the purely electrical nature of the crystal field perturbation – such perturbations do not couple to the electron spin. The ΔL and ΔJ selection rules derive from the pair of Wigner 6- j (Rotenberg *et al.*, 1969) symbols in Equation (7.64) as follows: First, a key property of the 6- j symbol

$$\left\{ \begin{matrix} j_1 & j_2 & j_3 \\ j'_1 & j'_2 & j'_3 \end{matrix} \right\} \quad (7.66)$$

is that the four triangle inequalities $|j_1 - j_2| \leq j_3 \leq |j_1 + j_2|$, $|j_1 - j'_2| \leq j'_3 \leq |j_1 + j'_2|$, $|j'_1 - j_2| \leq j_3 \leq |j'_1 + j_2|$, and $|j'_1 - j'_2| \leq j_3 \leq |j'_1 + j'_2|$ must be satisfied in order for the symbol to assume a nonzero value. Thus in the second 6- j factor above, for $j'_1 = j'_3 = l$ and $j_2 = \lambda$, we see that since $l = 3$ the maximum allowed value of λ is 6. This is why the sum in the Judd–Ofelt expression Equation (7.46) for the oscillator strength terminates at $\lambda = 6$. Next, note that if $j_1 = J$, $j_2 = \lambda$, and $j_3 = J'$ in the first 6- j symbol, a maximum of $\lambda = 6$ implies that the maximum $|J - J'|$ must be 6. A similar argument yields a maximum of 6 for $|L - L'|$.

As always, however, we need to remember that these rules must be applied to the full intermediate-coupling eigenstates. Thus since the spin-orbit and Coulomb interactions produce admixtures of Russell–Saunders terms of different L and S , simple inspection of the intermediate-coupling term labels can confirm whether a transition is allowed but cannot identify dipole-forbidden transitions. About the best that can be said is that since the intermediate-coupling term labels indicate the dominant Russell–Saunders contribution to the eigenstate, those intermediate-coupling terms that satisfy the selection rules might be supposed to yield relatively larger oscillator strengths, at least if the reduced matrix elements do not by chance conspire against it. In particular, transitions between terms of the same spin will tend on average to be stronger than those of different spin. As an example, Figure 7.9 shows the absorption spectrum of Pr^{3+} in a fluoride glass host. Note that the $S = 3$ ground state implies weaker transitions to $^1G_4$ and to $^1D_2$ than to the ^3P_J levels. This prediction is borne out in the measured spectrum.

As for the ΔJ selection rule, with no crystal field, J remains a good quantum number and thus transitions of $\Delta J > 6$ are strictly dipole-forbidden. Under the perturbation of the crystal field, however, a small degree of mixing of J levels is inevitable and very weak absorption or emission features may be observed.

The reader may recall that the foregoing was all based on a purely static model of the crystal field. Insofar as the largest contributions to the Judd–Ofelt parameters derive from $4f^{N-1}5d$ and $4f^{N-1}6p$ excited configurations (that is, $\Delta l = \pm 1$), only odd-parity crystal fields can yield electric-dipole transitions. Therefore rare-earth

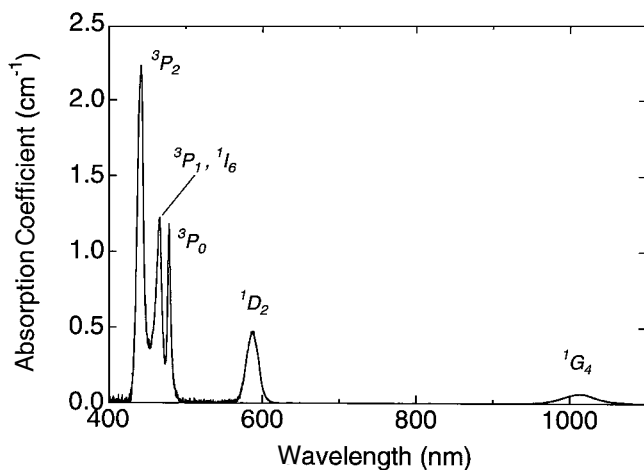


Figure 7.9: The visible and near-infrared absorption spectrum of Pr^{3+} in the fluorozirconate glass ZBLAN. The absorption features are labeled with the Russell–Saunders term of the upper J manifold; the ground term is $^3H_4$. The concentration of the dopant ions is 1 wt%.

ions in crystals with inversion symmetry should not exhibit optical transitions. That electric-dipole transitions in such hosts are nevertheless observed is believed to result from odd-parity *dynamical* contributions to the crystal field. Judd (1962) has shown that all of the results derived for a static crystal field are recovered for dynamically induced transitions; in particular, for hosts of arbitrary symmetry, the crystal field parameters $\bar{B}_{kq}$ serve to simultaneously parametrize both static and dynamic components, at least for fixed temperature. For crystalline hosts possessing inversion symmetry, lattice phonons break the local symmetry and odd-parity $\bar{B}_{kq}$ attain nonzero values. Nonzero (and temperature-dependent) Judd–Ofelt parameters result as well as temperature-dependent broadening of homogeneous linewidths, the latter phenomenon being likewise observed in lower-symmetry hosts. Aside from linewidth data, additional evidence for electron–phonon coupling is found in the appearance of weak phonon sidebands that accompany the purely electronic “zero-phonon” transitions we have been discussing so far. These sidebands result from the combination of an electronic transition with emission or absorption of a lattice phonon. At low temperature, only phonon emission occurs and sidebands appear at frequencies below the main transition; as a function of frequency, the sideband absorption closely follows the phonon density of states.

7.4 NONRADIATIVE RELAXATION

We have seen that a rare-earth ion embedded in a condensed host material will exhibit radiative transitions as a consequence of the static and dynamic crystal

fields impressed upon the ion. As just mentioned, temperature-dependent linewidths and phonon sidebands are additional signatures of this ion–host coupling. Another phenomenon related to the ion–host interaction, one of central importance in discussing the suitability of a particular transition to laser applications, is nonradiative relaxation. In this process, coupling of the electronic coordinates of the ion to the phonon coordinates of the host lattice makes possible the decay of an excited state through the emission of one or more phonons. If the initial and final electronic states are separated by energy ΔE , a quantity called the *energy gap*, the total energy of the emitted phonons is likewise ΔE , meaning that the transition involves no radiative emission. In general, radiative relaxation competes with nonradiative relaxation; the total relaxation rate is just the algebraic sum of the two rates:

$$\gamma_{\text{tot}} = \gamma_{\text{rad}} + \gamma_{\text{nr}} \quad (7.67)$$

The important practical consequence of nonradiative relaxation is that it can possibly deplete the population of a laser's upper state at a rate much faster than the radiative rate, thus spoiling that state's ability to store energy and exhibit a large stimulated emission cross-section.

Rare-earth ions have historically offered a particularly attractive means for investigating nonradiative processes in condensed matter. As a consequence of the weak coupling between the ions and the lattice, this interaction is treated well through perturbation theory and general analytic expressions have been found for the relaxation rate as a function of temperature and of the energy gap. For example, when emission of $N \geq 3$ phonons occurs during a nonradiative transition across an energy gap ΔE , it is found theoretically that at low temperatures the nonradiative relaxation rate is given by the simple form

$$\gamma_{\text{nr}}(T = 0) = K e^{-\alpha \Delta E} \quad (7.68)$$

a famous result termed the *energy-gap law*. The miracle of the energy-gap law is that the coefficients K and α depend only on the host medium and are largely independent of the particular impurity ion or transition of interest. Moreover, the quantity α systematically increases as the number of emitted phonons $N = \Delta E / \hbar \omega_{\text{eff}}$ increases, where ω_{eff} is some effective phonon frequency of multi-phonon emission. In fact, in most cases ω_{eff} is just the maximum phonon frequency that the host lattice can support, a reasonable expectation given that the transition rate is calculated in N th-order perturbation theory and thus is likely to be maximized when ω_{eff} assumes its largest possible value. However, some host materials may possess lower-energy phonons with either stronger electron–phonon coupling or higher densities of states than the highest-energy phonons and thus yield larger multi-phonon transition rates. Long wavelength lasers therefore require the use of host materials with low

Table 7.7. Measured parameters for multi-phonon emission for various rare-earth-doped host materials

Host	$\hbar\omega_{eff}$ (cm ⁻¹)	K (s ⁻¹)	β (cm)
Crystals			
LaBr ₃	175	1.2×10^{10}	1.9×10^{-2}
LaCl ₃	260	1.5×10^{10}	1.3×10^{-2}
LaF ₃	350	6.6×10^8	5.6×10^{-3}
LiYF ₃	400	3.5×10^7	3.8×10^{-3}
Y ₂ O ₃	550	2.7×10^8	3.8×10^{-3}
YAlO ₃	600	5.0×10^9	9.6×10^{-3}
Y ₃ Al ₅ O ₁₂	700	9.7×10^7	3.1×10^{-3}
Glasses			
Fluoride	500	1.6×10^{10}	5.2×10^{-3}
Germanate	900	3.4×10^{10}	4.9×10^{-3}
Silicate	1100	1.4×10^{12}	4.7×10^{-3}
Phosphate	1200	5.4×10^{12}	4.7×10^{-3}

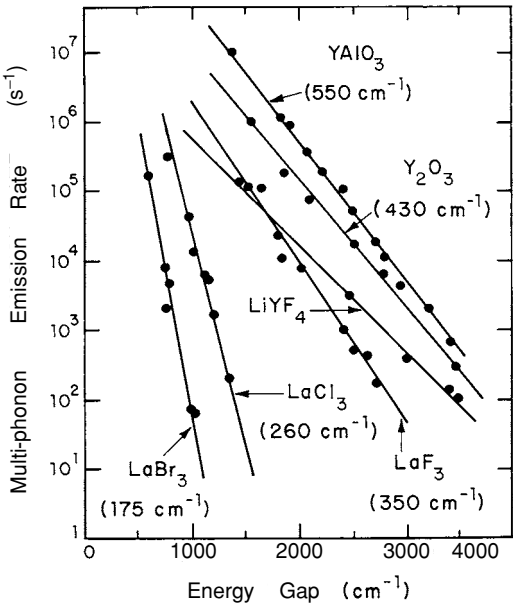


Figure 7.10: The energy-gap law illustrated for rare-earth ions in various crystalline host materials.

maximum phonon frequencies, that is, materials containing constituents of large atomic mass. This trend is illustrated in Table 7.7, which gives for different host materials corresponding values of ω_{eff} , K , and α . Figure 7.10 shows the energy-gap law in action for various host crystals.

With respect to the temperature dependence of the nonradiative relaxation rate, an equally elegant expression is derived:

$$\gamma_{nr}(T) = \gamma_{nr}(0) \left(1 + \frac{1}{e^{\hbar\omega_{eff}/kT} + 1} \right)^{\frac{\Delta E}{\hbar\omega_{eff}}} \quad (7.69)$$

In this result, the second term enclosed in parentheses is just the Bose–Einstein occupation factor for a phonon of frequency ω_{eff} ; it expresses the idea that thermal occupation of phonon modes stimulates nonradiative relaxation in a manner analogous to radiative stimulated emission. A rule of thumb regarding nonradiative relaxation in rare-earth materials states that at room temperature, if N is 5 or larger, then radiative relaxation will dominate nonradiative relaxation.

7.5 RADIATIONLESS ENERGY TRANSFER

All that we have been discussing so far involves radiative or nonradiative electronic transitions within a single rare-earth ion. Other dynamical phenomena become possible if we also consider coupling of one rare-earth ion to another. The essential consequence is that energy can be directly exchanged between the ions. If these ions are labeled A and B, such energy transfer may be indicated symbolically by



where the asterisk indicates an ion in a more highly excited state. The ions A and B may be different or identical.

Although interion coupling occurs through several mechanisms, by far the most important for practical applications in rare-earth materials arises from electrical interactions. Consider from a classical perspective the electric field surrounding an isolated and oscillating electric dipole, $\mu(t) = \mu_0 e^{-j\omega_0 t}$:

$$\tilde{\mathbf{E}}(\mathbf{r}, t) = e^{-j\omega_0 t} \left\{ \beta^2 (\mathbf{n} \times \mu_0) \times \mathbf{n} \frac{e^{j\beta r}}{r} + [3\mathbf{n}(\mathbf{n} \cdot \mu_0) - \mu_0] \left(\frac{1}{r^3} - \frac{j\beta}{r^2} \right) e^{j\beta r} \right\} \quad (7.71)$$

where $\mathbf{n} = \mathbf{r}/r$ is a unit vector that points from the location of the dipole towards the field point $\mathbf{r}$ and $\beta \equiv \omega_0/c$ (see Figure 7.11). The first term inside the curly brackets dominates at large r and describes electromagnetic radiation by the dipole; the wave vector for this monochromatic wave is β . For $r\beta \ll 1$, however, in the so-called near-field zone, the $1/r^3$ term dominates and the electric field becomes

$$\tilde{\mathbf{E}}(\mathbf{r}, t) = [3\mathbf{n}(\mathbf{n} \cdot \mu_0) - \mu_0] \frac{e^{-j\omega_0 t}}{r^3} \quad (7.72)$$

Now consider a neighboring dipole, μ' , one that is not oscillating, located at position

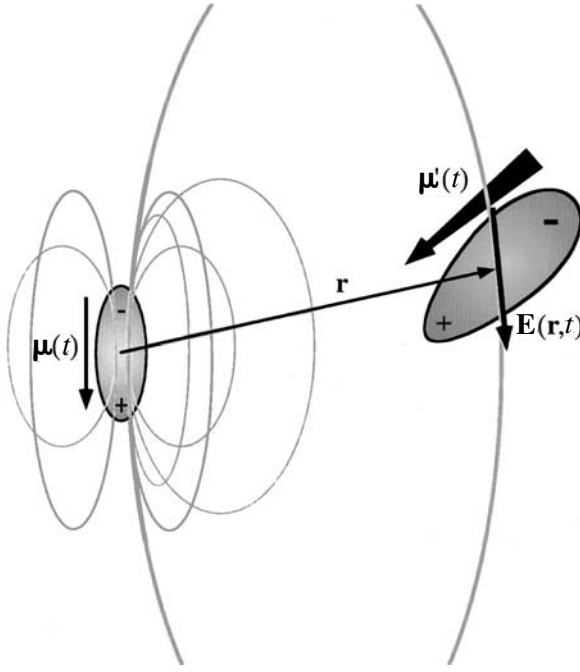


Figure 7.11: The interaction between the electric field of one dipole and the electric-dipole moment of another.

$\mathbf{R}$ within the near-field of the first dipole. The electrostatic energy of this dipole in the presence of the oscillating field of the first dipole is then

$$H = -\mu' \cdot \mathbf{E}(\mathbf{R}, t) = -[3(\mu' \cdot \mathbf{n})(\mathbf{n} \cdot \mu_0) - \mu' \cdot \mu_0] \frac{e^{-j\omega_0 t}}{R^3} \quad (7.73)$$

This last expression can be converted to quantum mechanical form by simply writing the dipole moments μ and μ' as operators. Viewing the two dipoles as rare-earth ions, the first of which is in an excited state $|A^*\rangle$ while the second is in a state $|B\rangle$ of lower energy, transitions of the interacting pair can occur in which energy is transferred from the first ion to the other. The transition rate is given by Fermi's Golden Rule, which ensures that energy is conserved during the exchange. The rate so obtained is

$$\begin{aligned} W_{transfer} &= \frac{2\pi}{\hbar} |\langle i | \hat{H} | f \rangle|^2 \rho_f \propto \frac{1}{R^6} |\langle A^* | \langle B | \hat{\mu}_A \hat{\mu}_B | A \rangle | B^* \rangle|^2 \rho_f \\ &= \frac{1}{R^6} |\langle A^* | \hat{\mu}_A | A \rangle|^2 |\langle B | \hat{\mu}_B | B^* \rangle|^2 \rho_f \end{aligned} \quad (7.74)$$

where the final state $|f\rangle$ is given by the product $|A\rangle|B^*\rangle$ in which the energy-donating ion has dropped to a lower-energy level whereas the accepting ion has been

raised to a higher one. One therefore finds in the literature the terms “donor” and “acceptor” often used to refer to the two ions, and almost equally often “sensitizer” and “activator,” respectively.

Note that the above form for the transition rate contains dipole-moment matrix elements just as do analogous expressions for radiative transitions. For this reason, the density of states factor in Equation (7.74) can then be plausibly argued as being equal to the overlap integral of the donor ion’s electric-dipole emission cross-section with the acceptor ion’s electric-dipole absorption cross-section. This argument has been made rigorous by Förster (1949) and Dexter (1953), who derived the complete expression for the electric dipole–dipole donor-to-acceptor transition rate in a medium of refractive index n :

$$W_{i \rightarrow f}^{DD} = \frac{3 \hbar^5 c^4}{4 \pi n^4} \frac{1}{\tau_d} \frac{1}{R^6} f_a \int_0^\infty \frac{L_d(E) L_a(E)}{E^4} dE \quad (7.75)$$

where τ_d is the radiative lifetime for the donor transition $A^* \rightarrow A$, f_a is the acceptor’s absorption oscillator strength on the transition $B \rightarrow B^*$, and $L_{d,a}(E)$ are normalized line shape functions $\int_0^\infty L_{d,a}(E) dE = 1$ for electric-dipole emission from the donor and electric-dipole absorption by the acceptor for their respective transitions. The spatial extent of Förster–Dexter transfer is typically 0 to ~ 100 Å.

There are other possible electrical interactions. Dexter (1953) also derived an expression for coupling of an electric dipole to an electric quadrupole, which showed a dependence on the interion separation of R^{-8} . Similarly, a quadrupole–quadrupole interaction yields a dependence of R^{-10} . In terms of the rate for energy transfer by dipole–dipole coupling, W^{DD} , the rates for these higher-order processes are given by

$$W_{i \rightarrow f}^{DQ} = W^{QD} = \frac{f_Q}{f_D} \left(\frac{\lambda}{R} \right)^2 W_{i \rightarrow f}^{DD} \quad \text{electric dipole–quadrupole coupling} \quad (7.76)$$

and

$$W_{i \rightarrow f}^{QQ} = \left(\frac{f_Q}{f_D} \right)^2 \left(\frac{\lambda}{R} \right)^4 W_{i \rightarrow f}^{DD} \quad \text{electric quadrupole–quadrupole coupling} \quad (7.77)$$

where $\lambda = h/E$ is the wavelength corresponding to the transition energy E and $f_{D,Q}$ is the oscillator strength for an electric-dipole or electric-quadrupole transition, respectively.

Because electric-dipole matrix elements are typically larger than for higher-order multi-poles, one expects dipole–dipole coupling to be most commonly observed. Nevertheless, unlike radiative transitions, high-order radiationless multi-pole transitions are readily observed, especially for rare-earth ions. This comes about because, as we have seen, electric-dipole transitions in rare earths are nominally forbidden

and thus exhibit oscillator strengths of only 10^{-6} . On the other hand, $4f \rightarrow 4f$ electric-quadrupole transitions are fully allowed and exhibit oscillator strengths on the order of 10^{-7} – 10^{-8} . Inspection of Equations (7.76) and (7.77) shows that dipole–quadrupole and quadrupole–quadrupole energy-transfer transition rates will exceed the dipole–dipole rate when the donor–acceptor separation R is less than $\sim \lambda/10$, a circumstance readily obtained in practice even at low concentrations. As one example, evidence for quadrupole–quadrupole coupling has been observed in Yb^{3+} -doped silica glass (Brundage and Yen, 1986).

The line shape overlap integral in Equation (7.75) for the donor-to-acceptor transition rate implies a resonance condition for energy transfer. If the two ions are identical and the upgoing transition of the acceptor is the reverse of the downgoing transition of the donor, then the transition rate is clearly maximized and independent of the linewidths, at least for homogeneously broadened energy levels. In the further event that all possible acceptor ions are in the same state, most importantly the ground state, the excitation energy initially located on the donor may repeatedly hop from one acceptor site to another. This process is called resonant migration; an ensemble of excitations undergoing resonant migration on an acceptor sublattice may be viewed as an excitonic gas.

In the opposite extreme where the line shape overlap integral tends to zero, the transition rate likewise vanishes within the Förster–Dexter model. This situation most likely occurs at low temperatures where homogeneous linewidths are small, or more transparently for donor and acceptor transitions widely separated in energy. Nevertheless, efficient radiationless energy transfer is still observed across large energy gaps because absorption or emission of lattice phonons can make up the energy deficit. Under such conditions, theoretical analogs of Förster–Dexter transfer can be derived. For example, in an endothermic one-phonon-assisted process the rate of energy transfer is proportional to the occupation number $n(\omega_{ph}, T) = 1/(e^{\hbar\omega_{ph}/kT} - 1)$ of the promoting phonon mode. Conversely, for an exothermic one-phonon process the transition rate goes as $1 + n(\omega_{ph}, T)$. In both cases, the characteristic temperature dependence of the Bose–Einstein factor can be exploited to experimentally detect either process. For large energy deficits, the situation is analogous to that of single-ion nonradiative decay and therefore energy transfer between rare-earth species recovers the energy-gap law $W_{i \rightarrow f} \propto e^{-\alpha \Delta E}$ and its affiliated temperature dependence (Miyakawa and Dexter, 1970). Yet another possibility arises when the energy deficit is so small that the phonon density of states approaches zero. In this circumstance, the one-phonon transfer rate is small but a two-phonon process – one in which one high-energy phonon is absorbed and another is emitted – can yield a much larger transition rate. The small energy deficit is then made up by the difference of the two phonon energies.

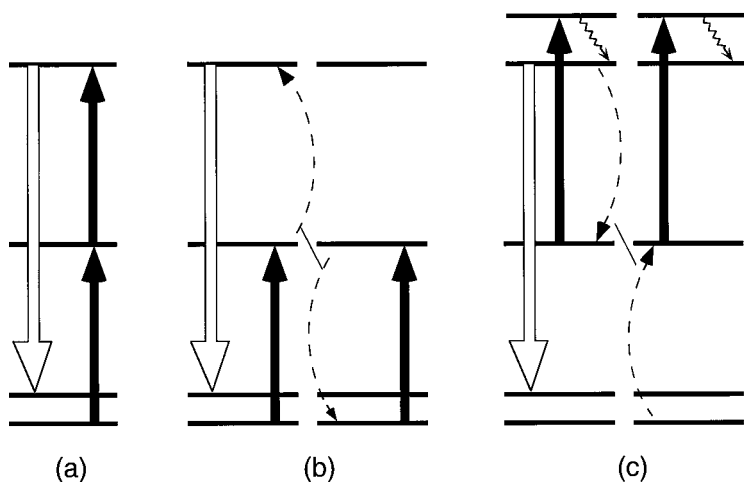


Figure 7.12: The three basic mechanisms of upconversion: (a) multi-photon absorption; (b) energy-transfer upconversion; (c) the photon avalanche mechanism.

7.6 MECHANISMS OF UPCONVERSION

At last we have in place all the pieces we need in order to discuss the conversion of two or more low-energy photons into a single high-energy photon. The essence of the process is to accumulate population in a high-lying state of the desired emissive ion; Figure 7.12 illustrates the three principal mechanisms by which this can be accomplished. These are resonant multi-photon absorption, cooperative energy-transfer upconversion, and the photon avalanche. The essential features of these mechanisms are described in detail as follows.

7.6.1 Resonant multi-photon absorption

The most obvious approach to building population in a high-lying state other than by direct one-photon pumping is through sequential absorption of multiple photons through one or more resonant intermediate states. For example, Figure 7.12(a) shows this process for two-photon pumping. In general, two separate pump lasers are required in order to drive this process; the operating wavelength of each laser is chosen so as to maximize the pumping rate to the upper level of the relevant transition. In the laser spectroscopy literature, the second step is often termed *excited-state absorption*, or ESA. In some instances, coincidental equivalence of the two wavelengths allows one pump laser to be used; in rare circumstances even three-photon pumping can be accomplished with a single pump laser (for example, Tm^{3+} , as discussed in Chapter 8). The likelihood that such single-wavelength

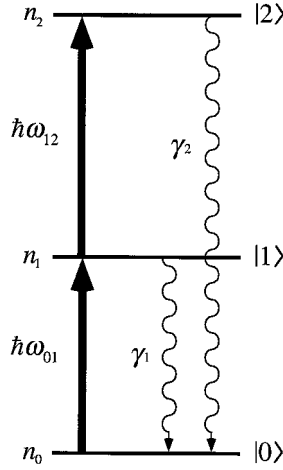


Figure 7.13: Energy-level diagram depicting pump and relaxation processes for two-photon upconversion pumping.

multi-photon pump processes will occur is significantly increased for disordered host materials owing to the broader linewidths observed in such hosts.

In order to gain a little more insight into upconversion by multi-photon resonant absorption, consider a three-level system with states labeled 0, 1, and 2 that is optically pumped by two lasers tuned to resonance with the ground- and excited-state absorption transitions, respectively (Figure 7.13). A rate equation for the change of the population density in the intermediate state $|1\rangle$ under cw pumping of the system takes the form

$$\dot{n}_1 = \frac{I_{01}}{\hbar\omega_{01}}[\sigma_{01}(\omega_{01})n_0 - \sigma_{10}(\omega_{01})n_1] - \frac{I_{12}}{\hbar\omega_{12}}[\sigma_{12}(\omega_{12})n_1 - \sigma_{21}(\omega_{12})n_2] - \gamma_1 n_1 \quad (7.78)$$

where n_i is the population density (number per unit volume) in the level i , I_{ij} is the intensity of the pump laser (of frequency ω_{ij}) resonant with the $i \rightarrow j$ transition, σ_{ij} is a cross-section for absorption (if $i < j$) or simulated emission (if $i > j$) on the $i \rightarrow j$ transition, and finally γ_1 is the total relaxation rate from the $|1\rangle$ state. A simpler expression obtains for the time rate of change of n_2 :

$$\dot{n}_2 = \frac{I_{12}}{\hbar\omega_{12}}[\sigma_{12}(\omega_{12})n_1 - \sigma_{21}(\omega_{12})n_2] - \gamma_2 n_2 \quad (7.79)$$

where γ_2 is the total relaxation rate of the $|2\rangle$ state; this relaxation is assumed for simplicity to occur exclusively to the ground state. The population in the three states also must satisfy the conservation condition

$$n_0 + n_1 + n_2 = N \quad (7.80)$$

where N is the total number of ions. Solving for the steady-state ($\dot{n}_1 = \dot{n}_2 = 0$) population densities attained by this cw doubled-pumped three-level system yields for the $|2\rangle$ state

$$n_2 = \frac{\frac{I_{01}}{I_{s,01}} \frac{I_{12}}{I_{s,12}} N}{1 + 2 \frac{I_{01}}{I_{s,01}} + \left(1 + \frac{\gamma_2}{\gamma_1}\right) \frac{I_{12}}{I_{s,12}} + 3 \frac{I_{01}}{I_{s,01}} \frac{I_{12}}{I_{s,12}}} \quad (7.81)$$

where for simplicity it has been assumed that $\sigma_{ij} \equiv \sigma_{ji}$, and $I_{s,ij} = \gamma_j \hbar \omega_{ij} / \sigma_{ij}$ is the saturation intensity for the $i \rightarrow j$ transition.

Equation (7.81) for the population density in the $|2\rangle$ state shows that in the limit where $I_{01} \ll I_{s,01}$ and $I_{12} \ll I_{s,12}$, that is, the linear pumping regime, n_2 is proportional to the bilinear product $I_{01} I_{12}$, implying that in the case where a single laser is used to drive both transitions, a quadratic dependence will be observed for the $|2\rangle$ state population density. Such a multi-linear or power-law dependence is in general characteristic of weakly pumped upconversion processes: for an n -photon process that yields population in a given state, an n th-order dependence on the pump intensities will be observed for the population density in that state. In the opposite limit of very large pump intensities, Equation (7.81) shows that for a three-level two-photon process $n_2 \rightarrow N/3$. It is easy to show that in this high-intensity limit all three population densities attain this value, suggesting that for such a simple upconversion model net population inversion is not possible.

Important variations on resonant multi-photon pumping deserve discussion. Consider now the four-state energy-level diagram depicted in Figure 7.14, where

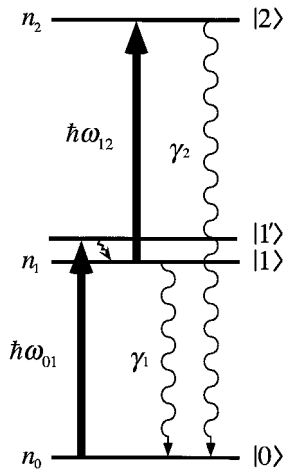


Figure 7.14: Energy-level diagram for a two-photon pumping mechanism capable of producing population inversion with respect to the ground state.

again for simplicity we take $\sigma_{ij} \equiv \sigma_{ji}$ and further assume that a single laser is sufficient to yield two-photon upconversion pumping. Moreover, the ground-state transition is taken to occur to a new state $|1'\rangle$ that relaxes so rapidly by nonradiative decay to state $|1\rangle$ that its population density is well approximated by 0. Under these conditions, the steady-state population densities must satisfy the algebraic equations

$$0 = -\frac{I\sigma_{01'}}{\hbar\omega_{01'}}n_0 + \gamma_1n_1 + \gamma_2n_2 \quad (7.82a)$$

$$0 = \frac{I\sigma_{12}}{\hbar\omega_{12}}(n_1 - n_2) - \gamma_2n_2 \quad (7.82b)$$

and

$$n_0 + n_1 + n_2 = N \quad (7.82c)$$

The reader may verify that for this model weak pumping again yields a quadratic dependence on pump intensity for the n_2 population. On the other hand, for $I \rightarrow \infty$, $n_2 \rightarrow N/2$ and most interestingly $n_0 \rightarrow 0$. We thus see in this circumstance the advantage afforded by the presence of a rapidly relaxing intermediate state lying within the upconversion chain. Not only has the population in the high-lying level increased but also a population inversion has developed between the $|2\rangle$ and $|0\rangle$ states (as well as between the less interesting $|2\rangle$ and $|1'\rangle$ states). This situation is reminiscent of one-photon pumped three- and four-level lasers where nonradiative relaxation plays an important role in the population kinetics.

As a last remark on this topic, we mention that ESA is sometimes actually deleterious to laser operation as it can deplete population in a desired upper laser level. For example, such a situation would arise in Figure 7.14 if $|2\rangle$ were the desired upper state yet a higher-lying state $|3\rangle$ yielded an ESA transition $2 \rightarrow 3$ resonant with one of the pump frequencies. Pump energy is thus wasted, even in the presence of rapid $3 \rightarrow 2$ relaxation. However, an even worse consequence of ESA in this context is that the laser output power may level off while the pump power is increased, in contradiction to simple laser models that normally predict the input and output powers to be proportional. Avoiding ESA transitions must always be kept in mind for optically pumped rare-earth-based lasers given the rich energy-level structure found in some ions.

7.6.2 Cooperative upconversion

The discussion in Section 7.5 describes the essential microscopic physics of radiationless energy transfer. In this section we further elaborate on how this mechanism forms the basis of an upconversion process. More concretely, as motivated by the

rate-equation analysis of multi-photon absorption in the previous section, we also wish to develop a rate-equation picture of the population kinetics of radiationless cooperative upconversion. The situation, however, is considerably more complicated than the case of multi-photon absorption; in many circumstances of practical interest, exact analytic solutions for the population kinetics are not available. Indeed, even different approximations that apply to the same physical situation yield qualitatively different analytic results, which unfortunately obscures how the *macroscopic* populations explicitly depend on the *microscopic* energy-transfer transition rates. (A good example of this problem will appear in the following discussion on the diffusion and hopping models of macroscopic energy transfer.) The situation is made worse if high excitation densities are considered, as might exist in a laser, or when nonuniform or nonrandom distributions of the impurity ions are involved. Nevertheless, it is still of great value in developing some intuition about radiationless energy transfer and upconversion processes to review the various models and observe how these models motivate at least a *phenomenological* rate-equation description of the population flow. Such a description will prove useful in discussing optical pumping of upconversion lasers based on radiationless transfer mechanisms.

To proceed, a few definitions and conditions must be stated:

- (1) The system to be considered is composed of a host matrix of volume V which is doped with a total of N_d donors and N_a acceptors. These numbers are intended to be time-independent quantities and thus do not reflect the state of excitation of the donors or acceptors. Moreover, we for now take the acceptor ions to be in the ground state and later in the section show how the more general case of excited acceptors can be incorporated into the phenomenological rate equations. It is through the rate equations that we will specifically model the upconversion population kinetics. The number of excited donors is denoted by N_d^* .
- (2) To simplify the following discussion, the donor species A is considered as distinct from the acceptor species, B. The results following from this assumption are readily reinterpreted in the case of identical donor and acceptor species. On the other hand, a crucial feature of our discussion will be the possibility that an excited donor A^* may transfer energy to a neighboring *unexcited donor* A, a process indicated symbolically by



and occurring at a rate given by the Förster–Dexter prescription. Such an exchange implies that the donor excitation can migrate from donor site to donor site until the excited donor undergoes the single ion relaxation step $A^* \rightarrow A$ or a true transfer to an acceptor, $A^* + B \rightarrow A + B^*$, occurs. For a given ion–ion separation, the former transition may occur much more rapidly than the latter given that one expects a high degree of overlap between the absorption and emission line shapes of the donor.

- (3) Only a weakly excited system is considered. Thus for all time, the number of donors in the ground state is approximated by N_d . The initial excitation of the donor system is assumed to occur with a δ -function laser pulse.
- (4) The donor and acceptor energy levels are considered to be homogeneously broadened. This excludes the possibility of spectral diffusion through inhomogeneously distributed states of the donor sublattice and ensures that the probability of transfer to acceptors depends only on the donor–acceptor separation.

As implied in item (2) above, the donor–donor and donor–acceptor transfers will in general occur at different rates for a given separation R between the participating ions. We denote these rates by the variables $k_{da} \equiv C_{da}/R^s$ and $k_{dd} \equiv C_{dd}/R^s$, where s is the multi-pole order of the transfer, assumed for simplicity to be the same for both processes. It is the relative magnitudes of these rates that determine the details of the observed population kinetics, as we now show.

Although not exhaustive of all possibilities, we consider four distinct regimes:

- (i) $C_{dd}/C_{da} = 0$, the direct-transfer regime;
- (ii) $0 < C_{dd}/C_{da} \ll 1$, the diffusion-limited transfer regime;
- (iii) $C_{dd}/C_{da} \geq 1$, the diffusion or hopping regime;
- (iv) $C_{dd}/C_{da} \gg 1$, the ultrafast migration regime.

In each of the above cases, the macroscopic kinetics depend differently on the number of donors and acceptors present, as well as on the values of the coefficients C_{dd} and C_{da} .

7.6.2.1 Case (i): Direct-transfer regime, $C_{dd}/C_{da} = 0$

This case applies when the transition rate for donor-to-donor transfer vanishes or equivalently when the donors are so dilute compared with the acceptors that little donor migration occurs. To begin, consider a single donor and acceptor pair in which the donor is excited at time $t = 0$. The probability that the donor is still observed in the excited state at time t can be immediately written down:

$$P_{A^*}(t) = e^{-t/\tau} e^{-k_{da}(R)t} \quad (7.84)$$

where τ is the lifetime of the donor excited state and the functional dependence of k_{da} on R is shown as a reminder of this quantity's origin in near-field multi-pole interactions. In order to derive a mean probability $\varphi(t)$ for observing the donor in the excited state, the previous expression must be averaged over all donor–acceptor separations R within the host volume V :

$$\varphi(t) = e^{-t/\tau} \int_V e^{-k_{da}(R)t} w(R) dV \quad (7.85)$$

Here, $w(R)$ represents the probability density for the acceptor to be located a distance R from the donor and is given for a random distribution of acceptors by $w(R) = 1/V$.

Increasing the number of acceptors to N_a and assuming a spherical sample volume with radius R_V , Equation (7.85) becomes

$$\varphi(t) = e^{-t/\tau} \left(\frac{4\pi}{V} \int_0^{R_V} e^{-k_{da}(R)t} R^2 dR \right)^{N_a} \quad (7.86)$$

Explicit evaluation of $\varphi(t)$ only requires insertion of a form for $k_{da}(R)$, followed by careful taking of limits $V \rightarrow \infty$ and $N_a \rightarrow \infty$. In the case of the general multi-pole interaction of order s , $k_{da}(R) = C_{da}/R^s$ and the final result is

$$\varphi(t) = e^{-t/\tau} e^{-\frac{4\pi}{3} n_a \Gamma(1-3/s) (C_{da} t)^{3/s}} \quad (7.87)$$

For the case of dipole–dipole coupling, $s = 6$ and the probability for observing the donor in the excited state reduces to

$$\varphi(t) = e^{-t/\tau} e^{-\frac{4\pi}{3} n_a \sqrt{\pi C_{da} t}} \quad (7.88)$$

which gives for the time dependence of the excited donor population simply

$$N_d^*(t) = N_d^*(0) \varphi(t) \quad (7.89)$$

The decay of the excited donors is thus nonexponential in the direct-transfer regime.

7.6.2.2 Case (ii): Diffusion-limited regime, $C_{dd}/C_{da} \ll 1$

This case applies when some degree of migration for the donor excitons occurs in the presence of a strong donor–acceptor interaction. One may therefore expect that following short-pulse excitation the decay of the donor population at early times will resemble that found in the direct-transfer regime. As the population density of donor excitons surrounding each acceptor is depleted, however, further donor–acceptor transfer will occur most probably through the eventual “diffusion” of donor excitons into the vicinity of acceptors. At late times, the population density of excited donors attains an approximately steady-state but nonuniform spatial distribution, implying that the late-time decay should be nearly exponential.

This “diffusion-limited” transfer problem can be formulated quantitatively by first reconsidering Equation (7.84) in the case of multiple acceptors:

$$P(\mathbf{r}, t) = e^{-t/\tau} e^{-\sum_{i=1}^{N_a} k_{da}(\mathbf{r}-\mathbf{r}_i)t} \quad (7.90)$$

where $\mathbf{r}$ denotes the position of an excited donor and $\mathbf{r}_i$ the position of the i th acceptor. At this point, the excited donors are assumed to constitute a smooth (but

not necessarily uniform) spatial distribution that is characterized by the density function $n_d^*(\mathbf{r}, t)$. The above equation may then be rewritten

$$n_d^*(\mathbf{r}, t) = n_d^*(\mathbf{r}, 0) e^{-t/\tau} e^{-\sum_{i=1}^{N_d} k_{da}(\mathbf{r}-\mathbf{r}_i)t} \quad (7.91)$$

Differentiating with respect to time,

$$\frac{\partial n_d^*(\mathbf{r}, t)}{\partial t} = -n_d^*(\mathbf{r}, t)/\tau - \sum_{i=1}^{N_d} k_{da}(\mathbf{r}-\mathbf{r}_i) n_d^*(\mathbf{r}, t) \quad (7.92)$$

At this point, in order to include the effect of donor migration, a phenomenological diffusion term is added to Equation (7.92) that may be likened to heat flow due to a nonuniform temperature distribution. The final result is thus

$$\frac{\partial n_d^*(\mathbf{r}, t)}{\partial t} = D \nabla^2 n_d^*(\mathbf{r}, t) - n_d^*(\mathbf{r}, t)/\tau - \sum_{i=1}^{N_d} k_{da}(\mathbf{r}-\mathbf{r}_i) n_d^*(\mathbf{r}, t) \quad (7.93)$$

The procedure by which we arrived at this expression is valid provided the background distribution of unexcited donors is sufficiently uniform that a narrow range of donor–donor transfer times is at play. This situation occurs most accurately when the host lattice itself forms the donor system, or less precisely when a very high concentration of randomly distributed donors exists. With this understanding, and for dipole–dipole coupling, a solution for the time dependence of the *spatially averaged* excited donor density is (Yokota and Tanimoto, 1967)

$$n_d^*(t) \equiv \overline{n_d^*(\mathbf{r}, t)} = n_d^*(0) e^{-t/\tau} e^{-\frac{4\pi}{3} n_a \sqrt{\pi C_{da} t} \left[\frac{1+10.87x(t)+15.5x(t)^2}{1+8.743x(t)} \right]} \quad (7.94)$$

where $x(t) = DC_{da}^{1/3} t^{2/3}$ and the diffusion coefficient D is found to be

$$D = \frac{1}{6} C_{dd} n_d (25.3/R_{nn}) \quad (7.95)$$

This solution was derived assuming an fcc lattice of donor ions (with nearest-neighbor distance R_{nn}) within which a small number of acceptors is embedded. Inspection of Equation (7.94) shows that at early times the effect of diffusion is unimportant and the nonexponential direct transfer result of Equation (7.88) dominates the donor decay. At late times, the diffusion term dominates and a near-exponential decay results. A good description of luminescence decay of Eu^{3+} ions in $\text{Eu}(\text{PO}_3)_3$ glass doped with Cr^{3+} acceptor ions is provided by this model (de Gennes, 1958).

7.6.2.3 Case (iii): *Hopping or diffusion regime*, $C_{dd}/C_{da} > 1$

This case deserves particular attention as it forms the basis for modeling of energy-transfer processes in many solid-state energy-transfer systems. The reason is the simple form assumed by population rate equations based on this picture. The essential idea is that the donor migration is sufficiently fast that a uniform spatial density of donor excitons is attained and therefore each exciton experiences the same average acceptor environment. The excited donor population thus decays exponentially. Another way to view this case is that a given donor exciton makes many “hops” on the donor sublattice before a donor–acceptor transition occurs. The situation is analogous to gas-phase reaction kinetics for two reacting species.

Recall Equation (7.87) for the time dependence of the decay of an excited donor into an acceptor excitation in the direct-transfer regime, except apply it to the case of donor-to-donor migration and neglect single-ion decay of the donor:

$$\varphi(t) = e^{-\frac{4\pi}{3}\Gamma(1-3/s)n_d(C_{dd}t)^{3/s}} \quad (7.96)$$

Thus this expression gives as a function of the multi-pole order s the probability that a donor exciton that has arrived at a particular site at time $t = 0$ still resides there at time t . The fundamental assumption of the hopping model is that while one recognizes that a distribution of hopping times would actually be observed in a real system, this complexity is removed by taking a suitably defined average hopping time as accurately reflecting the exciton motion. This average hopping time is readily calculated from Equation (7.96):

$$\tau_{hop} = \frac{\int_0^\infty t\varphi(t) dt}{\int_0^\infty \varphi(t) dt} = \frac{\Gamma(2s/3)}{\Gamma(s/3)} \left[\frac{4\pi}{3} n_d \Gamma(1-3/s) \right]^{-s/3} C_{dd}^{-1} \quad (7.97)$$

Now, the probability that an excited donor transfers its energy to an acceptor in the time τ_{hop} is given by

$$P = 1 - \varphi_a(\tau_{hop}) \quad (7.98)$$

where

$$\varphi_a(\tau_{hop}) = e^{-\frac{4\pi}{3}\Gamma(1-3/s)n_a(C_{da}\tau_{hop})^{3/s}} \quad (7.99)$$

is the probability that the excited donor has *not* transferred its energy to an acceptor in the absence of all other (unexcited) donors. Because we have supposed that $C_{dd} > C_{da}$, P is by hypothesis small; likewise is the argument of the exponential in Equation (7.99), and thus

$$P \cong \frac{4\pi}{3} n_a \Gamma(1-3/s) (C_{da}\tau_{hop})^{3/s} \quad (7.100)$$

To obtain the time rate of change of the excited-donor population, set

$$\frac{1}{n_d^*} \frac{dn_d^*}{dt} = \frac{dP}{dt} \cong -\frac{P}{\tau_{hop}} = -\frac{4\pi}{3} n_a \Gamma(1 - 3/s) (C_{dd} \tau_{hop})^{3/s} (\tau_{hop})^{-1} \quad (7.101)$$

Allowing also for the single-ion decay of the excited donors with time constant τ , the final result is

$$\frac{dn_d^*}{dt} = -\frac{n_d^*}{\tau} - \frac{4\pi}{3} \Gamma(1 - 3/s) (C_{dd} \tau_{hop})^{3/s-1} n_d n_d^* \quad (7.102)$$

which for dipole–dipole coupling gives

$$\frac{dn_d^*}{dt} = -\frac{n_d^*}{\tau} - \left(\frac{2\pi}{3}\right)^3 n_d \sqrt{6C_{dd}C_{da}n_a} n_d^* \quad (7.103)$$

The last expression shows that the time rate of change of the excited donor population depends on the bilinear product of the acceptor and excited-donor concentrations, just as they would in a gas-kinetics problem, and on the square root of the resonant and nonresonant transfer coefficients.

As a point of comparison, another approach to calculating the decay of the excited-donor population is possible with different starting assumptions. In a fast-diffusion picture of the exciton migration (as opposed to the slow-diffusion model considered above), one assumes a differential equation for the excited-donor population of the form

$$\frac{\partial n_d^*(\mathbf{r}, t)}{\partial t} = D \nabla^2 n_d^*(\mathbf{r}, t) - \frac{1}{\tau} n_d^*(\mathbf{r}, t) \quad (7.104)$$

to which is applied the boundary conditions

$$n_d^*(\mathbf{r}, 0) = n_{d,0}^*, n_d^*(r < R_c, t) = 0 \quad (7.105)$$

The latter condition supposes that within some critical distance R_c of an acceptor, the excited-donor population is zero. In this model, the critical distance as well as the diffusion coefficient D will depend on the geometry of the donor sublattice; their calculation requires simplifying assumptions akin to those used in Section 7.6.2.2. Other than single-exciton relaxation at the rate $1/\tau$, the rate of the excited-donor decay is given by the total flux of excitons that move across a spherical boundary of radius R_c surrounding each acceptor and summing over all acceptors.

This situation is reminiscent of the case considered in the previous section, except that the precise details of the donor-to-acceptor transfer embodied in the last term of Equation (7.93) have been subsumed into the simplifying boundary condition just described. The solution to Equation (7.104) as given by Chow and Powell (1980)

is

$$n_d^*(\mathbf{r}, t) = n_{d,0}^* \left[1 - \frac{R_c}{r} \operatorname{erfc} \left(\frac{r - R_c}{2\sqrt{Dt}} \right) \right] e^{-t/\tau} \quad (7.106)$$

where $\operatorname{erfc}(x)$ is the complementary error function for the argument x . From this result can be derived the total flux of excitons across the boundary defined by R_c :

$$\Phi(t) = 4\pi DR_c n_a n_{d,0}^* e^{-t/\tau} \left(1 + \frac{R_c}{\sqrt{\pi Dt}} \right) \quad (7.107)$$

In terms of the exciton density in regions of the host far removed from any acceptors, namely $n_{d,0}^* e^{-t/\tau}$, an effective time-dependent rate constant for donor–acceptor transfer may be written as

$$k_{da}(t) = 4\pi DR_c n_a \left(1 + \frac{R_c}{\sqrt{\pi Dt}} \right) \quad (7.108)$$

Now again since the exciton migration is presumed to be fast, we make the approximation $R_c/\sqrt{\pi Dt} \ll 1$ and thus the excited donors transfer energy to acceptors at the constant rate

$$k_{da} = 4\pi DR_c n_a \quad (7.109)$$

If one now assumes dipole–dipole coupling and a cubic donor lattice, the rate constant in this fast-diffusion model reduces to

$$k_{da} = 21n_d (C_{dd}^3 C_{da})^{1/4} n_a \quad (7.110)$$

Although this rate constant cannot be rigorously and quantitatively compared with the analogous quantity obtained with the hopping model (see Equation (7.103)), it is instructive to compare the functional dependencies. As before, the decay of the donor population is proportional to the product of the acceptor and excited-donor populations with a rate constant proportional to the total donor concentration. On the other hand, a different dependence on the transfer coefficients is found. Such a disagreement is no doubt a consequence of the different approximations made in the two approaches. Indeed Monte Carlo simulations of migration and donor–acceptor transfer yield a markedly different rate constant for donor–acceptor transfer than predicted by the hopping model.

7.6.2.4 Case (iv): Ultrafast migration regime, $C_{dd}/C_{da} \gg 1$

Equation (7.87) shows that in the direct-transfer regime the time rate of change of the excited-donor population diverges in the limit $t \rightarrow 0$. This anomalous behavior results from the assumption of a continuous distribution function $w(R)$ for the

acceptor position over the range $0 < R < \infty$, when in fact a discrete distribution is more accurate. Thus for very small values of the mean hopping time, the hopping model breaks down and a different approach is required. This case applies when the donor ions greatly outnumber the acceptor ions and otherwise the rate of donor–acceptor transfer is much less than the exciton hopping rate.

The idea here is that the donor migration is so fast that only the very early time behavior of direct transfer is appropriate in describing the excited donor decay. In other words, ultrafast migration leads to donor–acceptor transfer only for very close exciton–acceptor encounters.

In order to calculate the correct form for the early-time direct-transfer behavior, recall that for a single donor excited at time $t = 0$ in the presence of a single acceptor located a distance R away, the probability that the donor ion is still excited at time t is

$$P(t) = e^{-t/\tau} e^{-k_{da}(R)t} \quad (7.111)$$

At early times,

$$P(t) = 1 - k_{da}(R)t \quad (7.112)$$

and thus for an ensemble of acceptors in a volume V , the average probability that the donor is still excited is

$$\varphi(t) = 1 - \frac{1}{N_L} \sum_i k_{da}(R_i)t \quad (7.113)$$

where N_L is the total number of lattice sites and the sum is over all possible lattice sites (regardless of whether an acceptor actually resides there) *except* the donor site. Taking a time derivative and allowing for a total of N_a acceptors within V ,

$$\frac{d\varphi(t)}{dt} = -\frac{N_a}{N_L} \sum_i k_{da}(R_i) \quad (7.114)$$

In the limit $V \rightarrow \infty$,

$$\frac{d\varphi(t)}{dt} = \frac{1}{n_d^*} \frac{dn_d^*}{dt} = -\alpha n_a \sum_i k_{da}(R_i) \quad (7.115)$$

where α is a lattice-dependent constant. The exciton decay is thus exponential in this regime, but unlike in the hopping or diffusion models just discussed, a linear dependence on the donor–acceptor transfer rate is obtained whereas no dependence on the donor–donor rate is expected. For dipole–dipole coupling and an fcc lattice (Manz, 1977), the above expression becomes

$$\frac{dn_d^*}{dt} = -4.22n_a \frac{C_{da}}{R_{nn}^4} n_d^* \quad (7.116)$$

where R_{nn} is the nearest-neighbor distance in the lattice.

7.6.3 Rate equation formulation of upconversion by radiationless energy transfer

The various regimes of radiationless energy transfer considered in detail in the previous discussion reveal a wide range of possible models and attendant subtleties in their interpretation. For example, we saw in Section 7.6.2.3 that the macroscopic kinetics expected for the hopping and fast-diffusion pictures yielded two distinctly different dependencies on the microscopic transfer coefficients C_{dd} and C_{da} . Nevertheless, both pictures implied a simple rate-equation form for the excited donor population kinetics:

$$\frac{dn_d^*}{dt} = -\frac{1}{\tau}n_d^* - K_{da}n_d^*n_a \quad (7.117)$$

where K_{da} is a macroscopic rate constant that characterizes the donor–acceptor transfer process, a result similarly applicable in the ultrafast migration regime.

We are thus led to propose – independent of the microscopic details of the migration and transfer steps – that for many cases of interest a description of population kinetics in terms of phenomenological rate equations will be effective in predicting those kinetics. All that is required is to measure values of the phenomenological rate constants. Indeed, in practical laser problems involving rare-earth-based active gain media, such a rate-equation description has been widely applied and largely successful in guiding optimization of laser performance.

We hasten to add, however, that not all energy-transfer experiments involving rare earths are so easily interpreted in terms of rate equations, at least if one insists on such a description with time-independent rate constants. Moreover, recall that the four cases considered in the Section 7.6 assumed weak excitation of the donor system. In the laser context, such an assumption is likely to break down under conditions of strong optical pumping. Consider a donor system composed of two-level ions and a system of acceptors in which the hopping model provides a good description of the population kinetics under weak excitation. Optical saturation of the donors must change the kinetics as the density of unexcited donors is decreased thereby inhibiting donor migration. The kinetics may then proceed according to a direct transfer process or it may assume the appearance of ultrafast migration. Nevertheless, as recommended by its empirical successes and because of the ease with which we can make further theoretical progress, we in this section and the next adopt the rate-equation approximation in order to understand radiationless upconversion mechanisms.

To proceed, consider an ensemble of donors and acceptors distributed within a solid host. As before, denote a donor ion by A and an acceptor ion by B, the latter possibly being of the same or of a different ionic species as A. For simplicity, we indicate the states of excitation of these ions by one or more asterisks, thus A means that the donor is in its ground state, while A^* means that the donor ion is

in the (unique) excited state from which a radiationless transfer of energy to the acceptor may occur. In order to discuss upconversion, three excitation states of the acceptor must be considered: B , B^* , and B^{**} , where the last symbol indicates that the acceptor is in the desired final state, namely, the upper laser level. Hence the upconversion step may be written



In this expression, it is assumed that the needed prior excitation steps $A \rightarrow A^*$ and $B \rightarrow B^*$ result from single-photon excitation, although in general they may result from additional radiationless transfer steps analogous to Equation (7.118). The latter remark hints at the range of complexity that upconversion mechanisms involving energy transfer may display. For example, instead of the process just described, radiationless transfer might promote a laser-active ion from its ground state to an intermediate state, after which single-photon absorption delivers the ion to the upper laser level. Combined with intervening single-ion nonradiative transitions such as those considered previously in the context of two-photon pumping, even more complex population kinetics are possible. For now, however, we simply assume that the donor state A^* and the acceptor state B^* are created by ordinary single-photon absorption and that the upconversion step occurs by the energy transfer step Equation (7.118). The task at this point is simply to write down and solve rate equations for the population kinetics.

Suppose the energy levels of species A and species B are as shown in Figure 7.15 and that the total concentration of these species are N_A and N_B , respectively. Optical pumping occurs at the frequency ω_p , which is resonant with both the $A \rightarrow A^*$

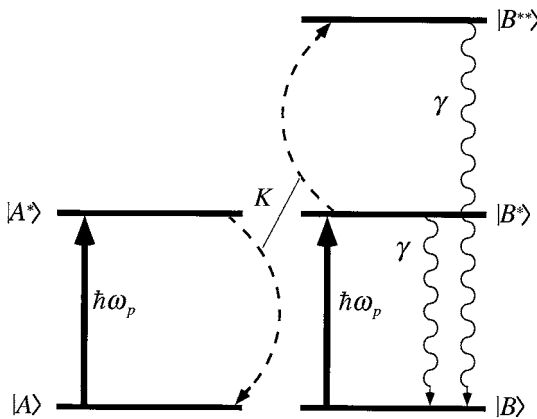


Figure 7.15: Energy-level diagram depicting pump and relaxation processes for an upconversion laser pumped with the help of radiationless energy transfer, as indicated by the linked dashed arrows.

and $B \rightarrow B^*$ transitions. Energy transfer from the donor A^* to the acceptor B^* is indicated by the linked dashed arrows and delivers the acceptor to the upper laser level B^{**} . In order to simplify the algebraic analysis, we will assume that the donor system is always optically saturated and that therefore the concentration of excited donors is pinned at $n_A^* \equiv N_A/2$. Rate equations for the populations in the three states of the B species are

$$\dot{n}_B^* = \frac{\sigma I}{\hbar\omega_p}(N_B - n_B^*) - Kn_A^*n_B^* - \gamma n_B^* \quad (7.119a)$$

$$\dot{n}_B^{**} = Kn_A^*n_B^* - \gamma n_B^{**} \quad (7.119b)$$

subject to the conservation condition

$$n_B + n_B^* + n_B^{**} = N_B \quad (7.119c)$$

In these expressions, K is a time-independent macroscopic rate constant that describes the donor–acceptor transfer, and we have assumed that single-ion decay to the ground state occurs for both excited states of the B species at the rate γ . Under steady-state conditions, $\dot{n}_B^* = \dot{n}_B^{**} = 0$, Equations (7.119) are readily solved for the population density in the upper laser level:

$$n_B^{**} = \frac{\frac{I}{I_s} \frac{KN_A}{\gamma}}{1 + 4\frac{I}{I_s} + \left(1 + \frac{I}{I_s}\right) \frac{KN_A}{\gamma}} N_B \quad (7.120)$$

where as before $I_s = \gamma\hbar\omega/\sigma$.

This last result displays an interesting feature. Consider Equation (7.120) in the limit of strong optical pumping, that is, $I \gg I_s$:

$$n_B^{**}(I \gg I_s) = \frac{KN_A}{4\gamma + KN_A} N_B \quad (7.121)$$

In this limit we see that if the product of the total donor concentration and macroscopic rate constant is large compared with the single-ion decay rate γ , then $n_B^{**} \rightarrow N_B$. This is a remarkable result: one sees that if good coupling between donors and acceptors exists, or if the donor concentration is large, then a 100% population inversion can be created in the B species. This circumstance would have been impossible if the system were pumped exclusively by optical means. On the other hand, we have ignored the possibility of *back transfer*, that is, the radiationless process $A + B^{**} \rightarrow A^* + B^*$, in formulating the rate equations. In general, such a process must be considered – it can be easily incorporated into the rate equations – but unlike optical pumping it is not thermodynamically required. For instance, if the forward transfer is a one-phonon-assisted *exothermic* process and if the energy

of that phonon is larger than kT , then the *endothermic* back-transfer process will be strongly inhibited. One can even imagine a laser system based on radiationless energy transfer between a pair of two-level systems that yields a population inversion in the acceptor species.

One can conceive other possible upconversion schemes based on a combination of optical pumping and energy transfer. For instance, it is possible that a laser-active ion may be effectively pumped with two or more purely radiationless transfers from the same donor state. Effective upconversion based on energy transfer may even be accomplished with a single species that performs the dual role of donor and acceptor. Examples of upconversion lasers based on all of these schemes will be described in the next chapter.

7.6.4 The photon avalanche

An important variation of cooperative upconversion that is also readily modeled with a system of rate equations deserves special attention. Known as the photon-avalanche mechanism, the basic idea is illustrated in Figure 7.16. We imagine two identical ions in sufficient proximity that good electric-multi-pole coupling exists between them. Ignoring for now the single-ion decay steps $2 \rightarrow 1$ and $1 \rightarrow 0$, the process begins by supposing that a small population already exists in the intermediate state $|1\rangle$ and that the pump laser is resonant with the $1 \rightarrow 2'$ transition,

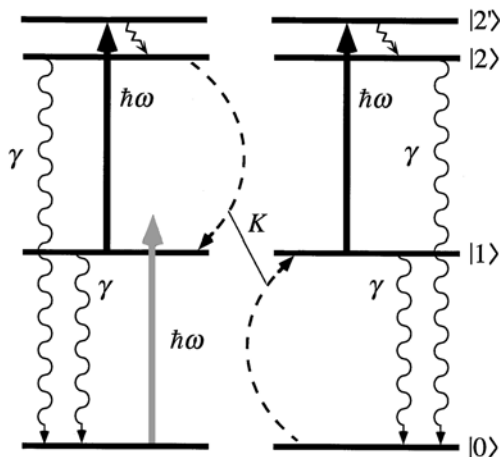


Figure 7.16: Energy-level diagram depicting pump and relaxation processes for a photon-avalanche pumping scheme. In particular, cross-relaxation, as indicated by the linked dashed arrows, creates population in the intermediate state $|1\rangle$, from which excited-state absorption of the pump radiation populates the upper laser level. A small seed population is established in state $|1\rangle$ through weak overlap of the pump frequency with the $0 \rightarrow 1$ transition (gray arrow).

as shown in the figure. Subsequent nonradiative relaxation of the $|2'\rangle$ state then delivers population to the upper laser level. Next, owing to multi-pole coupling between the ions, the radiationless *cross-relaxation* process comprising the $2 \rightarrow 1$ transition of one ion and the $0 \rightarrow 1$ transition for the second ion delivers both ions to the intermediate $|1\rangle$ state. With an intense pump laser resonant with the $1 \rightarrow 2'$ transition, however, both ions are rapidly delivered to the upper laser level. Thus despite a temporary depletion of population in the state $|2\rangle$, the net effect of the process up to this point may be viewed as the investment of one unit of $|2\rangle$ -state population in return for two units, at least as long as the optical pumping rate is larger than the spontaneous relaxation rates of either the $|2\rangle$ or especially the $|1\rangle$ state.

Recognize the implication of this involved sequence of events: a factor of 2 increase in the population of the high-lying upper laser level has been obtained without the need for direct optical pumping out of the ground state. More importantly, additional cycles of the process successively double and redouble the upper-state population, hence the moniker “photon avalanche” by which this phenomenon is known. A photon avalanche can rapidly build a large population in the upper laser level even when the initial “seed” population in the intermediate state is small. This seed population may arise from weak ground-state absorption of the (nonresonant) pump laser, or more simply it may exist as an equilibrium thermal population.

Of course, we have been discussing an idealized photon-avalanche process in which no population is lost anywhere in the sequence by either radiative or nonradiative relaxation back to the ground state. Good photon-avalanche pumping also requires that the impurity ions be distributed in a geometry favorable to rapid cross-relaxation. In particular, if one confines attention to a single pair of ions that have undergone one cycle of the avalanche, a third ion must be engaged before the process can continue. A large average ion density, ion clustering, or rapid resonant migration of either the $|1\rangle$ - or the $|2\rangle$ -state excitations is therefore necessary in order to further the avalanche build up.

However, it is not at all necessary for the avalanche “gain” to equal 2, as it was in our idealized description above. A rate-equation model of the photon avalanche mechanism, including the relaxation processes shown in Figure 7.16, is given by the set of equations

$$\dot{n}_2 = \frac{I\sigma}{\hbar\omega_p}n_1 - Kn_0n_2 - \gamma n_2 \quad (7.122a)$$

$$\dot{n}_0 = \frac{I\sigma_0}{\hbar\omega_p}n_0 - Kn_0n_2 + \gamma(n_0 + n_2) \quad (7.122b)$$

where we have supposed that very fast nonradiative relaxation occurs on the $2' \rightarrow 2$ transition. We have also supposed that seeding of the population in level 1

is accomplished by weak optical pumping from level 0 (i.e. $\sigma_0(\omega_p) \ll \sigma(\omega_p)$) with the same laser that is resonant with the $1 \rightarrow 2'$ transition. As usual, the population densities must satisfy the conservation rule

$$n_0 + n_1 + n_2 = N \quad (7.122c)$$

Setting the time derivatives to zero and, moreover, dividing the resulting equations by $N\gamma$, we have

$$0 = S\rho_0 - \kappa\rho_0\rho_2 - \rho_2 \quad (7.123a)$$

$$0 = -S_0\rho_0 - \kappa\rho_0\rho_2 + (\rho_0 + \rho_2) \quad (7.123b)$$

and

$$\rho_0 + \rho_1 + \rho_2 = 1 \quad (7.123c)$$

where $\rho_i = n_i/N$ is the relative population density in the state $|i\rangle$, $\kappa = KN/\gamma$ is the relative cross-relaxation rate, $S = I\sigma(\hbar\omega_p\gamma)^{-1}$ is the relative pump intensity, and $S_0 = I\sigma_0(\hbar\omega_p\gamma)^{-1}$ is the relative seed intensity. The solution for the relative population density in level 2 is

$$\rho_2 = \frac{\sqrt{4\kappa SS_0(S+1) + [(S+1)(S_0+1) - \kappa(S-1)]^2} - [(S+1)(S_0+1) - \kappa(S-1)]}{2\kappa(S+1)} \quad (7.124)$$

Figure 7.17 shows a plot of this quantity as a function of the relative pump intensity S for various values of the relative seed intensity S_0 (and fixed relative cross-relaxation rate, κ) and for various values of κ (for fixed S_0). Note that at high pump intensities and fixed κ , the population in the upper laser level is independent of the seed intensity, whereas for fixed seed intensity, the avalanche threshold and the limiting relative population density in level 2 is highly sensitive to the relative cross-relaxation rate.

A little more insight into the avalanche process can be gained by defining the avalanche gain as

$$G = \frac{S\rho_1}{\kappa\rho_0\rho_2} \quad (7.125)$$

Figure 7.18(a) shows a plot of the avalanche gain as a function of the relative pump intensity for a fixed relative cross-relaxation rate of $\kappa = 5$, while Figure 7.18(b) shows the level 2 population density as a function of the gain so calculated. The latter figure illustrates the clear threshold for population build up in the $|2\rangle$ state at a point well below the ideal avalanche gain of 2.

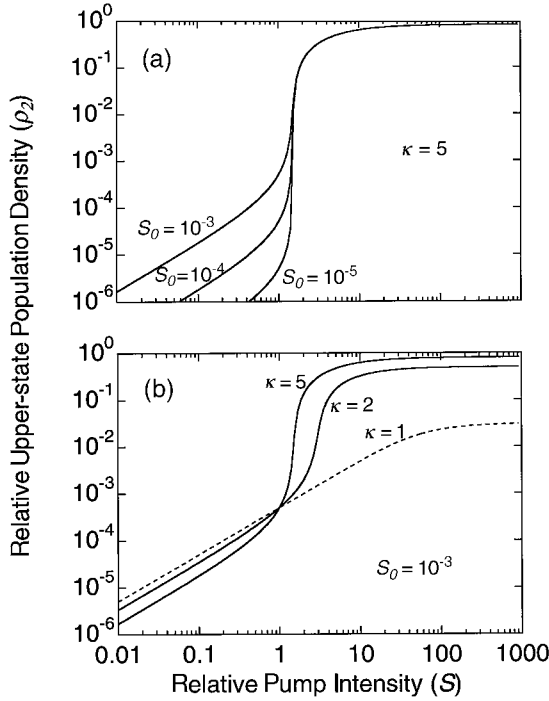


Figure 7.17: Relative population of the upper laser level in a system driven by a photon avalanche process. In (a), the effect of different rates of seed pumping is shown for a fixed rate of cross-relaxation, while in (b) the effect of different rates of cross-relaxation is shown for a fixed rate of seed pumping.

7.7 ESSENTIALS OF LASER PHYSICS

So far we have been exclusively discussing those issues in upconversion lasers that concern the creation of population in high-lying levels of gain-active rare-earth ions. In this last section we start with the assumption that optical gain has somehow been created in an appropriate medium and present the essentials of how laser oscillation and coherent optical output are then produced. In the spirit of previous sections we continue with a rate-equation description of the relevant physics, an approach that should make obvious how to construct a complete mathematical model of upconversion laser operation. Nonetheless the treatment here is by no means comprehensive and is intended only to provide enough practical knowledge to suggest how all the pieces fit together and to make more accessible the detailed discussions of upconversion laser operation appearing in the next chapter. For more in-depth understanding, readers are advised to consult the extensive accounts of laser physics and engineering such as those in Siegman (1986), Verdeyan (1981), and Powell (1998).

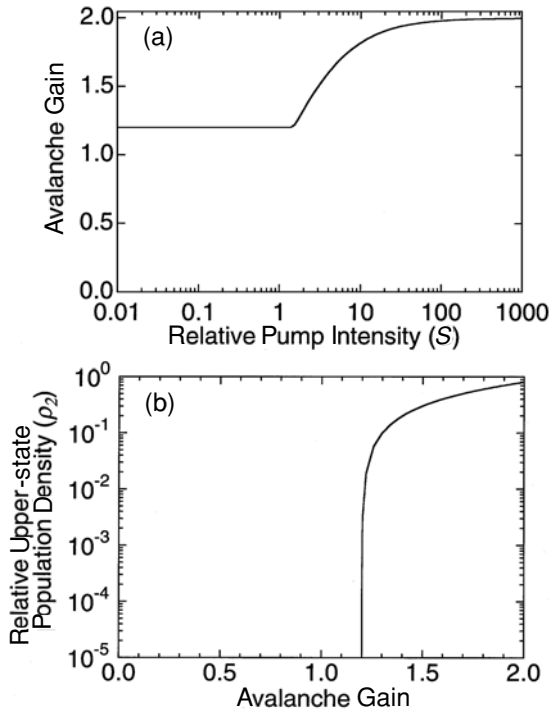


Figure 7.18: (a) Avalanche gain as a function of relative pump intensity for a relative cross-relaxation rate of $\kappa = 5$. (b) The relative population density obtained in the upper laser level for the avalanche gains shown in (a).

7.7.1 Qualitative picture

Two ingredients make a laser: amplification and feedback. An analogy familiar to everyone is the sound of a public address system and a microphone placed in the same room. The microphone picks up random noise in the room and thereby supplies a weak signal to an electronic audio amplifier. Upon amplification, which occurs over a bandwidth of perhaps 15 kHz, the electronic signal is converted back to an acoustic signal with the help of speakers placed in the room. If, after propagation and reflection off of the room's walls, the acoustic signal then arrives back at the microphone at a level *larger* than the original input, the process repeats indefinitely with the sound volume increasing without limit. Of course, in practice the acoustic power can never exceed the electrical power that supplies the amplifier, and therefore the amplifier output must saturate at some well-defined level determined by this supplied power. More significantly, the annoying sound heard by the room's occupants bears no relation to the original acoustic noise: although gain exists over much of the audible frequency range, the steady-state acoustic signal is a very loud simple tone of well-defined and stable frequency. The *amplitude* of this

tone relative to the power drawn from the line cord is determined by such factors as the gain and efficiency of the amplifier, the sensitivity of the microphone, and the acoustic reflectivity of the room walls. The *frequency* of the tone is determined by that combination of factors just named that yields the highest round-trip gain relative to the round-trip loss. Even if a small difference in gain or loss distinguishes two nearby frequencies, the frequency that experiences the lesser net gain will fade as power is increasingly devoted to sustaining the frequency of higher net gain.

In the optical realm, a population inversion in an atomic system will act as an amplifier for an input beam. If feedback to this amplifying medium is provided by a pair of mirrors that form a stable optical resonator,¹ feeding of some optical “noise” to the medium will quickly set the system into oscillation just as occurred in our acoustic analog. In the optical case, such noise may be derived from either blackbody radiation in the surroundings or spontaneous emission from the gain medium itself. (Similarly, in the acoustic analog, electronic noise in the amplifier might set the system into oscillation.) Presuming that the gain medium is continuously pumped by some means, the optical power within the cavity increases up to saturation just as before. If, moreover, the gain medium is homogeneously broadened, then a highly monochromatic optical field – at a frequency for which the net gain is a maximum – develops in the resonator for the same reasons as stated in our acoustic analog.

7.7.2 Rate equations for continuous-wave amplification and laser oscillation

7.7.2.1 Small signal gain

The first task is to reconsider a medium composed of a homogeneously broadened two-level system with lower state $|a\rangle$ and upper state $|b\rangle$ (Figure 7.19). Denote the population densities in these states by N_a and N_b . In the absence of population relaxation, the rate of change of these quantities when the medium is exposed to a monochromatic plane-wave field of intensity I and frequency ω is given in terms of absorption and stimulated-emission cross-sections:

$$\dot{N}_b = [N_a \sigma_{abs}(\omega) - N_b \sigma_{se}(\omega)] \left(\frac{I}{\hbar \omega} \right) \quad (7.126)$$

Written in this form, the rate equation may be interpreted in an intuitive way: The quantity $\Phi = I/\hbar\omega$ is just the flux of photons in the incident beam (units of photons per unit time per unit area) and thus the rate at which an absorber is excited

¹ The reader is referred to the references for discussions of the theory of optical resonators, in particular the books Siegman (1986) and Verdeyan (1981).

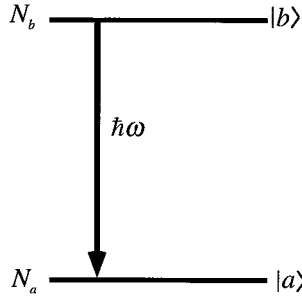


Figure 7.19: The two-level system as an optical amplifier. An applied field of frequency ω_0 induces stimulated emission for $N_b > N_a$.

by the incident field is given by the product of this flux and the effective “area” of the absorber, namely the absorption cross-section. Multiplication of this product by the total number of absorbers exposed to the beam yields the total number of absorption events per unit time. A similar argument holds for the rate of stimulated emission, making the total rate of change of the population in the $|b\rangle$ state the difference of the two rates. Count up all of the events occurring in a unit volume and Equation (7.126) results.

An important simplification of Equation (7.126) is obtained for nondegenerate levels; in this case, $\sigma_{se}(\omega) = \sigma_{abs}(\omega)$ and the rate of change of the upper-state population density is directly proportional to the inversion $N_b - N_a$. More generally, $\sigma_{se}(\omega) = (g_a/g_b)\sigma_{abs}(\omega)$ and the rate of change of the upper-state population density may be written

$$\dot{N}_b = \left[N_a - \frac{g_a}{g_b} N_b \right] \left(\frac{I \sigma_{abs}}{\hbar \omega} \right) = \left[\frac{g_b}{g_a} N_a - N_b \right] \left(\frac{I \sigma_{se}}{\hbar \omega} \right) \quad (7.127)$$

In an even more general case, the two cross-sections may refer to transitions between two broadband *manifolds* of sublevel states, provided the sublevel populations within these manifolds are rapidly thermalized and one agrees that N_a and N_b denote each manifold population as a whole.

In any case, we of course know that as the beam propagates through the medium its amplitude will grow or attenuate depending upon the sign of $\dot{N}_b$ that results from the medium’s response to the incident field. More specifically, for a thin slab of thickness dz the differential change of the beam intensity is

$$dI = -\hbar \omega \dot{N}_b = [N_b \sigma_{se}(\omega) - N_a \sigma_{abs}(\omega)] I dz \quad (7.128)$$

or in other words

$$\frac{dI}{dz} = g(\omega) I \quad (7.129)$$

where

$$g(\omega) \equiv N_b \sigma_{se}(\omega) - N_a \sigma_{abs}(\omega) \quad (7.130)$$

Assuming that the population densities N_a and N_b are only weakly perturbed by the incident beam (the *small-signal* limit) and, moreover, are constant throughout a finite length of the medium in the z direction, Equation (7.130) may be readily integrated to yield

$$I(z) = I_0 e^{g(\omega)z} \quad (7.131)$$

Hence for $N_a \sigma_{abs} > N_b \sigma_{se}$ the incident beam must diminish in amplitude on passing through the medium. On the other hand, if $N_b \sigma_{se} > N_a \sigma_{abs}$, the medium acts as an amplifier with a net gain of $G = e^{g(\omega)z}$. The quantity $g(\omega)$ in this weak-field limit is therefore termed the small-signal gain coefficient.

7.7.2.2 Large-signal gain and gain saturation in a three-level amplifier

In order to predict the behavior of an optical amplifier in the limit of a strong input field, however, a more explicit model of the amplifying medium is required. Figure 7.20 thus shows a specific energy-level structure for which we wish to write down a complete rate-equation description of pumping, relaxation, and stimulated emission processes in the amplifier. The atomic system consists of just three states, $|0\rangle$, $|1\rangle$, and $|2\rangle$, and is pumped by an unspecified mechanism that transfers population out of the ground state to the upper laser level always at the rate R , independent of the ground-state population. This is equivalent to the assumption that the system need only be weakly pumped. Spontaneous relaxation of state $|2\rangle$, which occurs at the rate γ_2 , is assumed to occur only to state $|1\rangle$. Similarly, γ_1 is the relaxation rate of state $|1\rangle$ to the ground state. In the presence of a unidirectional beam incident

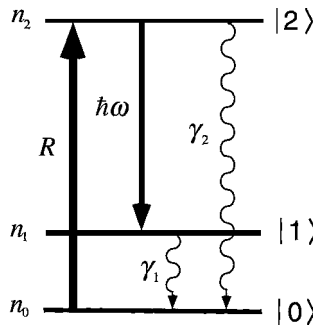


Figure 7.20: Three-level energy-level diagram depicting pumping and relaxation processes for an optical amplifier at the frequency ω_0 .

on a medium consisting of an ensemble of such three-level systems, the population densities in the two excited states must satisfy

$$\frac{dn_2}{dt} = R - \frac{I}{\hbar\omega} [\sigma_{se}(\omega)n_2 - \sigma_{abs}(\omega)n_1] - \gamma_2 n_2 \quad (7.132a)$$

$$\frac{dn_1}{dt} = \frac{I}{\hbar\omega} [\sigma_{se}(\omega)n_2 - \sigma_{abs}(\omega)n_1] + \gamma_2 n_2 - \gamma_1 n_1 \quad (7.132b)$$

where the incident beam is of intensity I and frequency ω resonant with the $2 \rightarrow 1$ transition. Now because we are ultimately interested in modeling a cw laser where steady-state conditions have been attained, the rate equations in this case may be readily solved for the population densities by setting the time derivatives of Equations (7.132) to zero. The result is a pair of algebraic equations whose solutions are

$$n_2 = \frac{R \left(\frac{1}{\gamma_2} + \frac{I}{I_s} \frac{\sigma_{abs}}{\sigma_{se} \gamma_1} \right)}{1 + I/I_s} \quad (7.133a)$$

$$n_1 = R/\gamma_1 \quad (7.133b)$$

The new quantity $I_s \equiv \hbar\omega\gamma_3/\sigma_{se}$ is of special significance in laser and optical amplifier problems; it is the *saturation intensity* and characterizes the intensity at which the incident beam reduces the level-2 population density by a factor of 2 below its value at low intensity. Another view is that the saturation intensity is that intensity equivalent to a flux of one photon passing through one stimulated-emission cross-section in one level-2 lifetime.

Equations (7.133) may now be used to write down the gain coefficient of the system at arbitrary intensity:

$$\begin{aligned} g(\omega) &= \sigma_{se}(\omega)n_2 - \sigma_{se}(\omega)n_1 \\ &= \frac{R \left[\frac{\sigma_{se}(\omega)}{\gamma_2} - \frac{\sigma_{abs}(\omega)}{\gamma_1} \right]}{1 + I/I_s} \\ &= \frac{g_0(\omega)}{1 + I/I_s} \end{aligned} \quad (7.134)$$

where

$$g_0(\omega) = R \left[\frac{\sigma_{se}(\omega)}{\gamma_2} - \frac{\sigma_{abs}(\omega)}{\gamma_1} \right] \quad (7.135)$$

is the small-signal gain coefficient defined earlier. We thus see that for $\sigma_{se}/\gamma_2 > \sigma_{abs}/\gamma_1$, the gain will be positive. In the more simple case where $\sigma_{se} \equiv \sigma_{abs}$, the condition for positive gain reduces to $\gamma_2^{-1} > \gamma_1^{-1}$. In short, if the level-2 lifetime

is larger than that of the lower laser level, that is, if the upper laser level acts as a *storage state* relative to the lower laser level, then the medium will amplify the applied field. In light of Equation (7.134), the strong-field expression for the rate of change of the applied field with distance through the medium thus becomes

$$\frac{dI}{dz} = \frac{g_0(\omega)}{1 + I/I_s} I \quad (7.136)$$

An interesting feature of this result is that once the beam intensity reaches sufficient amplitude, *gain saturation* occurs and the rate of increase of the intensity with distance diminishes. Indeed, the gain is reduced by a factor of 2 when $I = I_s$ and vanishes in the limit $I \rightarrow \infty$. Therefore should any loss mechanism exist in the beam path, the beam intensity must saturate at some finite value as it propagates through the medium.

As an important aside, note that our expression for I_s and the explicit functional dependence on intensity shown by the gain coefficient is actually more general than our simple model would appear to dictate: one typically finds for homogeneously broadened systems a large-signal gain coefficient in the form of Equation (7.134), except that the saturation intensity will assume the more general form

$$I_s = \hbar\omega/\tau_{eff}\sigma_{se} \quad (7.137)$$

where τ_{eff} is a characteristic equilibration time (absent any applied field) for the populations in the two states in response to an abrupt change in the pumping rate.

7.7.2.3 A model laser oscillator

So much for the amplification aspect of a laser oscillator; we now add the feedback ingredient. This is accomplished by enclosing the gain medium between a pair of mirrors that act as a stable cavity for the amplified optical field. As shown in Figure 7.21, the idea is that a simple noise signal due to either fluorescence from the

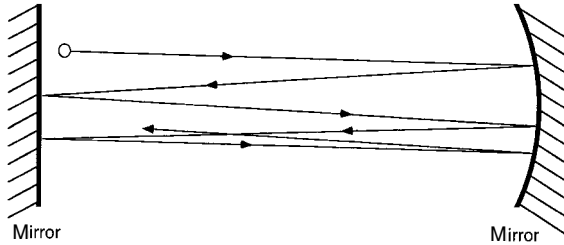


Figure 7.21: A stable optical cavity within which an initial light ray is perpetually reflected from the end mirrors. A gain medium within the cavity further amplifies the optical field after each reflection.

gain medium or from blackbody radiation present in the environment is amplified as it propagates through the medium. Upon reflection from one of the end mirrors back into the gain medium, the optical signal undergoes further amplification. Yet a third pass through the medium occurs after reflection from the second mirror. This cycle of reflection and amplification repeats indefinitely provided that a representative optical ray of the initial noise signal is perpetually trapped between the mirrors. Moreover, if both mirrors are perfectly reflecting and no scattering, diffractive, or absorptive loss is incurred by the beam within the medium, the intensity will increase without limit.

In practice, of course, we wish to extract optical power from the cavity. There are a number of ways this can be accomplished, but by far the simplest and most common is to replace one of the laser cavity end mirrors with a partial mirror, or *output coupler*, that reflects some fraction of the circulating intensity back into the gain medium while transmitting the balance to the outside world. In this circumstance, one sees that the circulating intensity will make an abrupt downward jump at the output coupler and then build up again during the subsequent round trip through the gain medium. Depending upon the saturation intensity and the output coupler's reflectivity, the circulating intensity must then attain some finite steady-state value at each location along the optical axis of the cavity and, moreover, will depend on the direction of propagation, at least in this one-dimensional picture.

These ideas can be readily made quantitative and interesting predictions thereby derived concerning the laser performance. Consider Figure 7.22, which shows an idealized laser oscillator composed of a gain medium and an optical cavity formed with a total reflector and an output coupler. The medium is homogeneously broadened with a gain coefficient given by an expression of the form Equation (7.134).

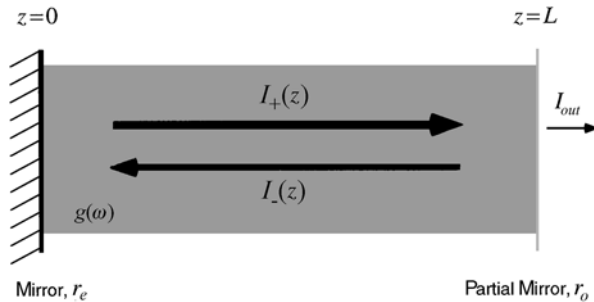


Figure 7.22: Idealized one-dimensional laser oscillator comprising an end mirror with reflectivity r_e , output coupler with reflectivity r_o (hence transmissivity $1 - r_o$), and a gain medium with gain coefficient $g(\omega)$. The forward and backward propagating field intensities are functions of position z within the cavity and denoted by $I_+(z)$ and $I_-(z)$, respectively.

The reflectivity of the total reflector, located on the left-hand side of the cavity at $z = 0$, is assumed to be imperfect and of reflectivity r_c while the reflectivity of the output coupler, located at $z = L$, is r_o . As shown in the figure, the circulating intensity consists of two pieces: a right-propagating component denoted by $I_+(z)$ and a left-propagating component denoted by $I_-(z)$. The intensity of the output beam is thus given by

$$I_{out} = (1 - r_o)I_+(z = L) \quad (7.138)$$

In turn, if the definitions $W_{\pm}(z) \equiv I_{\pm}(z)/I_s$ for the *relative* circulating intensities are made, Equation (7.136) implies that these quantities must satisfy the coupled equations

$$\frac{dW_+}{dz} = \frac{g_0(\omega)}{1 + (W_+ + W_-)} W_+ \quad (7.139a)$$

$$\frac{dW_-}{dz} = -\frac{g_0(\omega)}{1 + (W_+ + W_-)} W_- \quad (7.139b)$$

where in the second expression the minus sign appears because W_- increases in the negative z direction. Note also that the frequency ω is at the moment unspecified, but as we shall see later will be decided by the laser itself. Equations (7.139) immediately imply that

$$\frac{1}{W_+} \frac{dW_+}{dz} = -\frac{1}{W_-} \frac{dW_-}{dz} = \frac{g_0(\omega)}{1 + (W_+ + W_-)} \quad (7.140)$$

or equivalently that

$$\frac{d \ln(W_+)}{dz} = -\frac{d \ln(W_-)}{dz} \quad (7.141)$$

and thus finally that

$$\frac{d \ln(W_+ W_-)}{dz} = 0 \quad (7.142)$$

We therefore find the noteworthy result that even though the relative intensities are in general functions of position within the cavity, their product is equal to some unknown constant throughout the cavity:

$$W_+(z)W_-(z) = C \quad (7.143)$$

The differential equations for the relative intensities (Equations (7.139)) may then

be rewritten as

$$\begin{aligned} g_0(\omega) dz &= \frac{1 + W_+ + C/W_+}{W_+} dW_+ \\ &= -\frac{1 + W_- + C/W_-}{W_-} dW_- \end{aligned} \quad (7.144)$$

which on integration from $z = 0$ to $z = L$ readily yields the solutions

$$g_0(\omega)L = \ln \left[\frac{W_+(L)}{W_+(0)} \right] + [W_+(L) - W_+(0)] - C \left[\frac{1}{W_+(L)} - \frac{1}{W_+(0)} \right] \quad (7.145a)$$

and

$$-g_0(\omega)L = \ln \left[\frac{W_-(L)}{W_-(0)} \right] + [W_-(L) - W_-(0)] - C \left[\frac{1}{W_-(L)} - \frac{1}{W_-(0)} \right] \quad (7.145b)$$

Subtraction of these two equations gives

$$\begin{aligned} 2g_0(\omega)L &= \ln \left(\frac{1}{r_e r_o} \right) + (1 - r_o)W_+(L) - \left(1 - \frac{1}{r_e} \right)W_+(0) \\ &\quad - \frac{C}{W_+(L)} \left(1 - \frac{1}{r_o} \right) - \frac{C}{W_+(0)} (1 - r_e) \end{aligned} \quad (7.146)$$

where we have exploited the facts that $W_+(0) = r_e W_-(0)$ and $W_-(L) \equiv r_o W_+(L)$.

Equation (7.146) can be further modified by realizing that $W_-(L)W_+(L) = C$ as well as $W_-(0)W_+(0) = C$, so that both $r_o W_+(L)^2 = C$ and $W_+(0)^2 = r_e r_o W_+(L)^2$. Substitution of these identities into Equation (7.146) and solving for the relative intensity incident on the output coupler finally gives

$$W_+(L) = \frac{g_0(\omega) - \frac{1}{2} \ln \left(\frac{1}{r_e r_o} \right)}{1 - r_o + \sqrt{r_o/r_e} (1 - r_e)} \quad (7.147)$$

We thus obtain for the output intensity the result

$$\frac{I_{out}}{I_s} = (1 - r_o)W_+(L) = \frac{g_0(\omega) - \frac{1}{2} \ln \left(\frac{1}{r_e r_o} \right)}{1 + \sqrt{r_o/r_e} \left(\frac{1 - r_e}{1 - r_o} \right)} \quad (7.148)$$

As compact as is this last result, it tells us a number of facts about the laser's operation:

1 Threshold condition Note first that Equation (7.148) expresses a threshold condition for the small-signal gain coefficient in order for laser oscillation to occur, namely

$$g_0^{th} = \frac{1}{2L} \ln\left(\frac{1}{r_e r_o}\right) \quad (7.149)$$

Moreover, if the loss experienced by the circulating intensity upon reflection from the end reflector and the output coupler is hypothesized to be uniformly distributed over the length of the cavity, that is, if we write $r_e r_o = e^{-2\alpha_0 L}$, where α_0 is the loss per unit length, then $\alpha = \ln(1/r_e r_o)/2L$ and the threshold condition becomes simply

$$g_0^{th}(\omega) = \alpha_0 \quad (7.150)$$

In other words, oscillation will occur when the gain per unit length equals or exceeds the loss per unit length, or equivalently when the round-trip small-signal gain $G = e^{2g_0(\omega)L}$ equals or exceeds the round-trip loss. The situation is illustrated in Figure 7.23.

2 Slope efficiency If we adopt the simple three-level gain model of the previous section and substitute Equation (7.135) for the small-signal gain coefficient into

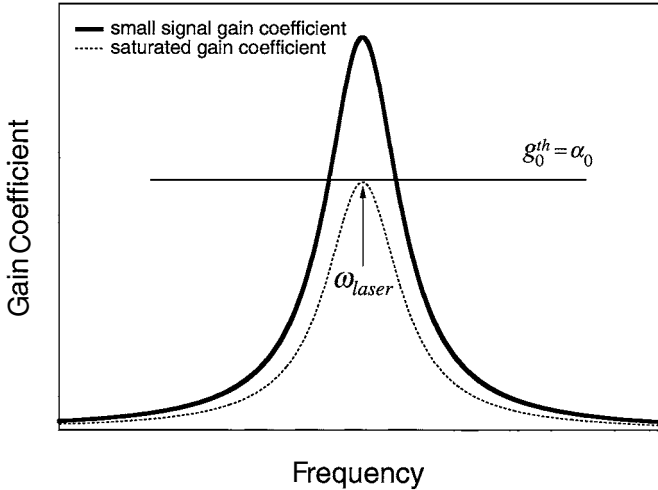


Figure 7.23: Small-signal and saturated gain coefficient as a function of frequency for a homogeneously broadened laser transition. Laser oscillation is possible at all frequencies for which the small-signal gain coefficient lies above the threshold value of α_0 . Oscillation is particularly favored at the maximum of the small-signal gain coefficient, thus yielding a saturated gain coefficient as shown by the dashed curve.

Equation (7.148) for the output intensity, the result is

$$\frac{I_{out}}{I_s} = \frac{R \left[\frac{\sigma_{se}(\omega)}{\gamma_2} - \frac{\sigma_{abs}(\omega)}{\gamma_1} \right] L - \frac{1}{2} \ln \left(\frac{1}{r_e r_o} \right)}{1 + \sqrt{r_o/r_e} \left(\frac{1 - r_e}{1 - r_o} \right)} \quad (7.151)$$

Further, if we suppose that the system is optically pumped with an intensity of I_p at a frequency ω_p resonant with the $0 \rightarrow 2$ transition, then the derivative of the output intensity with respect to the pump intensity is

$$\frac{dI_{out}}{dI_p} = \frac{\frac{I_s \sigma_{02}(\omega_p) n_0}{\hbar \omega_p} \left[\frac{\sigma_{se}(\omega)}{\gamma_2} - \frac{\sigma_{abs}(\omega)}{\gamma_1} \right] L}{1 + \sqrt{r_o/r_e} \left(\frac{1 - r_e}{1 - r_o} \right)} \quad (7.152)$$

In this expression, $\sigma_{02}(\omega_p)$ is the absorption cross-section of the $0 \rightarrow 2$ transition, assumed to be unsaturated by the pump laser. Equation (7.152) reveals that above threshold the output power of the laser will increase linearly with the pump power; the rate of increase is termed the *slope efficiency*. With respect to the practical engineering problems of constructing a commercially viable laser system, this quantity is of key significance as it tells us how economically the laser can be powered. A laser with good slope efficiency will be significantly less expensive to construct because the cost of the pump source is almost universally the highest of all the laser components. Still worse, this cost increases dramatically with the required power. We'll have more to say about slope efficiency when we later compare three- and four-level lasers.

3 Gain saturation Presuming that a pumping rate for the system has been imposed so as to yield a small-signal gain *above* the threshold condition, that is, if

$$G_0(\omega) = e^{2g_0(\omega)L} > 1/r_e r_o \quad (7.153)$$

then we know that when the laser reaches a steady state the actual round-trip *large-signal* gain $G(\omega)$ must be equal to the round-trip loss, that is, that

$$G(\omega) \equiv 1/r_e r_o \quad (7.154)$$

Because Equations (7.153) and (7.154) imply that $G(\omega) < G_0(\omega)$, the circulating intensity must partially saturate the round-trip gain to a point where the combined losses at the end reflector and output coupler are exactly compensated by the amplifying medium. This fact naturally holds even if other loss mechanisms exist within the cavity such as scattering and Brewster reflections. The dashed

curve in Figure 7.23 shows the effect of saturation on the gain line shape for a homogeneously broadened gain medium.

4 Oscillation frequency Presuming that the round-trip loss is frequency-independent, the laser will oscillate at the frequency where the small-signal gain is a maximum. This is a direct consequence of gain saturation: If the laser is somehow oscillating at a frequency other than at the peak of $g_0(\omega)$, then there will exist other frequencies where the large-signal gain must be larger. In this case, an optical signal experiencing this higher gain will build up quickly and overtake in amplitude the pre-existing signal. The medium then becomes more strongly saturated and the round-trip gain falls below threshold for the first signal. It is clear that only a very narrow band of frequencies near the peak of the small-signal gain curve will not suffer this fate, while due to gain saturation all other frequencies will experience gain below the oscillation threshold. This idea is illustrated in Figure 7.23. Gain saturation thus accounts, amongst other reasons, for the spectral bandwidth of laser oscillators being orders of magnitude narrower than the fluorescence linewidth. One can also see how to accomplish frequency tuning in a laser oscillator: By introducing frequency-dependent loss elements, the laser will choose the unique oscillation frequency where the product of the large-signal gain and the round-trip loss is unity.

5 Optimum output coupling Inspection of Equation (7.146) for the output intensity reveals that a maximum will occur for a unique value of the output coupler reflectivity. Figure 7.24 illustrates this point for interesting design parameters. In particular, note the natural result that higher gains (due, for example, to large stimulated-emission cross-sections) yield lower optimum reflectivities and higher output intensities, while perhaps more surprisingly higher internal loss also recommends lower optimum reflectivity.

7.7.2.4 Inhomogeneous broadening

Our consideration of exclusively homogeneously broadened transitions helps simplify the analysis of amplification, laser oscillation, and gain saturation because the amplified optical field interacts equivalently with *all* gain-active atoms in the medium. The salient complication for inhomogeneously broadened laser transitions is that we must consider the possibility that different subgroups of atoms will possess different center frequencies and therefore will contribute differently to the small-signal gain at a given frequency. What is more interesting, however, is that in the presence of strong inhomogeneous broadening multiple laser fields may *simultaneously* exist in the optical cavity with each field interacting with different

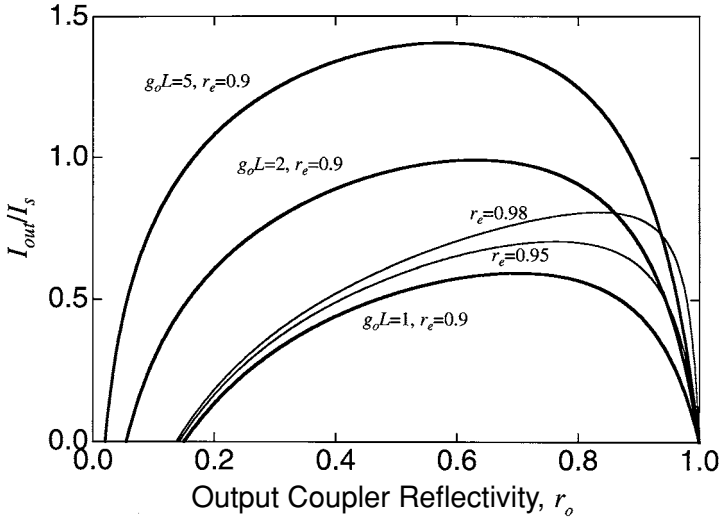


Figure 7.24: Relative output intensity of the one-dimensional model laser as a function of output coupler reflectivity, r_o . The bold curves show the output for fixed internal loss (quantified through the value of the end mirror reflectivity, r_e , and the indicated values of g_0L). The fine curves and the low-lying bold curve show the output for $g_0L = 1$ and three values of r_e .

atomic subgroups at their respective center frequencies. If the length of the cavity is L , then the cavity will support oscillation frequencies separated by the *longitudinal mode spacing* $\delta\omega = \pi c/Ln$, where n is the index of refraction of the gain medium.

Consistent with the location of the allowed longitudinal modes, gain saturation will occur independently over the full range of frequencies for which the small-signal gain lies above threshold. This situation is illustrated in Figure 7.25, which shows the effect of gain saturation of each homogeneously broadened subgroup on the overall inhomogeneously broadened small-signal gain line shape. In particular, *spectral holes* appear in the gain spectrum at each of the allowed longitudinal modes, a phenomenon commonly referred to as *saturation hole burning*.

We can take advantage of our study of homogeneously broadened systems to understand more quantitatively inhomogeneous saturation. We shall see that more subtle differences exist between the two cases than those just mentioned.

To proceed, represent a general distribution of transition center frequencies comprising the small-signal gain line shape of an inhomogeneously broadened system by the probability function $p(\omega_0)$. Thus if n_2 is the total number density of atoms found in the upper laser level, then $n_2 p(\omega_0) d\omega_0$ is the differential number of atoms per unit volume in the upper laser level whose center frequencies ω_0 fall within

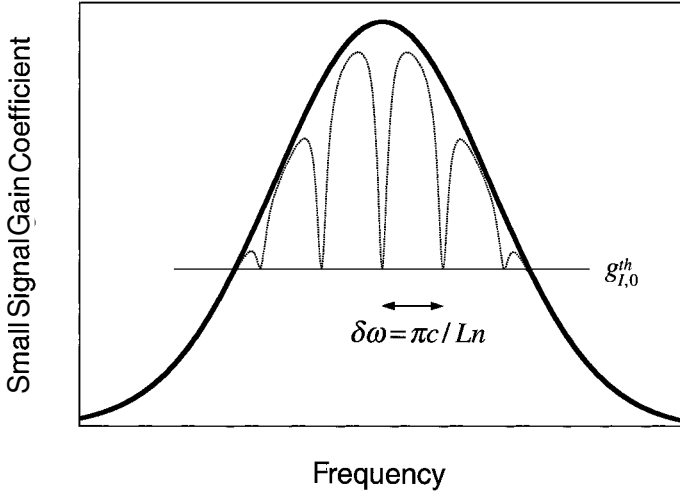


Figure 7.25: Gain saturation for an inhomogeneously broadened laser transition. Multiple frequencies may simultaneously oscillate, separated by the longitudinal mode spacing.

the frequency interval $d\omega_0$. Denoting the inhomogeneous gain coefficient by the symbol $g_I(\omega)$, this quantity in the presence of an applied field of intensity I_ω at the frequency ω is given by

$$g_I(\omega) = \int_0^\infty \frac{p(\omega_0)g_{h,0}(\omega_0, \omega)}{1 + I_\omega/I_s(\omega_0, \omega)} d\omega_0 \quad (7.155)$$

where

$$g_{h,0}(\omega_0, \omega) = \sigma_{se}(\omega_0, \omega)n_2 - \sigma_{abs}(\omega_0, \omega)n_1 \quad (7.156)$$

is the small-signal gain coefficient that would be observed at the frequency ω for atoms of center frequency ω_0 in the absence of inhomogeneous broadening. In turn, $I_s(\omega_0, \omega)$ is the saturation intensity for the subgroup of atoms of center frequency ω_0 when exposed to a beam of frequency ω . Taking the stimulated-emission cross-section to be of the Lorentzian form

$$\sigma_{se}(\omega_0, \omega) = \sigma_{se}^0 \frac{(\Delta\omega_h/2)^2}{(\omega_0 - \omega)^2 + (\Delta\omega_h/2)^2} \quad (7.157)$$

where σ_{se}^0 is the peak stimulated-emission cross-section and $\Delta\omega_h \equiv \gamma_{tot}$ is the full width at half maximum of the Lorentzian line shape function, then

$$g_I(\omega) = \sigma_{se}^0 \left(n_2 - \frac{g_2}{g_1} n_1 \right) \int_0^\infty \frac{p(\omega_0)(\Delta\omega_h/2)^2}{(\omega_0 - \omega)^2 + (\Delta\omega_h/2)^2 (1 + I_\omega/I_s^0)} d\omega_0 \quad (7.158)$$

Here I_s^0 is the saturation intensity at $\omega = \omega_0$, the peak of the homogeneous line shape function. Note also the interesting result that the denominator is of Lorentzian

form with an intensity-dependent width. This is the width of the “saturation hole” that is “burned” by the incident field. Denoting this width by

$$\Delta\omega_{hole}^2 \equiv \Delta\omega_h^2(1 + I/I_s^0) \quad (7.159)$$

Equation (7.158) may be rewritten

$$g_I(\omega) = \frac{\pi\sigma_{se}^0\Delta\omega_h}{4} \left(n_2 - \frac{g_2}{g_1}n_1 \right) \frac{\Delta\omega_h}{\Delta\omega_{hole}} \int_0^\infty p(\omega_0) \frac{(\Delta\omega_{hole}/2\pi)}{(\omega_0 - \omega)^2 + (\Delta\omega_{hole}/2)^2} d\omega_0 \quad (7.160)$$

This is as far as we can go without making additional assumptions. If, for example, we suppose that the inhomogeneous width is very much larger than the width of the saturation hole, then $p(\omega_0)$ varies slowly with respect to the Lorentzian factor by which it is multiplied in the above integrand. The gain coefficient then becomes

$$g_I(\omega) = \left\{ \frac{\pi\sigma_{se}^0\Delta\omega_h}{4} \left(n_2 - \frac{g_2}{g_1}n_1 \right) p(\omega) \right\} \frac{\Delta\omega_h}{\Delta\omega_{hole}} \quad (7.161)$$

The quantity in curly braces, however, can readily be shown to be just the small-signal gain coefficient $g_{I,0}(\omega)$ for an inhomogeneously broadened system, while according to Equation (7.159), $\Delta\omega_h/\Delta\omega_{hole} = \sqrt{1 + I_\omega/I_s^0}$. Making these substitutions into Equation (7.161) yields finally

$$g_I(\omega) = \frac{g_{I,0}(\omega)}{\sqrt{1 + I_\omega/I_s^0}} \quad (7.162)$$

Saturation of an inhomogeneously broadened transition therefore occurs more gradually than at the center frequency of a Lorentzian homogeneous system, a consequence of the interaction of the incident beam with homogeneous subgroups that neighbor the subgroup located at the signal frequency ω .

7.7.2.5 Three-level versus four-level systems

The three-state energy-level structure used in the previous sections to introduce the rate-equation approach to pumping and amplification (Figure 7.20) in laser gain media is rarely found in practice. No commercially available laser system operates in this way. A more likely possibility for a three-level laser is shown in Figure 7.26. In fact, the ruby laser, the first visible laser ever demonstrated, is well described by this model. As opposed to the previous three-level scheme, in this case the ground state serves as the lower laser level whereas the intermediate level serves as the upper laser level. As before, pumping is accomplished between the ground and highest excited state.

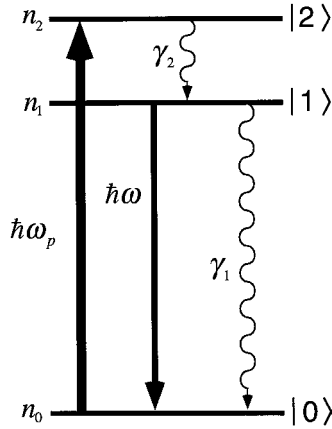


Figure 7.26: Energy-level diagram for the idealized three-level laser.

It is useful to investigate the gain characteristics in this new model as a function of pumping rate. Referring to Figure 7.26, we suppose that the relaxation processes are as shown, so that the rate equations are

$$\dot{n}_2 = \frac{I_p \sigma}{\hbar \omega_p} (n_0 - n_2) - \gamma_2 n_2 \quad (7.163a)$$

$$\dot{n}_1 = \gamma_2 n_2 - \gamma_1 n_1 \quad (7.163b)$$

where optical pumping with an intensity of I_p is applied at the frequency ω_p . The absorption cross-section on the $0 \rightarrow 2$ transition is σ , which is also assumed for convenience to be equal to the stimulated-emission cross-section of the reverse transition. Subject to the usual conservation condition

$$n_0 + n_1 + n_2 = N \quad (7.164)$$

the steady-state population inversion between the upper and lower laser levels is

$$n_1 - n_0 = N \frac{\frac{R}{\gamma_1} \left(1 - \frac{\gamma_1}{\gamma_2} \right) - 1}{\frac{R}{\gamma_1} \left(1 + \frac{2\gamma_1}{\gamma_2} \right) + 1} \quad (7.165)$$

where $R = I_p \sigma / \hbar \omega_p$ is the rate constant for pumping out of the ground state. For a “good” system, however, the relaxation rate out of the laser-pumped state will be large. In this circumstance, the numerator of the previous expression will be maximized while the denominator is minimized, both factors thus serving to maximize the population inversion. In the limit $\gamma_2 \gg \gamma_1$, one obtains the simple

result

$$n_1 - n_0 = N \frac{R/\gamma_1 - 1}{R/\gamma_1 + 1} \quad (7.166)$$

whereby a population inversion occurs only for $R/\gamma_1 > 1$.

Equation (7.166) reveals the principal disadvantage of three-level lasers, namely that a high rate of pumping is required before a population inversion is created with respect to the ground-state lower laser level. A related disadvantage is that true three-level laser systems are nearly universally operated in a pulsed mode as it is difficult to maintain a continuous inversion when the lower laser level acts as a storage state.

A little thought suggests that the disadvantages just named can be eliminated by adding one more energy level. The situation is depicted in Figure 7.27: By introducing an additional excited state to serve as the lower laser level, it is clear that generating a population inversion with respect to this state will be much easier given that the initial population in this state is zero. If also the rate of relaxation out of this new state back to the ground state is large, then continuous oscillation is likewise easier to accomplish.

Rate equations for this four-level scheme are easy to write down:

$$\dot{n}_3 = \frac{I_p \sigma}{\hbar \omega_p} (n_3 - n_0) - \gamma_3 n_3 \quad (7.167a)$$

$$\dot{n}_2 = \gamma_3 n_3 - \gamma_2 n_2 \quad (7.167b)$$

$$\dot{n}_1 = \gamma_2 n_2 - \gamma_1 n_1 \quad (7.167c)$$

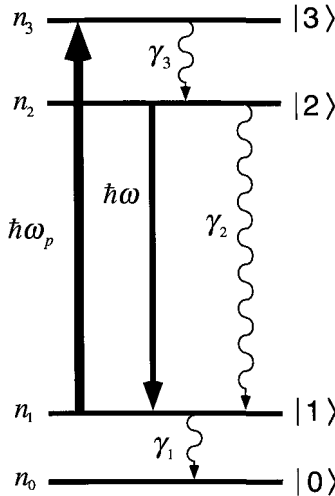


Figure 7.27: Energy-level diagram for the idealized four-level laser.

Proceeding as before, we invoke the conservation condition $n_0 + n_1 + n_2 + n_3 = N$, define the rate constant for optical pumping as $R = I_p \sigma / \hbar \omega_p$, and solve for the steady-state population inversion. Performing the algebra, the result is

$$n_2 - n_1 = N \frac{R(1 - \gamma_2/\gamma_1)}{R[1 + \gamma_2(1/\gamma_1 + 1/\gamma_3)] + \gamma_2} \quad (7.168)$$

This expression tells us that population inversion is achieved at *any* pumping rate so long as $\gamma_1 > \gamma_2$. A key advantage of a four-level laser system over the three-level system is thus made apparent: In addition to achieving inversion for any nonzero pumping rate, the essential task of creating gain in the system is shifted from that of providing a sufficiently powerful pumping mechanism to that of selecting appropriate material parameters. In particular, it is *easy* to find atomic systems for which $\gamma_1, \gamma_3 \gg \gamma_2$, in which case the inversion is

$$n_2 - n_1 = N \frac{R/\gamma_2}{R/\gamma_2 + 1} \quad (7.169)$$

Figure 7.28 compares the population inversions for three- and four-level laser models as a function of the relative pumping rate R/γ , where γ is the relaxation rate of the upper laser level. While many other laser level schemes may be readily

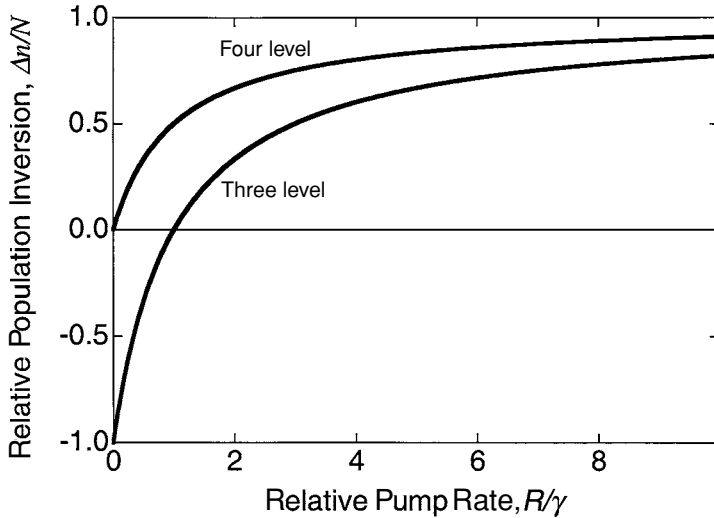


Figure 7.28: Comparison of the population inversions obtained as a function of relative pump rate, R/γ , for three-level and four-level laser models. Here γ is the relaxation rate of the upper laser level. The curves represent the case where the relaxation rate of the laser-pumped level and the lower laser level (in the four-level system) are large compared with γ .

imagined, in practice the four-level model exactly describes or well approximates the majority of laser gain media.

7.8 SUMMARY

Rare-earth spectroscopy is the fundamental underlying discipline of upconversion laser research. Key elements of this subject are that optical transitions in the visible and near-infrared range occur between spin–orbit levels belonging exclusively to the $4f$ shell of the trivalent ionic species. When rare-earth ions are dissolved in solid-state host materials, these levels are only weakly perturbed by the crystal field and thus appear very close in energy to their free space values. On the other hand, the breaking of spherical symmetry in a solid-state environment yields a “crystal-field splitting” of formerly degenerate spin–orbit levels; such symmetry breaking is in large part responsible for the optical activity of the ions. In particular, owing either to the intrinsic symmetry of the host lattice or to the modulation of the crystal field by phonons, few transitions remain dipole forbidden once a given rare-earth ion comes under the electrodynamic influence of a surrounding lattice.

As we saw, however, a compact understanding of the systematics of rare-earth spectroscopy is still possible through use of Judd–Ofelt theory and is specifically expressed by the three Judd–Ofelt parameters. Beyond the characterization of the strength of radiative transitions, the theory may also be applied to the rationalization of two-ion radiationless transitions, namely those arising from the interaction of one rare-earth ion’s electric field with the electric dipole moment of another. These energy-transfer processes between donor and acceptor ions, combined with single-ion optical pumping and the basic fact that just about any pair of rare-earth levels possess an electric-dipole matrix element means that a wealth of pathways for population dynamics exist even in lightly doped samples.

These simple features of rare-earth spectroscopy are what make for so many possible upconversion laser schemes. The game of producing significant populations in high-lying levels of rare-earth species goes no further than exploiting in combination appropriate resonant transitions – be they single-ion or derived from donor–acceptor energy-transfer mechanisms – and the need to beat out the gain quenching effects of unavoidable nonradiative transitions in the solid. Once an upconversion gain medium’s population dynamics are understood, conventional laser theory takes over. The combination of population dynamics and laser dynamics can be conveniently modeled with simple rate equations, although the resulting phenomena can be strikingly rich. In the next chapter we describe how these phenomena have been applied to the demonstration of working upconversion lasers.

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8

Upconversion lasers

The science of upconversion laser development can lay claim to many critical achievements on the path to a practical device. Among these are broad spectral coverage, room-temperature operation, single-wavelength pumping, and all-solid-state design. Arguable remaining milestones to overtake are high power, low cost, and easy manufacture. In this chapter we present a discussion of the origins and development of this subject with an eye toward conferring on researchers, who are new to the field, a comprehensive grasp of the published literature on upconversion laser experiments. After an account of the early history of upconversion laser research, we divide the topic into parts: the first comprises work involving bulk laser gain media – exclusively rare-earth-doped crystals – whereas the second comprises work on optical fiber gain media – primarily doped fluorozirconate glasses. Following a similar organization, nearly all the publications discussed in this chapter are reprinted under one cover in the anthology by Gosnell (2000). The chapter concludes with a brief discussion of potential directions for future research.

8.1 HISTORICAL INTRODUCTION

The notion of multi-photon upconversion in the solid state was first discussed by Bloembergen (1959) in the context of microwave and infrared quantum counters. The idea, depicted in Figure 8.1, is to exploit the photon counting capabilities of photomultiplier tubes sensitive only in the visible and near-infrared spectral range to detect upconversion emission. As seen in the figure, a single low-energy “signal” photon is converted to a single high-energy output photon when an upconverting pump photon supplied by an external source is simultaneously absorbed by the system. Transparent host materials doped with rare-earth and transition-metal ions were specifically named as potential detectors. The idea was further expanded upon by Porter and for the first time experimentally implemented by him (Porter 1961) in a Pr^{3+} -doped LaF_3 crystal pumped by a mercury discharge lamp. In this

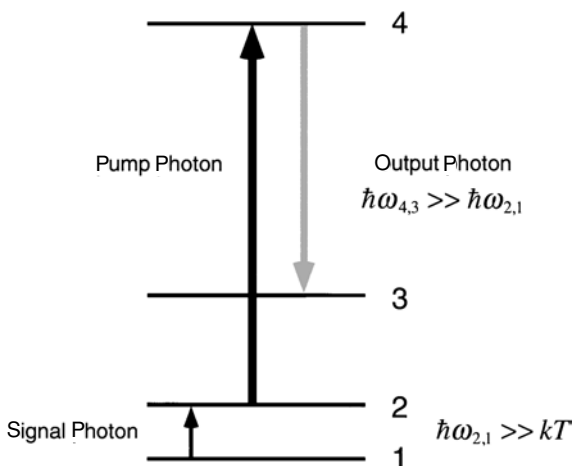


Figure 8.1: Generic energy-level diagram for a quantum counter: An infrared signal photon is detected in the visible by upconversion pumping of a high-lying energy state followed by short-wavelength radiative relaxation.

experiment, 2.3- μm radiation (coincident with the $\text{Pr}^{3+} \ ^3H_4 \rightarrow \ ^3H_6$ transition) from an incandescent lamp was detected at the upconverted wavelength of 618 nm.

Still in the context of quantum counters, the first description and realization of solid-state upconversion emission as mediated by radiationless energy transfer was given independently by Auzel (1966) in France and by the Russians Ovsyankin and Feofilov (1966). In particular, Auzel considered the absorption of signal photons at 970 nm by Yb^{3+} ions in a polycrystalline powder of $\text{WO}_4\text{Na}_{0.5}\text{Yb}_{0.5}$ doped with 2 wt% Er^{3+} (Figure 8.2). Upconversion then resulted through energy transfer from excited Yb^{3+} ions in the $^2F_{5/2}$ state to the $^4I_{11/2}$ state of Er^{3+} , followed by a second transfer from an excited Yb^{3+} ion to Er^{3+} that promoted the latter from its $^4I_{11/2}$ state to $^4F_{7/2}$. Subsequent nonradiative relaxation to the $^4S_{1/2}$ state and the thermally populated $^4H_{11/2}$ state then yielded upconversion emission on the famous green Er^{3+} transitions of $^4S_{1/2} \rightarrow ^4I_{15/2}$ (540 nm) and $^2H_{11/2} \rightarrow ^4I_{15/2}$ (525 nm). This experiment is especially interesting as it relies exclusively on *two* energy-transfer steps in order to pump the emitting states, in this way revealing the promise of such radiationless mechanisms in producing upconversion emission. Indeed, the Yb^{3+} ion later emerges as the most important of energy-transfer donor ions in upconversion laser experiments. Intensive research in upconversion phosphors and their applications followed for two decades after the pioneering experiments of 1966; Auzel (1973) and Wright (1986) have given comprehensive reviews of this work.

With the inspiration of the rapidly advancing research on quantum counters and upconversion phosphors, the first true upconversion laser experiments were

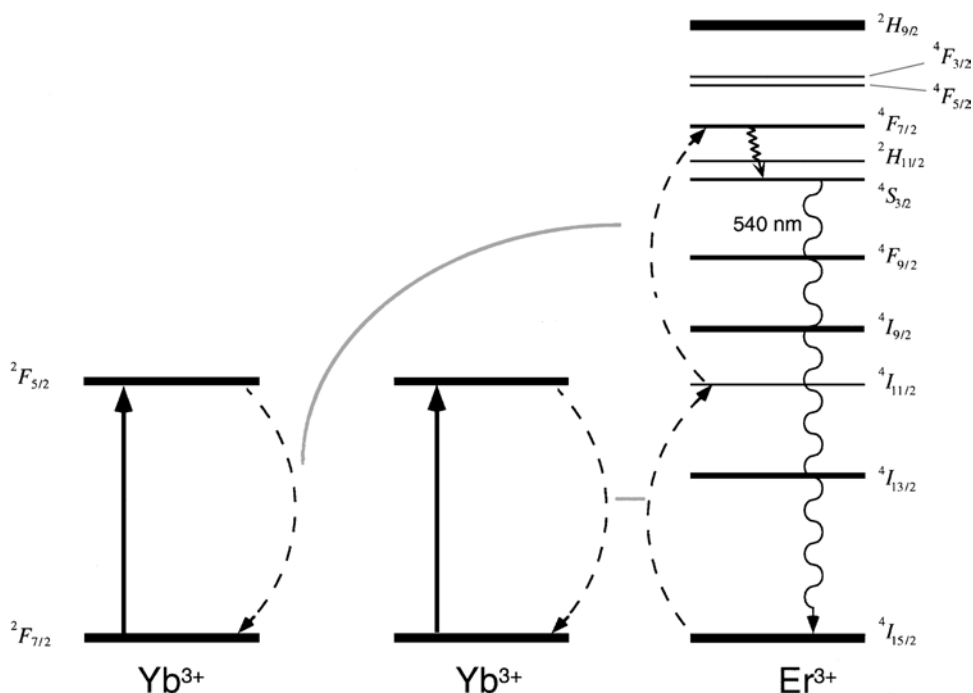


Figure 8.2: Mechanism for green upconversion luminescence by Er^{3+} ions. A sequential pair of radiationless energy-transfer steps from Yb^{3+} ions optically pumped in the infrared populates the Er^{3+} emitting level (Auzel, 1966).

undertaken in the early 1970s by Johnson and Guggenheim (1971). This singular work is especially remarkable in that nearly 20 years would pass before a visible upconversion laser would be demonstrated again.

Two laser crystals were prepared for these experiments. First, a BaY_2F_8 crystal was heavily doped with Yb^{3+} and lightly doped with Er^{3+} ions in order to yield a net composition of $\text{BaY}_{1.19}\text{Yb}_{0.75}\text{Er}_{0.06}\text{F}_8$, a choice no doubt motivated by Auzel's experiments five years before in which the goal was to induce multiple radiationless energy transfers to an acceptor ion from infrared-pumped Yb^{3+} . Dielectric coatings were then applied directly to the parallel faces of the prepared crystal, specifically a total reflector on one end and a 99.7% reflector at 670 nm on the other. At a sample temperature of 77 K, stimulated emission on the $^4F_{9/2} \rightarrow ^4I_{15/2}$ transition of Er^{3+} was then observed when the crystal was pumped in the infrared by a pulsed xenon flashlamp. The lamp's emission spectrum was filtered so as to remove all wavelengths shorter than 670 nm. That a true multiple energy-transfer mechanism involving Yb^{3+} and Er^{3+} ions was responsible for upconversion in this red laser system was asserted by the authors to be a necessary consequence of infrared pumping and the weak energy-transfer upconversion to be expected from Er^{3+} ions

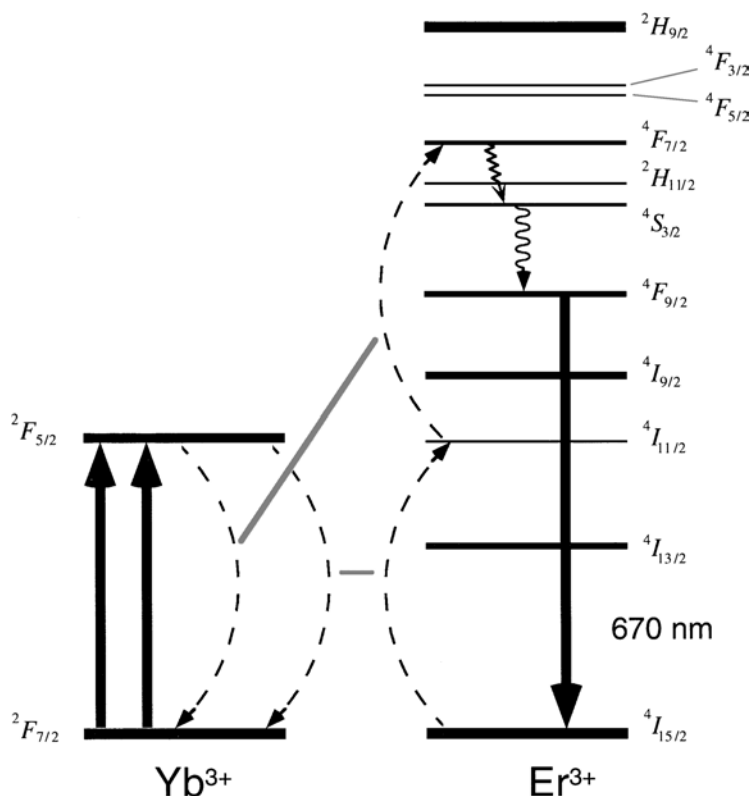


Figure 8.3: Red upconversion laser based on sequential energy transfer from infrared-pumped Yb^{3+} ions to Er^{3+} (Johnson and Guggenheim, 1971).

at such low concentration. An energy-level diagram illustrating the radiative and nonradiative transitions of this upconversion laser is shown in Figure 8.3.

In their second experiment, Johnson and Guggenheim prepared an analogous crystal of net composition $\text{BaY}_{1.4}\text{Yb}_{0.59}\text{Ho}_{0.01}\text{F}_8$ and again optically pumped a dielectric coated sample at 77 K with the near-infrared output of a pulsed xenon flashlamp. In this case, green laser output was obtained on the $^5\text{S}_2 \rightarrow ^5\text{I}_8$ transition at 551 nm. An interesting point regarding this Ho^{3+} laser is that when a wider-band flashlamp filter passing wavelengths longer than 400 nm was used, the laser threshold with respect to the lamp's output energy was reduced from 635 J to 355 J, thus indicating that even when direct pumping of the upper laser level was permitted, the majority of upper state pumping resulted from the Yb^{3+} -to- Ho^{3+} energy transfer upconversion process (Figure 8.4). This was another promising sign that upconversion lasers could be efficiently pumped and ultimately made technically practical. However, an important draw back of both the erbium and holmium lasers was that the ground crystal-field manifold served as the terminal

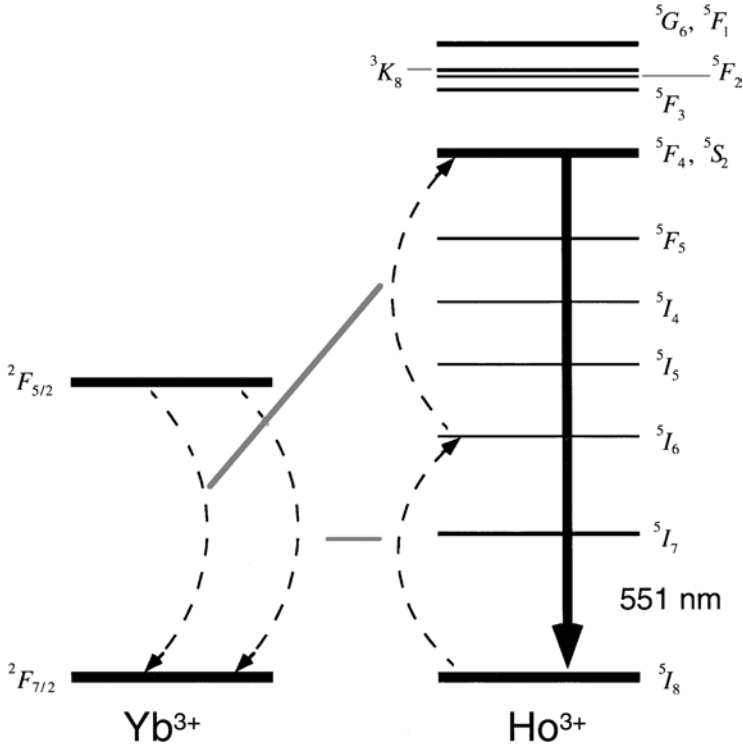
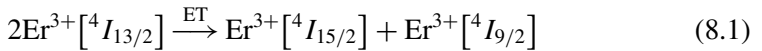


Figure 8.4: Green upconversion laser based on sequential energy transfer from infrared-pumped Yb^{3+} ions to Ho^{3+} (Johnson and Guggenheim, 1971).

laser level; sufficient gain was thus obtained only at cryogenic temperatures where quasi-four-level operation could be assured.

The fundamental feasibility of upconversion lasers having been nevertheless established, more than a decade would pass before further progress in the field would be made. An energy-transfer upconversion step was implicated in a 1983 report (Prokhorov, 1983 and references therein) in which properties of an $\text{Er}^{3+}:\text{YAG}$ laser operating on the ${}^4I_{11/2} \rightarrow {}^4I_{13/2}$ transition at $2.94\text{ }\mu\text{m}$ were investigated. Although this system was conventionally pumped into the upper laser level, cw lasing was observed despite the long lifetime of the lower laser level as a result of the rapid upconversion process

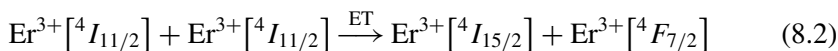


Because the $\text{Er}^{3+}[{}^4I_{9/2}]$ state so created quickly relaxed to the ${}^4I_{11/2}$ state, a further consequence of energy transfer was the recycling of pump energy back to the upper laser level. An almost exactly analogous mechanism was found responsible for *vibrational* upconversion pumping in CN^- -doped KBr (Tkach *et al.*, 1984).

In this case, pulsed excitation of the $\nu = 1$ vibrational state of the CN^- stretch mode at $4.8\text{ }\mu\text{m}$ led to phonon-assisted vibrational energy transfer between pairs of such singly-excited CN^- molecules. The result was upconversion pumping of the molecule's $\nu = 2$ state. Subsequent oscillation on the $\text{CN}^- 2 \rightarrow 1$ vibrational transition was then observed at $4.9\text{ }\mu\text{m}$, a wavelength nevertheless longer than the pump wavelength. Two years later, Pollack *et al.* (1986) exploited the same mechanism first discussed by Prokhorov (1983) in order to yield upconversion pumping of the $^4I_{11/2}$ state of Er^{3+} in CaF_2 doped at the 5–10 at% level. Optical pumping on the $^4I_{15/2} \rightarrow ^4I_{13/2}$ transition was provided by a pulsed Er^{3+} :glass laser operating at $1.54\text{ }\mu\text{m}$. This experiment is noteworthy in that room-temperature pulsed operation was obtained for the $2.8\text{ }\mu\text{m}$ $^4I_{11/2} \rightarrow ^4I_{13/2}$ transition. In their discussion of the laser's operation, the authors presented one of the earliest rate-equation models of upconversion for the explicit purpose of laser design. Room-temperature cw operation was later demonstrated in this same system (Xie and Rand, 1990) when pumped at $1.51\text{ }\mu\text{m}$ by a cw color-center laser.

Citing as motivation the advent of high-power GaAlAs diode lasers as potential infrared pump sources for visible light generation, a true visible upconversion laser pumped in the near-infrared was soon demonstrated by Silversmith *et al.* (1987), all working at IBM. It is here that the renaissance in upconversion laser development actually begins as the vision for a visible all-solid-state system is first stated in print. Using an YAlO_3 host material with dielectric cavity mirrors deposited directly onto the laser crystal, these authors demonstrated green laser emission on the $\text{Er}^{3+} ^4S_{3/2} \rightarrow ^4I_{15/2}$ transition at 550 nm. The laser was pumped (see Figure 8.5) by two-photon absorption from two independent cw dye lasers, one operating at 792 nm and the other at 840 nm – handy wavelengths because these were available from the new AlGaAs lasers. Green laser output was observed for laser-crystal temperatures ranging from 10 to 77 K, with a maximum output of 1 mW cw having been measured for ~ 400 mW of total pump power at a sample temperature of 30 K.

However, as strong green fluorescence was observed when the sample was exposed to only the shorter pump wavelength, the authors recognized that a phonon-assisted radiationless energy-transfer process also participated in pumping the upper laser level. Although other possibilities exist, the most likely energy-transfer pathway behind this observation is



after which nonradiative relaxation of the remaining excited ions builds population in the $^4S_{3/2}$ upper laser level. On the other hand, energy-transfer processes

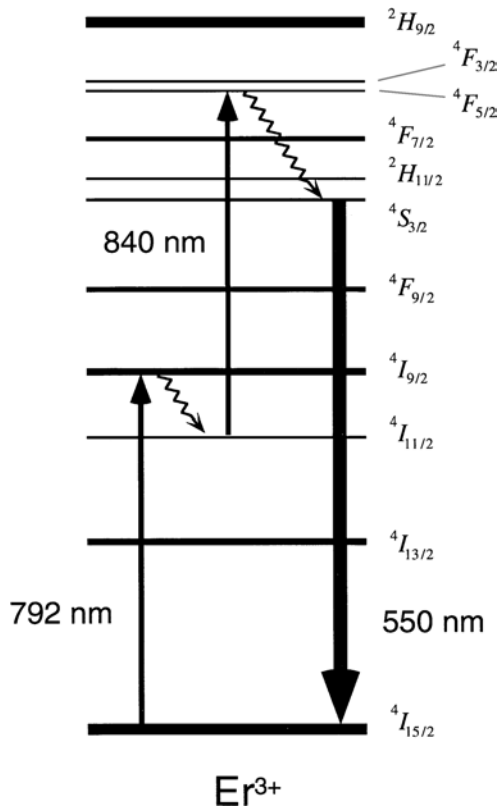


Figure 8.5: Green upconversion laser based on sequential two-photon pumping of Er^{3+} (Silversmith *et al.*, 1987).

deleterious to green laser operation also played a role in this experiment. In particular, fluorescence was observed from the high-lying $^2H_{9/2}$ and $^2P_{3/2}$ states – states not otherwise accessible given the wavelengths of the two pump lasers – thus indicating an indirect loss of pump energy that the authors proposed as a possible explanation for a 9% deficit in the estimated population inversion.

Although this first cw visible upconversion laser exhibited a relatively high total threshold pump power of 200–300 mW, and, moreover, required very low temperatures for optimum output, the essential practicality of coherent infrared-to-visible upconversion had been convincingly demonstrated. In response, a burst of activity from several research groups soon followed. Indeed, within a year Macfarlane *et al.* (1988) demonstrated a 380-nm violet laser based on $\text{Nd}^{3+}:\text{LaF}_3$ with a cw output power of 12 mW and went on to demonstrate a single-wavelength-pumped infrared-to-green upconversion laser (Lenth *et al.*, 1988) based on energy transfer in $\text{Er}^{3+}:\text{LiYF}_4$ (specifically Equation (8.2)). The latter experiment is interesting in that it underscored the importance of the host medium in determining the efficiency

of upconversion energy-transfer mechanisms. With an Er^{3+} concentration identical to that of their earlier work with $\text{Er}^{3+}:\text{YAlO}_3$, namely 1 at%, infrared pumping at 802 nm *alone* yielded a threshold pump power of just 80 mW, optimum operation at 45 K, and a total output power of 5 mW for a total pump power of only 240 mW.

Meanwhile, Pollack, Chang, and colleagues expanded their studies of upconversion-pumped infrared lasers to include new host materials, higher concentrations, and detailed rate-equation analysis (Pollack and Chang, 1988, 1990, Pollack *et al.*, 1989). Most significantly, in their 1989 work, these researchers demonstrated true upconversion laser output at 0.85 and 1.23 μm (in addition to output at 1.73 μm) for an $\text{Er}(5\%):\text{LiYF}_4$ laser crystal at 110 K, again pumped by a pulsed 1.53- μm source. What is compelling about this experiment, however, is that a *three-fold* energy-transfer mechanism is needed to explain the creation of population in the $^4S_{3/2}$ upper laser level – the same upper level as serves for the 550-nm green transition. Specifically, the authors argue for the simultaneous direct conversion of three Er^{3+} ions initially in the $^4I_{13/2}$ state: two into the ground state while the third ion is promoted to the $^4S_{3/2}$ state. The following year, Xie and Rand (1990a) claimed exactly the same mechanism to account for their observation of cw upconversion output at 855 nm in $\text{Er}(5\%):\text{CaF}_2$ pumped at 1.51 μm . These workers later demonstrated visible output from such a “trio” upconversion laser (Xie and Rand, 1992).

Returning to early work focused on visible Er^{3+} transitions, McFarlane (1989) at the Hughes Research Laboratories also investigated LiYF_4 (aka YLF) host materials, but at a concentration 5 times higher than that used by the IBM group. He first observed cw output at cryogenic temperatures for the $^4F_{9/2} \rightarrow ^4I_{15/2}$ transition of Er^{3+} at 671 nm when a dielectric-coated laser crystal was pumped at 791 nm. Aided by multiple steps of single-ion nonradiative relaxation during optical pumping, energy-transfer upconversion and a sequential photon absorption process were proposed as joint pumping mechanisms for the $^4F_{9/2}$ upper laser level. However, when additional feedback at 551 nm was provided by an auxiliary mirror mounted external to the sample cryostat, self-Q-switched output on the $^4S_{3/2} \rightarrow ^4I_{15/2}$ green transition occurred simultaneously with 671-nm output. This appears to be the first report of dual-wavelength oscillation in an upconversion laser. The Q-switching behavior for the green output was ultimately attributed to excited-state saturated absorption at 551 nm on the $^4I_{13/2} \rightarrow ^2H_{9/2}$ transition, as had been previously noted (Lenth *et al.*, 1988).

Trivalent thulium also made its first appearance in an upconversion laser in 1989. Using pulsed dye lasers operating at 781 and 649 nm, Nguyen *et al.* (1989) exploited sequential multi-photon absorption to optically pump a $\text{Tm}^{3+}(1\%):\text{YLF}$ crystal held at 75 K and obtained 180 μJ of pulsed deep blue output on the

dopant ion's $^1D_2 \rightarrow ^3F_4$ transition at 450 nm. Remarkably, this same group soon reported room-temperature operation of this device (Nguyen *et al.*, 1990).

By all accounts the most significant accomplishment published in 1989, again by the pioneering IBM group (Tong *et al.*, 1989), was the first demonstration of an all-solid-state visible upconversion laser, the pump source being an AlGaAs diode-laser array operating at 791 nm. Using an YLF laser crystal doped with Er^{3+} at 1 at%, 100 μW of self-Q-switched output was obtained on the familiar 551-nm green transition at a sample temperature of 40 K. As with previous experiments involving YLF host crystals, both cooperative energy transfer and doubly-resonant multi-photon absorption provided upconversion pumping of the upper laser level. Changing over to dye-laser pumping at 791 nm, the same laser crystal exhibited a remarkably low pump-power threshold of 17 mW, a clear benchmark of the rapid advances the field was making in these early years.

But just a few months later a revolutionary approach in upconversion laser design was first applied. Motivated by the development of 1.55- μm erbium-doped silica-fiber lasers and amplifiers, which exhibited remarkably low thresholds and high gains, Allain *et al.* (1990) working at the Centre National d'Études des Telecommunications (CNET) investigated the possibility of upconversion pumping in a fiber geometry. High gain is expected in fiber-based active devices because of the natural confinement of pump power to a very small diameter ($\sim 1\text{--}5\ \mu\text{m}$) over meters of fiber length. Large population densities are therefore maintained over large distances, implying that extraordinary round-trip gains can be created with relatively little pump power, even in three-level laser systems. Moreover, the enormous economic potential of light-wave telecommunications had driven the refinement of single-mode silica optical fiber to the point where the theoretical limits of optical loss could be purchased for just a few dollars per meter. Both high gain and low loss are thus the hallmarks of silica-fiber lasers and amplifiers.

However, owing to the need for energy storage by intermediate states within the upconversion chain, Allain *et al.* recognized that a silica-fiber host with its 1100-cm^{-1} maximum phonon frequency would likely yield nonradiative relaxation rates too high to permit effective upconversion pumping. Indeed, phonon frequencies in the bulk upconversion laser crystals explored up to then all lay below about 600-cm^{-1} . Instead, these workers turned to newly developed fibers based on the low-phonon fluorozirconate glass ZBLAN ($\text{ZrF}_4\text{-BaF}_2\text{-LaF}_3\text{-AlF}_3\text{-NaF}$). Although in 1990 optical losses in doped ZBLAN fiber well exceeded theoretical limits, its $\sim 650\text{-cm}^{-1}$ maximum phonon frequency ensured excited-state lifetimes comparable to those of the bulk crystals already successfully used in upconversion laser experiments.

In their first experiment, the CNET group demonstrated both 455-nm ($^1D_2 \rightarrow ^3F_4$) and 480-nm ($^1G_4 \rightarrow ^3H_6$) blue laser output from a Tm^{3+} -doped ZBLAN-fiber

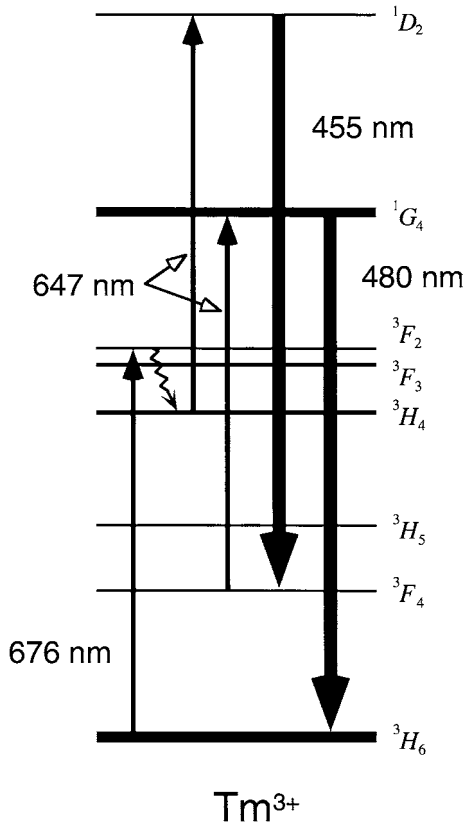


Figure 8.6: Dual-wavelength blue upconversion laser based on sequential two-photon pumping of Tm^{3+} ions in a ZBLAN fiber. Two possible upconversion pathways populate the upper laser levels (Allain *et al.*, 1990).

multi-photon pumped with a cw krypton-ion laser running simultaneously on two red lines at 647 and 676 nm (Figure 8.6). The optical cavity was formed of two flat mirrors placed in contact against the ends of a 170-cm length of fiber; this fiber was then immersed in a liquid nitrogen bath in order to enhance absorption of pump light and reduce nonradiative loss. An oscilloscope image of the optical output wave form showed that when the pump beam was turned on first a short burst of output at 455 nm occurred, followed by a continuous spiking output at 480 nm. Up to 400- μW average power was measured at 480 nm; the laser threshold was just 90 mW of injected pump power into the fiber.

While this level of performance was less than what already had been achieved in the earlier bulk upconversion laser experiments, the CNET group just two months after the submission of their Tm^{3+} results unambiguously proved the merits of the fiber-laser format by demonstrating the world's first cw, room-temperature, visible upconversion laser (Allain *et al.*, 1990a). Based on a 1-m length of ZBLAN fiber

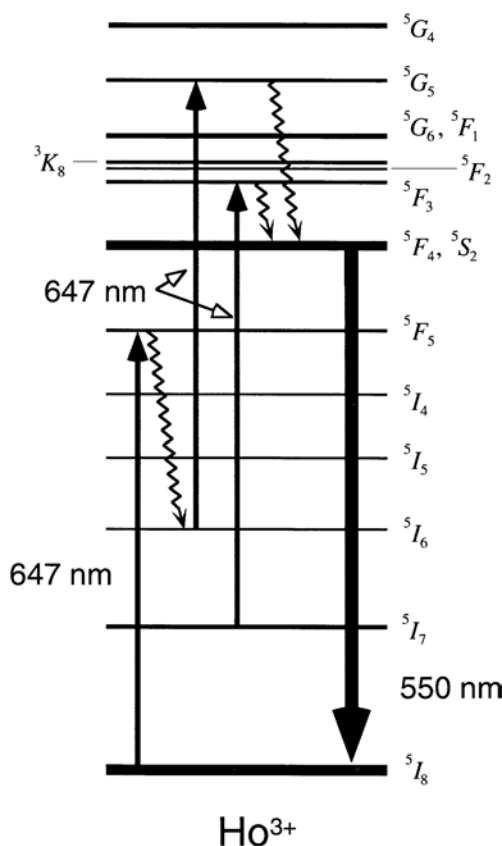


Figure 8.7: Green upconversion laser based on single-wavelength sequential two-photon pumping of Ho^{3+} ions. Two different pathways build population in the upper laser level (Allain *et al.*, 1990a).

doped with 1 wt% Ho^{3+} , a maximum cw output of 10 mW was obtained on the 550-nm $^5S_2 \rightarrow ^5I_8$ transition when the fiber was multi-photon pumped with a Kr^+ laser operating at 647 nm (Figure 8.7). The laser exhibited a substantial 20% slope efficiency with respect to the pump power as coupled into the fiber and was tunable in the green over the (540–553)-nm wavelength band.

An account of a praseodymium-based upconversion laser was likewise first published in early 1990 (Koch *et al.*, 1990). Operating at 644 nm and pumped by a 677-nm cw dye laser, this experiment is most noteworthy for its exploitation of a photon-avalanche pump mechanism, one of the first examples of this pumping scheme in an upconversion laser. The details are depicted in Figure 8.8: The pump laser is tuned to the $\text{Pr}^{3+} \ ^3F_3 \rightarrow \ ^3P_1$ ESA transition, which weakly populates the $^3P_0$ and $^3P_1$ states commensurate with a small population initially in the $^3F_3$ state. Unfortunately, the specifics of the required cross-relaxation

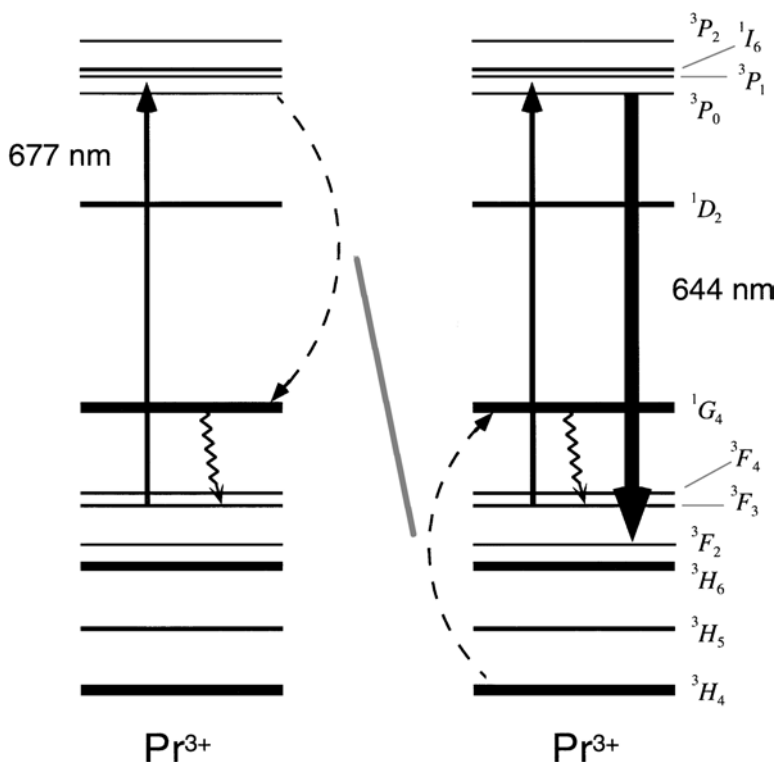


Figure 8.8: Photon-avalanche pump mechanism for a Pr^{3+} -based red upconversion laser (Koch *et al.*, 1990).

mechanisms that help to repopulate the $^3F_3$ state were not known at the time of this experiment; nevertheless, the authors point out the interesting possibility that the $^3P_0$ upper laser level need not be involved. In any case, the $\text{Pr}^{3+}(7\%):\text{LaCl}_3$ laser crystal produced a remarkable 230 mW of output power at 80 K for just 900 mW of pump power.

Aided by the versatile Ti:S laser made commercially available in the late 1980s, another high-power upconversion-laser result was soon reported by McFarlane *et al.* (1990). Working again with what has ultimately emerged as the most popular of bulk upconversion laser materials, namely $\text{Er}^{3+}:\text{LiYF}_4$, the Hughes group demonstrated 430 mW of green output for a 5-mol% sample held at 70 K and pumped with 2.1 W at 797 nm. The output consisted of a 100-kHz train of 100-ns Q-switched pulses as has been observed previously.

Not to be outdone, the IBM group countered with their own results based on high-power Ti:S pumping (Hebert *et al.*, 1990), in this case demonstrating two new laser transitions in $\text{Er}(1\%):\text{YLF}$. These were the blue $^2P_{3/2} \rightarrow ^4I_{11/2}$ transition at 470 nm and the green $^2H_{9/2} \rightarrow ^4I_{13/2}$ transition at 561 nm. In the case of the

blue laser experiment – the first to use an infrared pump source – the most complex upconversion pumping mechanism up to that time was deduced: In order to populate the high-lying $^3P_{3/2}$ state of Er^{3+} , the energy of at least four Er^{3+} ions directly or indirectly pumped to the $^4I_{11/2}$ state must be pooled by multiple energy-transfer steps onto a single ion. Another remarkable result obtained with the blue transition, this time by using a red pump laser tuned to 653 nm, was a 10-mW pump-power threshold measured at a sample temperature of 16 K. As for the green transition, infrared Ti:S pumping and the aforementioned complex population kinetics led to simultaneous green output on the $^2H_{9/2} \rightarrow ^4I_{13/2}$ transition and on the familiar $^4S_{3/2} \rightarrow ^4I_{15/2}$ transition, at least for sample temperatures in the range of 25–40 K. Below 25 K, the new 560-nm transition dominated, while above 40 K the 551-nm output took over.

Hence in just a few years the eventual fortunes of practical blue and green solid-state lasers had been soundly established through successful demonstrations of room-temperature operation, low pump thresholds, good slope efficiencies, and infrared pumping compatible with available semiconductor diode lasers. On the latter point, prospects could only improve as high-power, high-efficiency laser diodes based on new semiconductor materials and new design and manufacturing techniques would in subsequent years dramatically expand the range of pump wavelengths potentially applicable to the upconversion problem. Analytical models of upconversion laser kinetics had also been successfully developed and verified so that equally good prospects existed for rationalizing upconversion laser design. By 1990, the three upconversion pump mechanisms of sequential multi-photon absorption, radiationless energy transfer, and the related photon-avalanche process had each been exploited to demonstrate true upconversion laser output. In the transitional year of 1990, Lenth and Macfarlane (1990) for the first time took stock of all three upconversion pumping mechanisms, as if to place the field's first punctuation mark and allow everyone to take a breath. Numerous reviews (France, 1991, Monerie, 1991, Oomen and Lous, 1992, Piehler, 1993, Macfarlane, 1994, Hanna and Tropper, 1995, Goldner and Pellé, 1996, Hanna, 1996, Scheps, 1996) have since appeared offering updates on the field's rapid progress.

8.2 BULK UPCONVERSION LASERS

We now depart from a strictly chronological account of upconversion laser research and focus attention on the details of particular active ions and laser hosts. To this end, in this section we present the results of experiments on bulk upconversion lasers, the adjective “bulk” here denoting the rough equivalence of the three sample dimensions and the absence of any waveguiding function for either the pump beam or the intracavity oscillating mode. Owing to the narrower absorption and gain line

shapes characteristic of rare-earth-impurity transitions in crystalline materials, such hosts are used exclusively in this class of experiments.

Laser cavities constructed around bulk gain media usually assume one of two configurations. These are depicted in Figure 8.9. The main difference between the two types is whether the cavity mirrors are deposited directly onto curved surfaces ground and polished on the laser crystal (a “monolithic” laser cavity, Figure 8.9(a)) or whether external optics are used to form the cavity with the gain medium (and frequently cryostat windows) sandwiched between them (Figure 8.9(b)). In the latter case, the crystal faces are flat and may either be anti-reflection coated or simply tilted to Brewster’s angle in order to reduce intracavity losses. Sometimes a combination of the two approaches is used, in which case the high-reflectivity end-mirror is deposited directly onto one flat face of the laser crystal, an anti-reflection coating may be applied to the second flat face, and an externally mounted output coupler completes the cavity (Figure 8.9(c)). In all three cases, the high pump intensities dictated by the nonlinear character of upconversion processes often favor short cavity lengths and their commensurate narrow gain regions. Cavity lengths on the order of 1 cm or so are therefore common. Table 8.1 gives a comprehensive summary of bulk upconversion laser experiments performed as of this writing.

8.2.1 Upconversion pumped Er^{3+} infrared lasers

Ironically one of the most heavily investigated upconversion laser systems, namely Er^{3+} in various hosts pumped at $\sim 1.5\ \mu\text{m}$, yields laser output at primarily infrared wavelengths. Perhaps more ironic is that in many of these experiments the output wavelength is *longer* than the input wavelength, thus somewhat stretching the intent of the moniker upconversion laser. The reason behind this irony is that Er^{3+} ions pumped from the ground state to their first excited Stark manifolds exhibit upconversion due exclusively to cooperative energy transfer – no other processes complicate the upconversion kinetics. The low energy of the fundamental optical excitation no doubt places further restrictions on the complexity of possible upconversion pathways. As a result, the system is especially amenable to mathematical modeling, allowing one to interpret and perhaps quantify upconversion effects. For its pedagogical value we discuss in this section such “upconversion-pumped” infrared lasers.

S. A. Pollack and D. B. Chang, along with various colleagues of theirs at the Hughes Aircraft Company have been the leading proponents of this work. Important additional contributions have been made by P. Xie and S. C. Rand, most notably in the realm of continuously pumped laser systems as compared with the exclusively pulsed experiments conducted by the Hughes group.

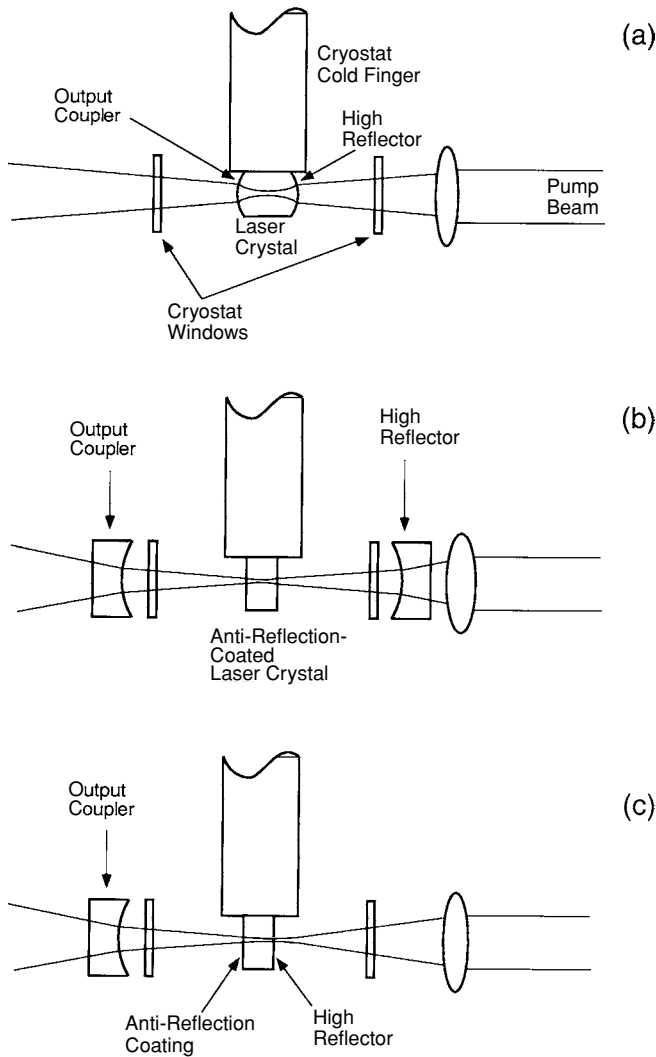


Figure 8.9: Basic laser cavity configurations for upconversion lasers based on doped bulk crystals. (a) A monolithic laser cavity: Dielectric mirrors are deposited directly onto the polished curved surfaces of the laser crystal. The high-reflector coating is chosen to maximize transmission of pump radiation while offering high reflectivity at the output wavelength. (b) Laser cavity with external cavity mirrors. Cavity losses are minimized by depositing anti-reflection coatings onto the polished flat surfaces of the laser crystal and onto the cryostat windows. Alternatively, the laser crystal may be tilted to Brewster's angle, which confers a preferred polarization to the oscillating mode and recommends that the pump-laser polarization be correctly oriented in order to optimize pump efficiency. (c) A hybrid monolithic-external cavity configuration in which the high reflector is deposited directly onto the laser crystal but the output coupler is mounted externally.

Table 8.1. *Bulk upconversion laser experiments*

Ion	Codopant	Host	Pump mechanism	Pump source	Pump wavelength	Operating temperature	Transition, wavelength	Max output power or energy	Reference
Er^{3+}	Yb^{3+}	BaY_2F_8	ET	pulsed xenon flashlamp	~ 960 nm	77 K	$^4F_{9/2} \rightarrow ^4I_{15/2}$	unreported	Johnson and Guggenheim, 1971 Pollack <i>et al.</i> , 1986
Er^{3+}		CaF_2	ET	pulsed Er^{3+} -glass laser	1.54 μm	298 K	$^4I_{11/2} \rightarrow ^4I_{13/2}$	unreported	
Er^{3+}		YAlO_3	MP + ET	cw dye lasers	$792 + 840$ nm	20–77 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$	1 mW (@ 30 K)	Silversmith <i>et al.</i> , 1987
Er^{3+}		LiYF_4	ET	pulsed Er^{3+} -glass laser	1.53 μm	100, 300 K	$^4I_{11/2} \rightarrow ^4I_{13/2}$	unreported	Pollack and Chang, 1988
		YAG	ET			100, 300 K	$^4I_{11/2} \rightarrow ^4I_{13/2}$	unreported	
		CaF_2	ET			100, 300 K	$^4I_{11/2} \rightarrow ^4I_{13/2}$	unreported	
		SrF_2	ET			100, 300 K	$^4I_{11/2} \rightarrow ^4I_{13/2}$	unreported	
Er^{3+}		LiYF_4	ET	cw dye laser	802 nm	10–90 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$	5 mW (@ 50 K)	Lenth <i>et al.</i> , 1988
Er^{3+}		LiYF_4	ET	pulsed Er^{3+} -glass laser	1.53 μm	110 K	$^4S_{3/2} \rightarrow ^4I_{9/2}$	unreported	Pollack <i>et al.</i> , 1989
							$^4S_{3/2} \rightarrow ^4I_{11/2}$	unreported	
							$^4S_{3/2} \rightarrow ^4I_{13/2}$	unreported	
Er^{3+}		LiYF_4	ET + MP	cw dye laser	791 nm	15–60 K	$^4F_{9/2} \rightarrow ^4I_{15/2}$	unreported	McFarlane, 1989
						15–85 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$	2.5 mW (average pulsed output @ 65 K)	
Er^{3+}		LiYF_4	ET + MP	cw AlGaAs laser	791 nm	40 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$	100 μW (average pulsed output @ 40 K)	Tong <i>et al.</i> , 1989
						<90 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$	4.2 mW (average pulsed output @ 40 K)	

Er ³⁺	BaY ₂ F ₈	ET	pulsed Er ³⁺ :glass laser	1.53 μm	100, 300 K	⁴ I _{11/2} → ⁴ I _{13/2} 2.80 μm	unreported	Pollack <i>et al.</i> , 1990
					100 K	⁴ S _{3/2} → ⁴ I _{9/2} 1.73 μm	unreported	
Er ³⁺	LiYF ₄	ET + MP	cw Ti:sapphire laser	797 nm	41 K	² H _{9/2} → ⁴ I _{11/2} 702 nm	40 mW	McFarlane <i>et al.</i> , 1990
					41 K	⁴ F _{9/2} → ⁴ I _{15/2} 668 nm	26 mW	
					41 K	² P _{3/2} → ⁴ F _{9/2} 618 nm	500 μW	
					70 K	⁴ S _{3/2} → ⁴ I _{15/2} 551 nm	430 mW (average pulsed output)	
Er ³⁺	CaF ₂	ET	cw color-center laser	1.51 μm	298 K	⁴ I _{11/2} → ⁴ I _{13/2} 2.75 μm	9 mW	Xie and Rand, 1990
Er ³⁺	LiYF ₄	ET	cw Ti:sapphire laser	797, 969 nm	10–40 K	² H _{9/2} → ⁴ I _{13/2} 561 nm	3.5 mW (@ 10 K)	Hebert <i>et al.</i> , 1990
			cw dye laser	653 nm	10–35 K	² P _{3/2} → ⁴ I _{11/2} 470 nm	6 mW (@ 16 K)	
			cw Ti:sapphire laser	797, 969 nm	10 K	² P _{3/2} → ⁴ I _{11/2} 470 nm	2 mW (969 nm pump)	
Er ³⁺	CaF ₂	ET	cw color-center laser	1.51 μm	77 K	⁴ S _{3/2} → ⁴ I _{13/2} 855 nm	64 mW	Xie and Rand, 1990a
Er ³⁺	LiYF ₄	ET	cw Ti:sapphire laser	791 nm	49 K	⁴ S _{3/2} → ⁴ I _{15/2} 551 nm	500 mW	McFarlane, 1991
Er ³⁺	YAG		pulsed Er ³⁺ :glass laser	1.53 μm	77, 300 K	⁴ I _{11/2} → ⁴ I _{13/2} 2.9 μm	unreported	Pollack <i>et al.</i> , 1991
	YSGG				77, 300 K	⁴ I _{11/2} → ⁴ I _{13/2} 2.8 μm	unreported	
Er ³⁺	LiYF ₄	ET	cw color-center laser	1.55 μm	77–200 K	⁴ S _{3/2} → ⁴ I _{13/2} 855 nm	120 mW (@ 77 K)	Xie and Rand, 1990
Er ³⁺	LiYF ₄	ET	cw color-center laser	1.55 μm	9–95 K	⁴ S _{3/2} → ⁴ I _{15/2} 544 nm	32 mW (@ 9 K)	Xie and Rand, 1990a
Er ³⁺	LiYF ₄	ET	cw color-center laser	1.55 μm	9 K	⁴ S _{3/2} → ⁴ I _{15/2} 544 nm	600 nJ Q-switched	Xie and Rand, 1992
					9–15 K	⁴ S _{3/2} → ⁴ I _{15/2} 544 nm	2 mW cw mode locked (@ 9 K)	

Table 8.1. (*cont.*)

Ion	Codopant	Host	Pump mechanism	Pump source	Pump wavelength	Operating temperature	Transition, wavelength	Max output power or energy	Reference
Er^{3+}		LiYF_4	ET	cw color-center laser	1.55 μm	2–35 K	$^2H_{9/2} \rightarrow ^4I_{13/2}$ 561 nm $^4S_{3/2} \rightarrow ^4I_{15/2}$ 551 nm $^2P_{3/2} \rightarrow ^4I_{11/2}$ 470 nm $^4S_{3/2} \rightarrow ^4I_{15/2}$ 551 nm $^2P_{3/2} \rightarrow ^4I_{11/2}$ 470 nm	12 mW peak chopped cw output @ 25 K 10 mW average pulsed output @ 35 K 630 μW peak chopped cw output @ 20 K unreported	Macfarlane <i>et al.</i> , 1992
		BaY_2F_8	PA, ET	cw Ti:sapphire laser	779–808 nm	20 K			McFarlane, 1992, 1994
			MP + ET					unreported	
	Er^{3+}	LiYF_4	ET	cw AlGaAs laser	797 nm	48 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 551 nm	100 mW	Stephens and McFarlane, 1993
	Er^{3+}	LiYF_4	MP + ET	pulsed Ti:sapphire laser + cw Kr^{+} laser	810 + 647 nm	298 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 551 nm $^4S_{3/2} \rightarrow ^4I_{15/2}$ 562 nm $^4S_{3/2} \rightarrow ^4I_{15/2}$ 561 nm $^2H_{9/2} \rightarrow ^4I_{11/2}$ 702 nm $^4S_{3/2} \rightarrow ^4I_{15/2}$ 551 nm $^4S_{3/2} \rightarrow ^4I_{15/2}$ 550 nm	0.95 mJ 140 μJ unreported 380 μW 40 mW 121 mW (@ 34 K)	Brede <i>et al.</i> , 1993, Danger <i>et al.</i> , 1994
Er^{3+}		YAG						unreported	
Er^{3+}		LiYF_4	ET	cw color-center laser	1.55 μm	10 K	$^2H_{9/2} \rightarrow ^4I_{11/2}$		Xie and Rand, 1993
Er^{3+}		LiYF_4	MP	cw Ti:sapphire laser	810 nm	298 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$		Heine <i>et al.</i> , 1994
Er^{3+}		YAlO_3	ET	cw Ti:sapphire laser	807 nm	7–63 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$		Scheps, 1994
Er^{3+}			PA		791 nm	7 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 551 nm	75 mW 37 mW	Möbert <i>et al.</i> , 1997
	Yb^{3+}	LiYF_4	ET(+ESA)	cw Ti:sapphire laser	955–974 nm	298 K			
Ho^{3+}		BaY_2F_8	ET	pulsed xenon flash-lamp	~960 nm	77 K	$^5S_2 \rightarrow ^5I_8$ 552 nm $^3P_0 \rightarrow ^3F_2$ 644 nm	unreported 230 mW (@ 80 K)	Johnson and Guggenheim, 1971 Koch <i>et al.</i> , 1990
Pr^{3+}		LaCl_3	PA	cw dye laser	677 nm	80–210 K			

Pt ³⁺	LiYF ₄	MP	cw Yb:YAG + cw Ti:sapphire laser	1030 + 867 nm	298 K	$^3P_0 \rightarrow ^3F_{2,3}$ 640 nm $^3P_0 \rightarrow ^3H_6$ 607 nm $^1D_2 \rightarrow ^3F_4$ 450 nm $^1G_4 \rightarrow ^3F_4$ 649 nm $^1D_2 \rightarrow ^3F_4$ 450 nm $^1G_4 \rightarrow ^3H_6$ 483 nm	2.2 mW 3.5 mW 180 μ J unreported 10 mW average pulsed output @ 15 K 30 mW average pulsed output @ 26 K 70 μ W average pulsed output unreported 365 μ J unreported unreported unreported unreported unreported unreported unreported unreported	Binun <i>et al.</i> , 1996 Nguyen <i>et al.</i> , 1989 Antipenko <i>et al.</i> , 1990 Hebert <i>et al.</i> , 1992 Scott <i>et al.</i> , 1993 Knipfer <i>et al.</i> , 1994 Thrash and Johnson, 1994
Tm ³⁺	LiYF ₄	MP	pulsed dye laser	781 + 649 nm	75 K	$^1D_2 \rightarrow ^3F_4$ 450 nm		
Tm ³⁺	Yb ³⁺ BaYb ₂ F ₈	ET	pulsed Nd laser	1.05 μ m	298 K	$^1G_4 \rightarrow ^3F_4$ 649 nm $^1D_2 \rightarrow ^3F_4$ 450 nm		
Tm ³⁺	LiYF ₄	MP	cw Ti:sapphire + dye lasers	784 + 648 nm	<160 K	$^1D_2 \rightarrow ^3F_4$ 450 nm		
		PA	cw dye laser	648 nm		$^1G_4 \rightarrow ^3H_6$ 483 nm		
Tm ³⁺	YAG	MP + ET	cw Ti:sapphire + dye lasers	785 + 638 nm	10–30 K	$^1G_4 \rightarrow ^3H_6$ 486 nm $^1D_2 \rightarrow ^3F_4$ 453 nm $^1D_2 \rightarrow ^3F_4$ 453 nm $^1D_2 \rightarrow ^3F_4$ 453 nm $^1G_4 \rightarrow ^3H_5$ 453 nm		
Tm ³⁺	LiYF ₄	MP	pulsed Ti:sapphire + dye lasers	780 + 650 nm	300 K	$^1D_2 \rightarrow ^3F_4$ 453 nm $^1D_2 \rightarrow ^3F_4$ 453 nm $^1G_4 \rightarrow ^3F_4$ 799 nm		
Tm ³⁺	LiYF ₄	MP	pulsed Ti:sapphire + dye lasers	780 + 650 nm	300 K	$^1D_2 \rightarrow ^3F_4$ 453 nm $^1D_2 \rightarrow ^3F_4$ 453 nm $^1G_4 \rightarrow ^3F_4$ 799 nm		
Tm ³⁺	LiGdF ₄	MP	pulsed Ti:sapphire + dye lasers	780 + 650nm	300 K	$^1D_2 \rightarrow ^3F_4$ 453 nm $^1D_2 \rightarrow ^3F_4$ 453 nm $^1G_4 \rightarrow ^3F_4$ 799 nm		
Tm ³⁺	Yb ³⁺ BaY ₂ F ₈	ET	cw Ti:sapphire laser	960 nm	298 K	$^1G_4 \rightarrow ^3H_5$ 453 nm $^1D_2 \rightarrow ^3F_4$ 453 nm $^1G_4 \rightarrow ^3F_4$ 799 nm 649 nm $^1D_2 \rightarrow ^3H_5$ 512 nm $^1G_4 \rightarrow ^3H_6$ 482 nm $^1D_2 \rightarrow ^3F_4$ 456 nm $^1G_4 \rightarrow ^3F_4$ 649 nm $^1D_2 \rightarrow ^3H_5$ 512 nm $^1D_2 \rightarrow ^3F_4$ 456 nm $^4D_{3/2} \rightarrow ^4I_{1/2}$ 380 nm		
			cw diode laser	960 nm				
Nd ³⁺	LaF ₃	MP	cw dye laser	788 + 591 nm, 578 nm	20–90 K	$^4D_{3/2} \rightarrow ^4I_{1/2}$ 380 nm	12 mW (@ 20 K)	Macfarlane <i>et al.</i> , 1988

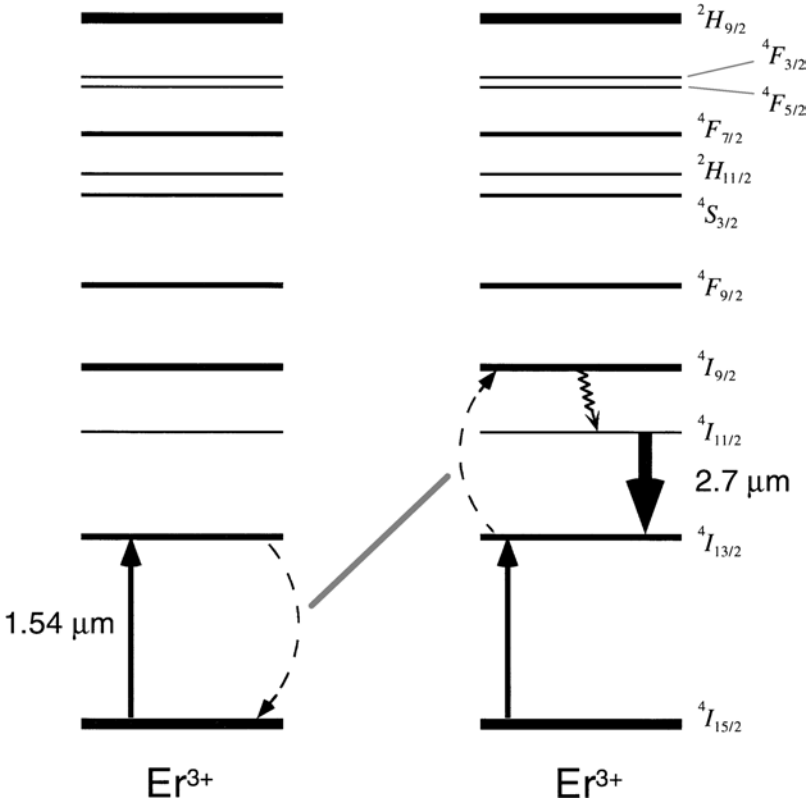
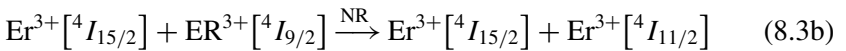
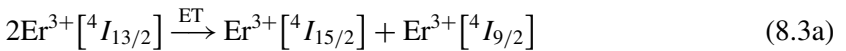


Figure 8.10: Energy-transfer upconversion pumping of Er^{3+} . Optical excitation at $1.54 \mu\text{m}$ yields long-wavelength infrared output originating in a high-lying state (Pollack *et al.*, 1986).

The first step in all experiments is optical pumping on the $4I_{15/2} \rightarrow 4I_{13/2}$ ground-state transition, as shown in Figure 8.10. In their seminal paper on this topic (Pollack *et al.*, 1986) the Hughes group used a pulsed Er:glass laser operating at $1.54 \mu\text{m}$ to drive this transition in CaF_2 single crystals doped at the level of 5–10 at%. Upconversion pumping then resulted from the following sequence of steps (see Figure 8.10):



where the abbreviations ET and NR denote radiationless energy transfer and non-radiative relaxation, respectively. A population inversion thus arose between the $4I_{11/2}$ state and the laser-pumped $4I_{13/2}$ state; placing the pumped crystal between separate curved cavity mirrors (Figure 8.9(b)) yielded pulsed laser output at room

temperature on the $2.8\text{-}\mu\text{m } ^4I_{11/2} \rightarrow ^4I_{13/2}$ transition. In order to minimize the pump threshold energy, the output coupler reflectivity in this experiment was 99.5%. Upon comparison of the relative timing of the $\sim 10\text{-ms}$ pump pulse and the output pulse, the latter was found to lag the leading edge of the pump by several milliseconds. This delay decreased with increasing pump-pulse energy, clearly implying that an energy-transfer mechanism was working behind the scenes to create the inversion.

Note in this experiment that the optically-pumped state is the same as the lower laser level. Denoting the population densities of the $^4I_{15/2}$, $^4I_{13/2}$, $^4I_{11/2}$, and $^4I_{9/2}$ states by the symbols N_0 , N_1 , N_2 , and N_3 , respectively, the Hughes group wrote down a set of rate equations to describe the upconversion kinetics:

$$\frac{dN_1}{dt} = -\alpha N_1^2 - \beta N_1 + B_{01}I_{01}(N_0 - N_1) - B_{12}I_{12}(N_1 - N_2) \quad (8.4a)$$

$$\frac{dN_2}{dt} = \alpha' N_3 - \alpha'' N_2 + B_{12}I_{12}(N_1 - N_2) \quad (8.4b)$$

$$\frac{dN_3}{dt} = \frac{\alpha N_1^2}{2} - \alpha_{TOT} N_3 + B_{13}I_{13}(N_1 - N_3) \quad (8.4c)$$

where α is the rate constant for upconversion as mediated by energy transfer, $\beta \equiv 1/\tau_1$ is the total rate of spontaneous decay of the $^4I_{13/2}$ state, and the $B_{ij}I_{ij}(N_i - N_j)$ terms denote the transition rates due to the net stimulated absorption of pump radiation of intensity I_{ij} at the relevant transition frequencies. The B_{ij} are just the Einstein B coefficients divided by the speed of light. In Equation (8.4b), α' denotes the rate at which the population density of the $^4I_{11/2}$ increases because of the spontaneous decay of the $^4I_{9/2}$ state, while α'' denotes the rate of total spontaneous decay of the $^4I_{11/2}$ state. Finally in Equation (8.4c), α_{TOT} is the total rate of spontaneous decay of the $^4I_{9/2}$ state.

In order to proceed with their analysis, the Hughes group assumed that: (1) the pump pulse was a time-domain δ -function and thus only N_1 would change in the presence of pump radiation; and (2) α_{TOT} was large owing to the expected large value of α'' resulting from nonradiative relaxation. In this case Equation (8.4c) reduces to

$$N_3 \approx \frac{\alpha}{\alpha_{TOT}} \frac{N_1^2}{2} \quad (8.5)$$

Under these approximations, and for a pump pulse width of τ , a population inversion would evolve in the system for times t lying in the range

$$\frac{\exp(2\alpha_{TOT}/\alpha')}{\alpha B_{01} N_0 I \tau} < t < \beta^{-1} \quad (8.6)$$

or in other words inversion would occur for a minimum pump fluence of

$$(I\tau)_{\min} = \frac{\beta \exp(2\alpha_{\text{TOT}}/\alpha')}{\alpha B_{01} N_0} \quad (8.7)$$

This last expression thus emphasizes the benefit of a large upconversion coefficient and a large concentration of active ions for achieving a low pump threshold. The Hughes group thus used the highest Er^{3+} concentration possible consistent with good optical quality for the laser crystal.

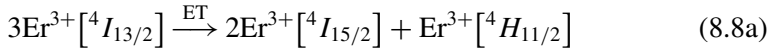
A last point worth making about this upconversion system is that the spontaneous lifetime of the terminal state was approximately twice that of the initial state. Under normal circumstances, such a situation makes the creation of a population inversion more difficult. Even when an inversion initially exists, the onset of oscillation will quickly build a stored population in the lower laser level and destroy the inversion, a circumstance called self-termination or self-saturation. That pulsed laser oscillation on the $2.7\text{-}\mu\text{m } ^4I_{11/2} \rightarrow ^4I_{13/2}$ transition had been nevertheless observed in 1967 was explained at the time by the rapid pumping of the upper laser level combined with the multi-level structure of the $^4I_{11/2}$ and $^4I_{13/2}$ Stark manifolds. The latter features were hypothesized to explain a red shifting of the laser output spectrum as a result of successive self-termination of short wavelength transitions to low-lying members of the terminal manifold. In the Hughes experiments, however, no such red shifting was observed, suggesting that upconversion was effective in depleting the lower laser level and that therefore cw oscillation might be possible on the $^4I_{11/2} \rightarrow ^4I_{13/2}$ transition.

Pollack and Chang went on to investigate pulsed pumping of the $\text{Er}^{3+} ^4I_{13/2}$ state in other host materials (Pollack and Chang, 1988, Pollack and Chang, 1990, Pollack *et al.*, 1990, 1991). In particular, efforts were made to measure the upconversion coefficient α for a variety of oxide and fluoride crystals. However, in a careful study of upconversion fluorescence waveforms, Pollack and Chang (1990) concluded that only in BaY_2F_8 did a simple model analogous to Equations (8.4) successfully explain the data – in this case, an upconversion coefficient of $\alpha = 3.7 \times 10^{-17} \text{ cm}^3/\text{s}$ was determined. Other materials yielded data that could be only approximately interpreted in terms of simple two-ion upconversion. This is because researchers ignored the complicating effects of three-ion cooperative summing and subsequent cross-relaxation of the resulting higher-lying excitations. Nevertheless, important conclusions regarding materials properties could be at least qualitatively obtained.

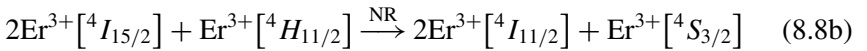
For example, upconversion coefficients for CaF_2 and SrF_2 were found to fall in the region of $10^{-15} \text{ cm}^3/\text{s}$, about two orders of magnitude larger than for BaY_2F_8 and indeed larger than all the other materials that Pollack and Chang (1990) investigated. This large difference was attributed to the need for charge compensation

in the CaF_2 and SrF_2 crystals when the triply-charged Er^{3+} ion substituted for the doubly-charged alkaline-earth cations. The result was that the rare-earth ion in these hosts occupied sites of varying crystal field symmetry – cubic, hexagonal, trigonal, etc. – and thus spectroscopic bands were broader and smoother owing to the mixture of crystal-field splittings in the different sites. The spectral overlap factors in the Förster–Dexter theory (Chapter 7) are therefore correspondingly larger than would be obtained for a single-site symmetry. On the other hand, such broadening reduced the stimulated-emission cross-section for the laser transition, in the process largely neutralizing the advantage of the increased rate of energy transfer. Hence pump thresholds (at room temperature) for Er^{3+} (5%)-doped CaF_2 , SrF_2 , and LiYF_4 laser rods were found in one study (Pollack and Chang, 1988) to all fall near about 50 mJ. By contrast, an Er^{3+} (33%):YAG laser rod in this same study yielded a pump threshold of 450 mJ, a result of the larger phonon frequencies and thus more rapid non-radiative relaxation in this material. An even higher pump threshold was found for Er^{3+} (33%): YAlO_3 (Pollack and Chang, 1990), a material that combined both poor upconversion efficiency and a short upper-level lifetime. Not all oxides, however, proved to be poor host materials for upconversion pumping of the $\text{Er}^{3+} {}^4I_{11/2}$ state. Because yttrium scandium gadolinium garnet ($\text{Y}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$, or YSGG) exhibits both a large upper-state lifetime and good upconversion efficiency, a 15-mJ pump threshold was measured for this material at room temperature (Pollack *et al.*, 1991).

As implied earlier, the two-ion upconversion step Equation (8.3a) is not the only energy-transfer phenomenon at work in these pulsed pumping experiments. The three-ion process



which is followed by the rapid nonradiative relaxation step



has led to the demonstration of upconversion-pumped lasers operating on the ${}^4S_{3/2} \rightarrow {}^4I_J$ transitions (for $J = 9/2, 11/2, 13/2$) at 1.73 μm , 1.23 μm , and 0.85 μm , respectively. For sample temperatures of ~ 100 K, both LiYF_4 (Pollack *et al.*, 1989) and BaY_2F_8 (Pollack *et al.*, 1990) host crystals were investigated, the former material exhibiting true upconversion output (Figure 8.11). The cooperative three-ion mechanism Equation (8.8a) was favored over other possible upconversion pathways, including sequential energy transfers onto a single Er^{3+} ion, owing to the rapid rise of fluorescence originating in the ${}^4S_{3/2}$ state.

While the Hughes group cornered the market on 1.5- μm pulsed upconversion-pumped erbium-based materials, Xie and Rand in a series of four short papers investigated cw infrared lasers upconversion pumped at 1.5 μm . In the first of these

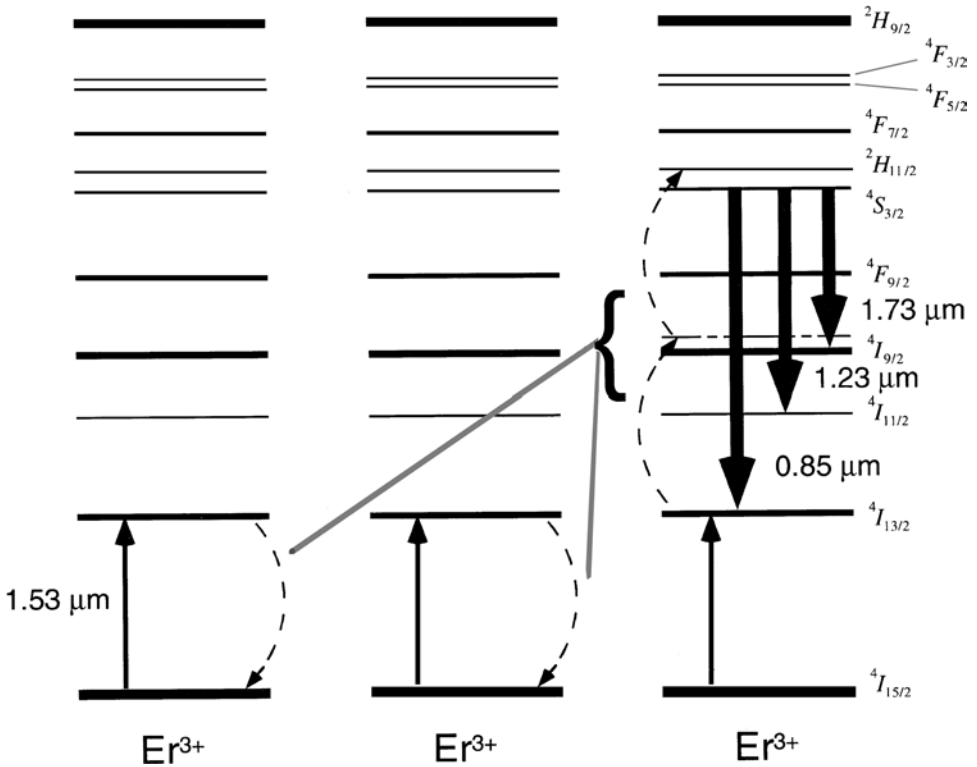


Figure 8.11: Cooperative radiationless energy transfer in an infrared-pumped Er^{3+} triplet. The mechanism is to be distinguished from sequential energy-transfer events, hence the use of the dashed “virtual state” shown in the figure. True upconversion output is obtained in the near infrared (Pollack *et al.*, 1990).

(Xie and Rand, 1990), the authors sought to definitively answer the question whether continuous wave output could be obtained on the otherwise self-terminating $2.7\text{-}\mu\text{m}$ $^4I_{11/2} \rightarrow ^4I_{13/2}$ transition when the lower laser level is continuously pumped. In other words, while the natural lifetime of the lower laser level was ~ 2 times larger than that of the upper laser level, could cooperative energy transfer reduce the effective lifetime of the lower laser level to the point where steady-state inversion and oscillation could be maintained? To answer this question, Xie and Rand wrote down rate equations analogous to Equations (8.4) – but ones which also included stimulated emission terms for the laser transition – then set the time derivatives to zero, and thereby derived an inequality that the upconversion coefficient would have to satisfy in order for continuous oscillation to be sustained. However, these researchers recognized that cw oscillation may be unstable in this nonlinear system and went on to perform a stability analysis that proved that chaotic output or periodic pulsing would not occur provided the lifetime of the $^4I_{9/2}$ state were sufficiently

small. This was indeed the case for the $\text{Er}^{3+}(5\%):\text{CaF}_2$ sample used in this study and cw oscillation in a purely upconversion-pumped self-terminating system was demonstrated for the first time. In particular, a cw NaCl color-center laser tuned to $1.51\ \mu\text{m}$ was used as a pump source; the laser cavity consisted of a 3-mm-thick disk with appropriate dielectric coatings deposited directly on the sample. At room temperature, output coupling of 2% yielded a maximum 9 mW of output at $2.75\ \mu\text{m}$ for 200 mW of pump power absorbed by the sample; the overall efficiency was thus 4.5%.

Xie and Rand also investigated the possibility of continuous output on the $^4S_{3/2} \rightarrow ^4I_{13/2}$ transition at 855 nm following three-fold cooperative upconversion (Equation (8.8a)) in the same material system (Xie and Rand, 1990a). With a sample prepared in a manner similar to their pair-pumped laser, except with mirror coatings reflecting at $0.85\ \mu\text{m}$ and output coupling of 0.5%, 64 mW of output at 855 nm was measured for a pump power of 235 mW at a sample temperature of 77 K. With an overall efficiency of 26% and a slope efficiency of 30%, this laser exhibited higher efficiency and a lower threshold (10 mW) than the pair laser, albeit this performance having been attained at much lower sample temperature. Switching to a YLF host doped at 5 mol% and employing a much improved three-mirror cavity design that compensated for astigmatism in the optical path (Figure 8.12), 120 mW of output power was produced for a pump power of 640 mW

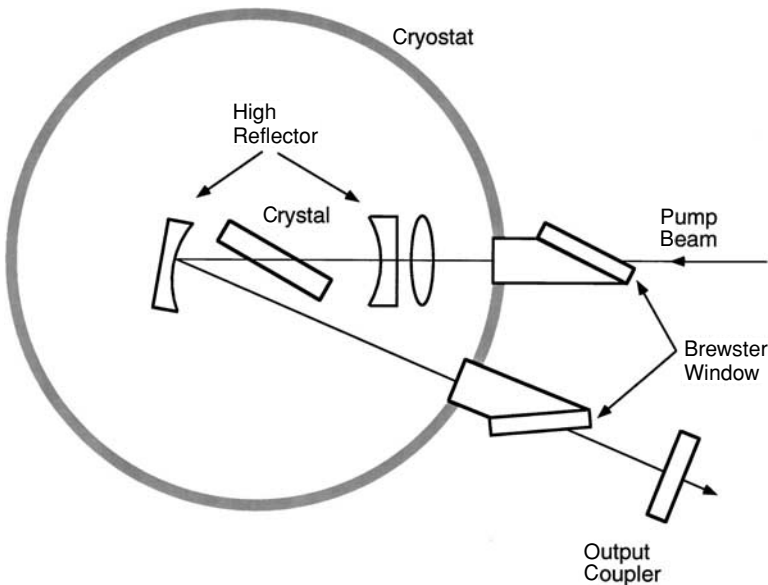
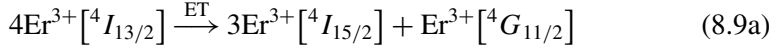


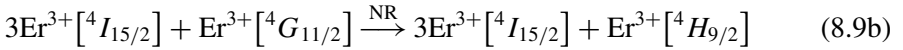
Figure 8.12: Laser cavity design used by Xie and Rand (1990) that compensates for astigmatism introduced by the Brewster-mounted laser crystal and the cryostat's input/output windows.

at a sample temperature of 77 K. The threshold pump was just 5 mW with the optimum output coupling of 23%; oscillation was observed up to a temperature of 200 K.

Taking multi-atom cooperative upconversion in YLF crystals one more step, Xie and Rand (1993) demonstrated cw output on the $^2H_{9/2} \rightarrow ^4I_{11/2}$ Er^{3+} transition at 702 nm, which resulted from upper-level pumping due to the four-ion process



followed by



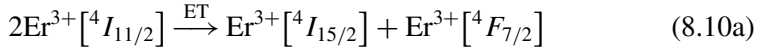
Based on a measurement of the upconversion excitation spectrum, which showed a one-to-one correspondence with the $^4I_{15/2} \rightarrow ^4I_{13/2}$ absorption spectrum, this “quartet” mechanism was favored over a time-ordered sequence of energy-transfer steps or multi-photon absorption. While only 380 μW of output was observed, Xie and Rand recognized that multi-atom cooperative upconversion processes could be intrinsically more efficient than sequential multi-photon absorption in populating high-lying states. Such is the case when subsequent upconversion emission terminates on any energy level that participates in the upconversion. In the case of Er^{3+} and four-fold upconversion, for example, a terminal laser level of $^4I_{13/2}$ (555 nm) would exploit this “photon-recycling” enhancement.

8.2.2 Er^{3+} visible upconversion lasers

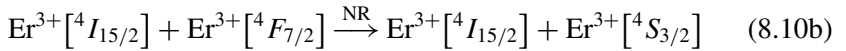
8.2.2.1 $\text{Er}^{3+}:\text{YAlO}_3$; $^4S_{3/2} \rightarrow ^4I_{15/2}$ transition at 550 nm

In the first report of cw green upconversion output based on the $\text{Er}^{3+} \ ^4S_{3/2} \rightarrow ^4I_{15/2}$ transition, Silversmith *et al.* (1987) used a monolithic cavity design in which dielectric mirrors were deposited directly onto the faces of a 3-mm-long YAlO_3 (YALO) crystal doped at 1 mol% Er^{3+} (Figure 8.9(a)). A stable optical resonator was created in this sample by polishing both faces to a radius of 2 cm; the output coupling was 1%. Although a radiationless energy-transfer mechanism was sure to have played a role in upconversion pumping of this system (green fluorescence was readily observed when a single pump laser tuned to 792.1 nm illuminated the sample – plus no doubly resonant ESA transition existed), this mechanism alone did not yield gain above the oscillation threshold. Addition of a second laser, tuned to the $^4I_{11/2} \rightarrow ^4F_{5/2}$ transition at 839.8 (Figure 8.5; see also Lenth *et al.* (1988)), created a two-photon upconversion pathway to the $^4S_{3/2}$ upper laser level. True cw output of 1 mW in the green was observed at a sample temperature of 30 K for 200 mW of pump power from each infrared laser.

Little further work has been performed with YALO host crystals since this seminal work. Seven years after the IBM experiments, however, R. Scheps conducted a comprehensive study of green upconversion oscillation in a single-wavelength-pumped 10-mm Er:YALO rod doped at 1.5 at% (Scheps, 1994). As a pump source, a Ti:S laser was tuned to coincide with various Stark transitions originating in the lowest-lying Kramers doublet (out of a total of eight) belonging to the ground-state manifold and terminating in one of five such levels belonging to the $^4I_{9/2}$ manifold. In order to distinguish one transition from another, the various possibilities were designated $^4I_{15/2}(1) \rightarrow ^4I_{9/2}(n)$, with $n = 1, 2, 3, 4$, or 5 ; the corresponding wavelengths ranged from 806.9 nm to 785.4, respectively. Thus the promise discovered in the earlier IBM work of upconversion pumping of the $^4S_{3/2}$ state based exclusively on energy transfer, specifically



followed by



was first realized in these much later experiments. Interestingly, quite low threshold pump powers were observed, being as low as 80 mW for a sample temperature of 34 K and as low as 130 mW at a temperature of 7 K. These results were obtained despite the high output coupling of 10%, which was provided by a separate dielectric mirror mounted outside the cryostat (the high reflectivity end reflector was deposited directly onto the crystal's rear surface; see Figure 8.9(c)). A maximum output power of 121 mW was measured for an absorbed pump power of 918 mW – an overall efficiency of 13%. Such dramatically improved results as compared with those of the IBM group no doubt resulted from the laser crystal's higher optical quality, with some improvement perhaps deriving from the higher impurity concentration. Scheps also discovered several pump wavelengths in the vicinity of 800 nm that gave rise to photon-avalanche pumping of the upper laser level. Although none of these yielded higher output power than any of the $^4I_{15/2}(1) \rightarrow ^4I_{9/2}(n)$ pump wavelengths, as measured with respect to the absorbed pump-laser power, overall output efficiencies as high as 16% were found.

8.2.2.2 $\text{Er}^{3+}:\text{YLF}; ^4S_{3/2} \rightarrow ^4I_{15/2}$ transition at 551 nm

As the previous discussion tacitly suggests, the greatest advantage of upconversion pumping by energy transfer in comparison to sequential multi-photon absorption is that typically only a single pump laser is required. Unlike the initial results on YALO host crystals, YLF doped with Er^{3+} ions readily exhibited the advantage

just named and quickly became the most studied of bulk upconversion laser materials. Evidently the energy-transfer mechanism at work in YLF crystals is of much greater efficiency than that in YALO crystals, although the exact reasons behind this difference have not been specifically addressed. In any case, the temporary continuation of oscillation on the $^4S_{3/2} \rightarrow ^4I_{15/2}$ transition after abrupt shut-off of a pump laser tuned to the $^4I_{15/2} \rightarrow ^4I_{9/2}$ transition was definitive evidence that upconversion by energy transfer was operative in the Er:YLF system (Lenth *et al.*, 1988). Indeed, using the same sample geometry and Er^{3+} concentration as in the previous year's YALO work, Lenth *et al.* obtained 5 mW of green output at a sample temperature of 50 K with just 240 mW of pump power at 802 nm. The green output, however, consisted of an unexplained quasi-continuous train of 80–150-ns pulses with a repetition rate of 40–250 kHz.

However, such a simple interpretation of the pumping mechanisms in YLF crystals proved short-lived. Subsequent work by R. A. McFarlane at the Hughes Research Laboratories soon demonstrated that upconversion pumping of Er:YLF at ~ 800 nm resulted from more than just energy transfer according to Equations (8.10) (McFarlane, 1989). In particular, McFarlane found evidence for the sequential two-photon mechanism

$$\text{Er}^{3+}[^4I_{15/2}] + \hbar\omega_{\text{pump}} \rightarrow \text{Er}^{3+}[^4I_{9/2}] \quad (8.11a)$$

$$\text{Er}^{3+}[^4I_{9/2}] \xrightarrow{\text{NR or R}} \text{Er}^{3+}[^4I_{13/2}] \quad (8.11b)$$

$$\text{Er}^{3+}[^4I_{13/2}] + \hbar\omega_{\text{pump}} \rightarrow \text{Er}^{3+}[^2H_{11/2}] \quad (8.11c)$$

$$\text{Er}^{3+}[^2H_{11/2}] \xrightarrow{\text{NR}} \text{Er}^{3+}[^4S_{3/2}] \quad (8.11d)$$

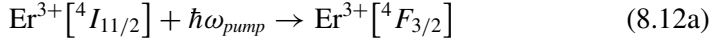
or

$$\text{Er}^{3+}[^2H_{11/2}] \xrightarrow{\text{NR or R}} \text{Er}^{3+}[^4F_{9/2}] \quad (8.11e)$$

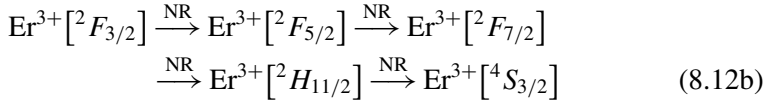
where the symbol R denotes spontaneous emission. The very last step just listed gave rise to inversion on the $^4F_{9/2} \rightarrow ^4I_{15/2}$ transition and strictly cw oscillation was observed at 671 nm in a 5%-doped monolithic laser cavity at temperatures up to 60 K. When an external mirror providing feedback at 551 nm was added to the optical system, pulsed output similar to that obtained by the IBM group was observed. This Q-switched behavior was explained by McFarlane as resulting from excited-state absorption of the 551-nm upconversion output on the $\text{Er}^{3+} ^4I_{13/2} \rightarrow ^2H_{9/2}$ transition. Optical saturation of this transition favors accumulation of the green output into a high-power saturating pulse. The process repeats as sufficient population is rebuilt in the $^4S_{3/2}$ state. In turn, the Q-switched pulse train imparts a time dependence to the $^4I_{13/2}$ population and hence a time dependence to the 671-nm

output as a consequence of the multi-photon absorption pathway Equations (8.11). Thus the upconversion laser output in this case was especially revealing of the pumping mechanisms at play.

In 1993, yet a third upconversion pumping mechanism in Er:YLF was identified by a German group working at the University of Hamburg (Brede *et al.*, 1993). Using a pulsed Ti:S laser and a 1%-doped crystal at room temperature, the ESA step



followed by the nonradiative cascade



was found to be the most important pump process in this lightly doped sample. The principal evidence for this conclusion was the observation that maximum green laser output (at room temperature!) was obtained at a pump wavelength of 810 nm, a wavelength not directly resonant with the $^4I_{15/2} \rightarrow ^4I_{9/2}$ ground-state transition. The reduced Er^{3+} concentration relative to that used in previous work clearly helped to inhibit the familiar energy-transfer process Equation (8.10a); that the 810 nm pump wavelength was so effective in creating a population inversion can be readily attributed to the thermal excitation of previously unoccupied Stark levels in both the ground and excited manifolds. More opportunities for doubly-resonant two-photon transitions thus exist at higher temperatures. Additional time-resolved spectroscopic studies further supported the importance of the ESA step Equation (8.12a) (Danger *et al.*, 1994). In Danger *et al.* (1994), a sizable 13% slope efficiency with respect to the absorbed pump laser power was measured for an optical cavity optimized for low pump threshold. The only real disadvantage in this experiment was that the low Er^{3+} concentration yielded only 15% absorption of the pump energy.

Taking advantage of this new upconversion mechanism, just a year later the Hamburg group demonstrated cw oscillation at 551 nm in a room-temperature $\text{Er}^{3+}(1\%):\text{YLF}$ laser crystal (Heine *et al.*, 1994). In order to improve absorption of the 810-nm pump radiation, the output coupler was designed to reflect 97% of the incident pump power – which entered the crystal through the end reflector – back into the gain region. With output coupling of 6.6%, 40 mW of true cw output was obtained for a pump power of 2.8 W derived from a cw Ti:S laser; the overall efficiency was 10% measured with respect to the absorbed laser power. That self-Q-switching was not observed in this experiment was likely

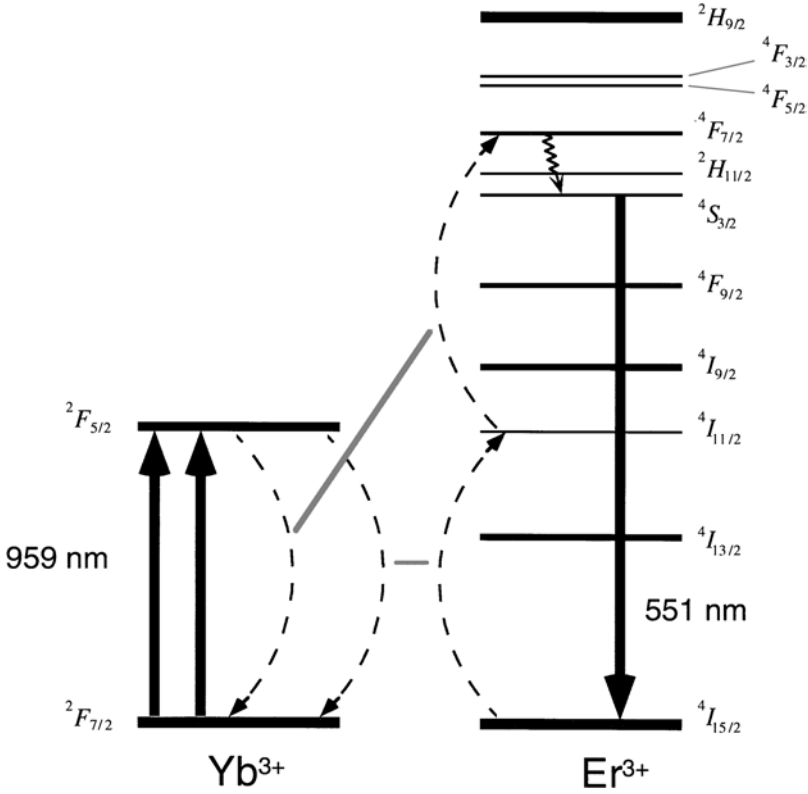
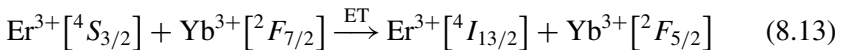


Figure 8.13: Green upconversion laser based on sequential energy-transfer upconversion pumping of Er^{3+} by infrared-pumped Yb^{3+} ions (Möbert *et al.*, 1997).

a result of the low erbium concentration and perhaps thermal broadening and population effects that one expects to reduce absorption cross-sections at high temperatures.

The work of this group culminated in 1997 (Möbert *et al.*, 1997) when still more impressive room-temperature results were obtained by adding Yb^{3+} as a codopant (at 3 at%) to an Er^{3+} (1%):YLF sample and exploiting the energy-transfer pump mechanism depicted in Figure 8.13. In this case, pumping at 959 nm into a 3-mm crystal yielded 37 mW of output, a threshold of 570 mW with respect to the *incident* pump power, 57% absorption of the pump light, and a slope efficiency of 3.4% with respect to the incident power. Unfortunately, the back transfer cross-relaxation process



helped to deplete the population in the upper laser level. By contrast, a comparable laser crystal with no Yb^{3+} codoping yielded a higher threshold power and half

the output power. In the latter case, a doubly-resonant two-photon upconversion process was responsible for filling of the upper laser level.

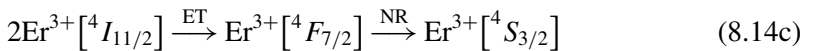
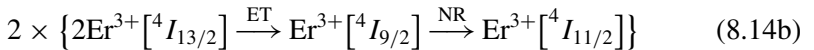
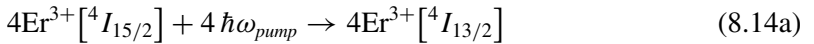
Returning to cw experiments performed at cryogenic temperatures, McFarlane (1991) following his initial work exploited a high-power cw Ti:S laser in order to yield the highest output power ever measured for a bulk upconversion laser. Using a 5-mm laser crystal doped at 5 mol%, 500 mW of green output was obtained with 4.4 W of pump power at 796.9 nm. Dielectric coatings were deposited directly on the plane parallel endfaces of the crystal; one end was coated with a 550-nm high reflector while the other was anti-reflection coated. With the sample mounted in a cryostat fitted with anti-reflection-coated access windows, optimum output coupling of 10% was provided by an external partial reflector (Figure 8.9(c)). Maximum output was obtained at 49 K and a linear dependence of output power on pump power was observed over the full range of applied pump powers above threshold. The latter fact is a common feature of upconversion laser experiments: although one expects a nonlinear dependence of output vs input given the multi-photon character of upconversion pumping, saturation of one or more of the upconversion steps will tend to linearize the output. The concept of slope efficiency is therefore still useful under these circumstances; McFarlane quoted a value of 14% for this quantity. However, perhaps the most important result of this high-power experiment was that no evidence was discovered for saturation of the output all the way up to the maximum pump power. Moreover, although the self-Q-switched behavior characteristic of 551-nm oscillation in Er:YLF was likewise observed at most pump powers, at the highest powers true cw output was achieved. On the other hand, losses in this experiment were higher than expected – the lowest threshold pump power was a sizable 80 mW despite the choice of a 99.5% reflectivity output coupler. One suggested source for this loss was energy transfer out of the $^4S_{3/2}$ upper laser level.

Another key result obtained with the $^4S_{3/2} \rightarrow ^4I_{15/2}$ Er³⁺ green transition in YLF laser crystals has been successful diode laser pumping on the $^4I_{15/2} \rightarrow ^4I_{9/2}$ transition. Using a 2-mm monolithic laser crystal with 0.5% output coupling, Tong *et al.* (1989) measured a threshold pump power of 34 mW at a sample temperature of 40 K using as a pump source an AlGaAs diode array. Although a slope efficiency of only 1% with respect to absorbed laser power was obtained, this may have been as much a consequence of low output coupling than inefficient pumping. Nevertheless, substitution for the diode laser of a cw dye laser reduced the threshold to 17 mW and increased the slope efficiency to 4%, thus indicating the degree to which the poorer beam quality or broader linewidth of the semiconductor pump affected the laser's performance. Stephens and McFarlane (1993), however, obtained dramatically improved results by carefully controlling the frequency and linewidth of a 3-W diode pump laser. These workers obtained an output power of

100 mW in the green in a 5-mm-long Er(5%):YLF monolithic laser cavity held at 48 K; a slope efficiency of 8% was measured for output coupling of 0.5%.

Extending their work on 1.5- μm pumping of Er:YLF to the visible domain, Xie and Rand (1990a) used a 3-mm crystal doped at 5% to demonstrate cooperative three-ion upconversion and green laser output at 551 ($^4S_{3/2}(1) \rightarrow ^4I_{15/2}(4)$) and 544 nm ($^4S_{3/2}(1) \rightarrow ^4I_{15/2}(6)$). Maximum output power of 32 mW was obtained at 9 K with 16% output coupling; the oscillation wavelength shifted from 544 to 551 nm above 15 K. The output power varied linearly with pump power with a slope efficiency of 11.6%, a fact the authors attributed to saturation of the pump transition below the threshold pump power of 20–50 mW. An interesting feature of this experiment was that true cw oscillation was observed in contrast to the self-Q-switched operation seen in most all other studies of this material, especially at the high doping level of 5%. Because substantial excited-state absorption at the oscillation wavelength still existed on the $^4I_{13/2} \rightarrow ^2H_{9/2}$ transition, Xie and Rand attributed the absence of self-pulsing to the different initial conditions achieved by 1.5- μm pumping and the predominance of cooperative upconversion in creating the inversion. In particular, the latter effect yielded a slower response to changes in the intracavity intensity as compared with sequential multi-photon absorption, hence instabilities leading to self-pulsing may have been effectively damped out. In a second publication describing visible laser emission from this system, Xie and Rand (1992) added an intracavity acousto-optic modulator to the optical setup and demonstrated both Q-switched and mode-locked operation. A minimum pulse width of 180 ps was deduced for operation in the latter mode.

It is worth pointing out, however, that some disagreement exists concerning the importance of the triplet mechanism proposed by Xie and Rand. In their own investigation of 1.5- μm pumping of Er³⁺(5%):YLF, Macfarlane *et al.* (1992) believed that *sequential* energy-transfer steps were responsible for building of population in the $^4S_{3/2}$ upper laser level. Specifically, these authors favored the four-fold mechanism

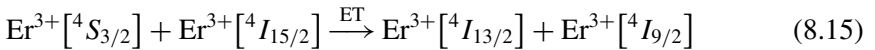


where for clarity ions delivered to the ground state are not explicitly displayed. A maximum output power of 10 mW was measured using a monolithic laser cavity at 35 K. Moreover, in contrast to the observations of Xie and Rand, the more typical self-pulsing behavior was obtained, even though experimental conditions were similar in the two experiments.

8.2.2.3 $\text{Er}^{3+}; {}^4\text{S}_{3/2} \rightarrow {}^4\text{I}_{15/2}$ transition in other host materials

Along with their room-temperature results in Er:YLF, Brede *et al.* (1993) and Danger *et al.* (1993) investigated pulsed visible output on the ${}^4\text{S}_{3/2} \rightarrow {}^4\text{I}_{15/2}$ green transition in YLF's sister compound KYF₄ as well as in YAG. Using a pulsed Ti:S laser tuned to 812 nm, a KYF₄ crystal doped with 1 at% Er^{3+} yielded 140 μJ of output at 561 nm, about a factor of 7 poorer performance than was obtained with their comparably doped YLF crystal. This result was due to significantly lower absorption of pump radiation in the KYF₄ sample; recall the doubly-resonant multi-photon absorption mechanism comprising Equation (8.11a) and Equation (8.12a). On the other hand, no output at all was obtained by single wavelength pumping of an $\text{Er}^{3+}(0.5\%):\text{YAG}$ sample, a result the authors partly attributed to the short 15- μs upper-level lifetime in this host material as compared to the 50- μs width of the pump pulse. However, addition of auxiliary cw pumping with a krypton ion laser ultimately induced oscillation in this system.

In addition to his studies of the YLF host medium, McFarlane (1994) published results of extensive spectroscopic studies and upconversion laser experiments on $\text{Er}^{3+}(5\%):\text{BaY}_2\text{F}_8$. Of greatest interest in this work was the observation of photon-avalanche pumping of the ${}^4\text{S}_{3/2}$ upper laser level for certain wavelengths shorter than those affiliated with the ${}^4\text{I}_{15/2} \rightarrow {}^4\text{I}_{9/2}$ ground-state absorption around 790 nm. In particular, on focusing the pump beam new features appeared in the excitation spectrum that could be assigned to the excited-state transitions ${}^4\text{I}_{11/2} \rightarrow {}^4\text{F}_{7/2}$ and ${}^4\text{I}_{13/2} \rightarrow {}^4\text{H}_{11/2}$ yet which did not significantly overlap any ground-state transitions. This implied the existence of an efficient cross-relaxation mechanism, possibly



that yielded large populations in the long-lived ${}^4\text{I}_{11/2}$ and ${}^4\text{I}_{13/2}$ excited states.

8.2.2.4 Er^{3+} ; other visible wavelengths

While the bulk of Er^{3+} -based visible upconversion laser experiments have been concerned with the high-gain ${}^4\text{S}_{3/2} \rightarrow {}^4\text{I}_{15/2}$ transition, upconversion output has been obtained at additional wavelengths as well. Aside from the 670-nm result obtained by Johnson and Guggenheim (1971), the first of these were reported by the prolific Hughes researchers, in this case working with their favorite composition $\text{Er}^{3+}(5\%):\text{YLF}$ (McFarlane, 1989, McFarlane *et al.*, 1990). With a 5-mm-long monolithic laser cavity held at 41 K, 26 mW of cw output on the ${}^4\text{F}_{9/2} \rightarrow {}^4\text{I}_{15/2}$ transition at 668 nm was obtained for 0.5% output coupling and optical pumping with a

2.1-W Ti:S laser tuned to the $^4I_{15/2} \rightarrow ^4I_{9/2}$ transition at 797 nm. Other red wavelengths produced with this same cavity, laser temperature, pump source, and pump power included 702 nm ($^2H_{9/2} \rightarrow ^4I_{11/2}$, 50 mW) and 618 nm ($^2P_{3/2} \rightarrow ^4F_{9/2}$, 500 μ W). Pumping mechanisms for the latter two transitions are not discussed in detail, although the energy transfer and multi-photon steps previously described by McFarlane (1989) were certainly of central importance.

The equally prolific IBM group was the first to produce blue output from an Er^{3+} upconversion laser (Hebert *et al.*, 1990): At an operating temperature of 16 K, a 3.5-mm monolithic YLF laser cavity with output coupling of 0.5% yielded 6 mW of output on the $^2P_{3/2} \rightarrow ^4I_{11/2}$ transition at 470 nm for optical pumping at 653 nm. Moreover, 2 mW of output was measured on this transition with this same laser at 10 K for Ti:S pumping at 969 nm. In the former experiment, Hebert *et al.* proposed the following sequence as the means by which population is built in the high-lying $^2P_{3/2}$ manifold:

$$2\text{Er}^{3+}[^4I_{15/2}] + 2\hbar\omega_{\text{pump}}(653 \text{ nm}) \rightarrow 2\text{Er}^{3+}[^4F_{9/2}] \quad (8.16a)$$

$$\begin{aligned} 3\text{Er}^{3+}[^2F_{9/2}] &\xrightarrow{\text{NR}} 2\text{Er}^{3+}[^4F_{9/2}] + \text{Er}^{3+}[^4F_{9/2}] \xrightarrow{\text{NR}} 2\text{Er}^{3+}[^4I_{9/2}] \\ &+ \text{Er}^{3+}[^4F_{9/2}] \xrightarrow{\text{NR}} 2\text{Er}^{3+}[^4I_{11/2}] + \text{Er}^{3+}[^4F_{9/2}] \end{aligned} \quad (8.16b)$$

$$\begin{aligned} 2\text{Er}^{3+}[^4I_{11/2}] + \text{Er}^{3+}[^4F_{9/2}] &\xrightarrow{\text{ET}} \text{Er}^{3+}[^4F_{7/2}] + \text{Er}^{3+}[^4F_{9/2}] \\ &\xrightarrow{\text{NR}} \text{Er}^{3+}[^4S_{3/2}] + \text{Er}^{3+}[^4F_{9/2}] \end{aligned} \quad (8.16c)$$

$$\text{Er}^{3+}[^4S_{3/2}] + \text{Er}^{3+}[^4F_{9/2}] \xrightarrow{\text{ET}} \text{Er}^{3+}[^2K_{13/2}] \xrightarrow{\text{NR}} \text{Er}^{3+}[^2P_{3/2}] \quad (8.16d)$$

where for clarity all Er^{3+} ions delivered to the ground state by energy transfer are not explicitly shown. Despite this complicated upconversion pathway, however, a threshold pump power of just 10 mW was measured. As for the infrared pumping experiment, an analogous mechanism was proposed, except that occupation of the $^4I_{11/2}$ was established by direct ground-state absorption at the 969-nm pump wavelength and pumping of the $^4F_{9/2}$ state was accomplished by the energy-transfer step

$$\text{Er}^{3+}[^4I_{11/2}] + \text{Er}^{3+}[^4I_{13/2}] \xrightarrow{\text{ET}} \text{Er}^{3+}[^4F_{9/2}] \quad (8.17)$$

Hence the latter four-photon process exhibited a higher threshold pump power of 200 mW as compared to the three-photon process Equations (8.16). (McFarlane (1994) likewise observed 470-nm output in his study of infrared pumped $\text{Er}^{3+}(5\%):\text{BaY}_2\text{F}_8$.) Hebert *et al.* further demonstrated yellow output at 560 nm in $\text{Er}:\text{YLF}$; in this case, upconversion pumping was accomplished according to the

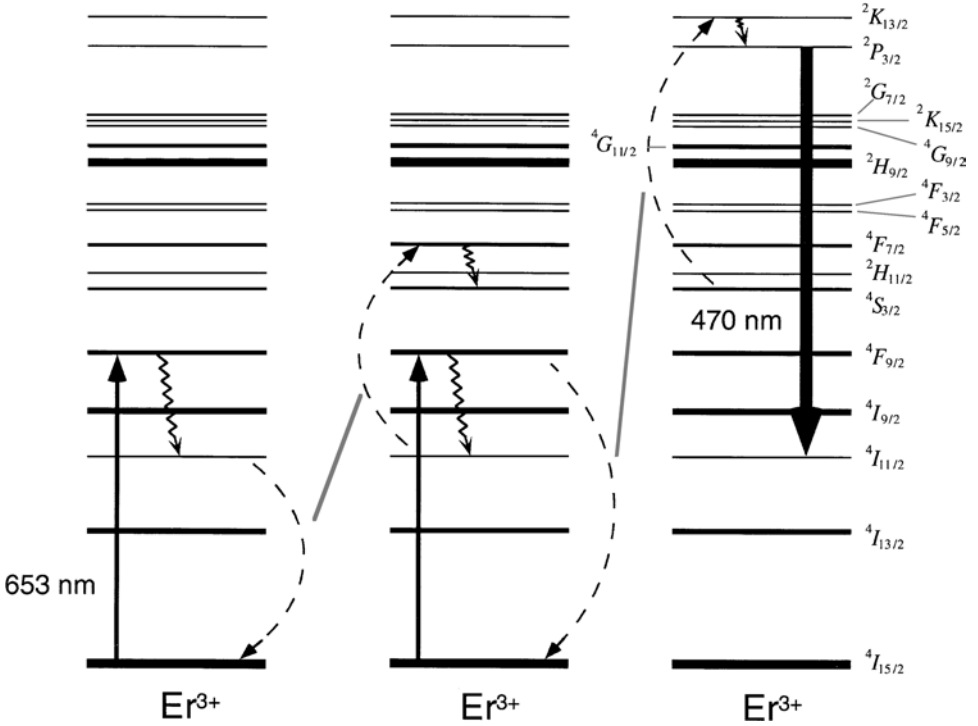
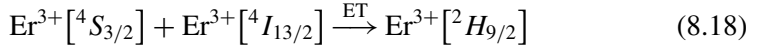


Figure 8.14: Complex upconversion pathway yielding population in the ${}^2P_{3/2}$ state of Er^{3+} . Note that the energy-transfer step shown in the two leftmost diagrams yields population in the ${}^4S_{3/2}$ state, whereas the energy transfer shown in the two rightmost diagrams shows how an ion already in the ${}^4S_{3/2}$ state is promoted to the upper laser level.

sequence of steps depicted in Figure 8.14 (Hebert *et al.*, 1990). An output power of 3.5 mW was measured on the ${}^2H_{9/2} \rightarrow {}^4I_{13/2}$ transition at a sample temperature of 10 K.

The IBM group went on to investigate upconversion output on the 470-, 551-, and 560-nm transitions following cw pumping at 1.5 μm in Er^{3+} (5%):YLF (Macfarlane *et al.*, 1992). Thus the energy of five pump photons is needed to populate the ${}^2P_{3/2}$ state, yet remarkably the threshold pump power for 470-nm output was just 80 mW. As mentioned at the conclusion of Section 8.2.2.2, these workers preferred to explain upconversion in this system in terms of sequential energy-transfer steps instead of n -fold cooperative upconversion. Macfarlane *et al.* therefore proposed the combination of processes Equations (8.14) and Equation (8.16d) as the mechanism behind population of the high-lying ${}^2P_{3/2}$ state; a total of eight 1.5- μm pump photons are required. A maximum 470-nm output power of $\sim 600 \mu\text{W}$ was measured at a pump power of 350 mW. As for output on the 560-nm yellow transition,

occupation of the $^2H_{9/2}$ upper laser level was accomplished by combining the steps of Equations (8.14) with the energy-transfer process



A maximum output of 12 mW was measured at 25 K for a pump power of 350 mW.

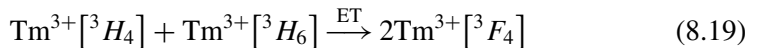
8.2.3 Tm^{3+} upconversion lasers

In the following discussion and figures, we have uniformly adopted the convention that the term label for the first-excited manifold of Tm^{3+} is $^3F_4$ while the label for the third-excited manifold is $^3H_4$.

8.2.3.1 $\text{Tm}^{3+}; ^1D_2 \rightarrow ^3F_4$ transition at ~ 450 nm

The first report of upconversion output from a Tm^{3+} -based laser was published by Nguyen *et al.* (1989) working at Los Alamos National Laboratory. Using an uncoated $\text{Tm}^{3+}(1\%):\text{YLF}$ crystal with plane-parallel faces, an optical cavity was constructed by surrounding this crystal with a pair of concave external dielectric mirrors, as shown in Figure 8.9(b). With 10% output coupling and optical pumping with a pair of pulsed dye lasers tuned to 781 and 649 nm, Nguyen *et al.* obtained pulsed blue output on the $^1D_2 \rightarrow ^3F_4$ transition at 450 nm. Figure 8.15 illustrates the two-photon sequential-absorption mechanism yielding population build up in the $^1D_2$ upper laser level. At a sample temperature of 75 K, 180 μJ of blue output was measured for 9.9 mJ of the infrared pump wavelength and 3.5 mJ of the red wavelength. Upon reducing the output coupling to 5%, doubling the crystal length from 1 to 2 cm, and increasing the doping level to 1.5%, Nguyen *et al.* (1990) obtained pulsed blue upconversion output at room temperature.

In later experiments of nearly identical design, Knüpfer *et al.* (1994) also achieved room-temperature operation at 450 nm with $\text{Tm}^{3+}(1%):\text{YLF}$, $\text{Tm}^{3+}(1\%), \text{Yb}^{3+}(10%):\text{YLF}$, and $\text{Tm}^{3+}:\text{LiGdF}_4$ laser crystals. For reasons not understood at the time, the best results were obtained with the double-doped sample: Output coupling of 0.5% yielded 2.5 μJ of 450-nm output for a total absorbed pump power from both pulsed pump lasers of ~ 2 mJ. A key issue in this work was the relative timing of the two pump lasers, one a pulsed DCM dye laser operating at 650 nm and the second a pulsed Ti:S laser. This issue derived from the gain quenching effect of the cross-relaxation step



which depletes the optically pumped intermediate $^3H_4$ state. The task was to optimize the arrival time of the 40-ns 650-nm pump pulse (which drove the $^3H_4 \rightarrow ^1D_2$

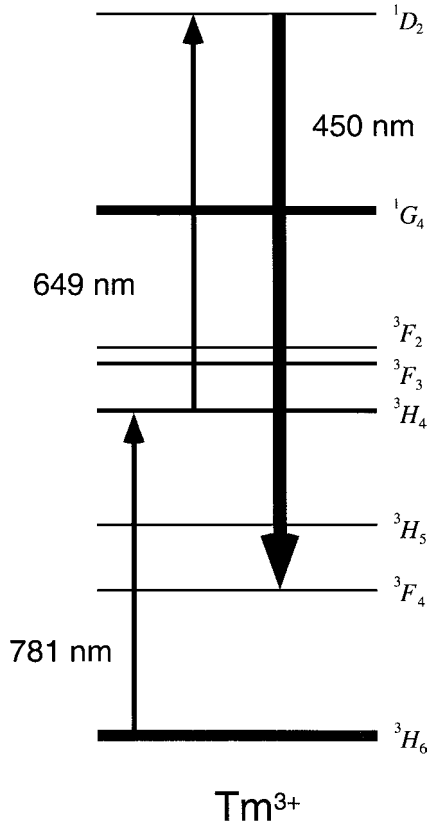
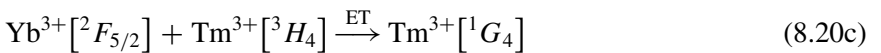
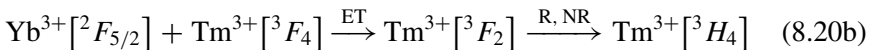
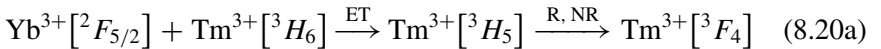


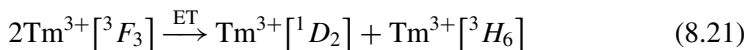
Figure 8.15: Pulsed sequential two-photon pumping mechanism used to yield blue upconversion output from Tm^{3+} (Nguyen *et al.*, 1989).

ESA transition) in light of Equation (8.19) and the 60- μs pulse width of the 780-nm pulse (which drove the $^3H_6 \rightarrow ^3H_4$ ground-state absorption transition). A delay of 70 μs yielded the maximum blue output.

Thrash and Johnson (1994) reported further positive results with Yb^{3+} double-doped material at room temperature, this time obtained with single-wavelength cw pumping of a BaY_2F_8 crystal. Using a Ti:S laser tuned to excite only the $^2F_{7/2} \rightarrow ^2F_{5/2}$ transition of Yb^{3+} at 960 nm, a triplet of energy-transfer events was proposed to explain visible fluorescence and upconversion laser output originating in the $^1G_4$ state. Specifically, the authors indicated the upconversion sequence



where superfluous ground-state species have not been written explicitly. This multiple-transfer mechanism yielded cw oscillation on the $^1G_4 \rightarrow ^3H_5$ transition (799 nm), cw oscillation on the $^1G_4 \rightarrow ^3F_4$ transition (649 nm), and pulsed oscillation on the $^1G_4 \rightarrow ^3H_6$ transition (482 nm). However, in order to explain fluorescence and laser output originating from the $^1D_2$ level, two additional upconversion pumping steps were proposed. The first of these was the phonon-assisted cross-relaxation step

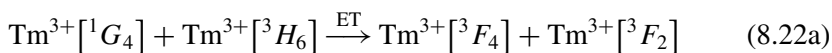


The second step required simultaneous cw oscillation at 649 nm, in which case ESA occurred on the nearly resonant $^3H_4 \rightarrow ^1D_2$ transition. Indeed, room-temperature output on the 456-nm $^1D_2 \rightarrow ^3F_4$ transition did not occur unless the 649-nm laser was on; still, the output consisted of a series of intermittent pulses. The laser cavity in this case consisted of a thin platelet (~ 1 mm) of $\text{BaY}_{1.0}\text{Yb}_{0.99}\text{Tm}_{0.01}\text{F}_8$ pressed against a flat end mirror reflective at both 649 and 456 nm. The cavity was closed with a concave high reflector in order to minimize the pump threshold. Replacement of the cw Ti:S pump laser with a pair of 100-mW diode lasers operating at the same wavelength successfully yielded oscillation at 456 nm, one of the few demonstrations of an all-solid-state visible upconversion laser operating at room temperature.

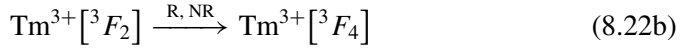
Returning to experiments involving singly doped YLF crystals, the IBM group (Hebert *et al.*, 1992) investigated two-photon 785-plus-648-nm sequential upconversion pumping using a cw DCM dye laser and a cw Ti:S laser. With a 3-mm monolithic laser cavity (Figure 8.9(a)) doped at 1.8 at% Tm^{3+} and held at 15 K, 9 mW of 450-nm output was obtained with output coupling of 0.5% and a total pump power of ~ 570 mW. The output, however, consisted of a semirandom 100–500-kHz train of pulses each 80–100 ns in width.

8.2.3.2 $\text{Tm}^{3+}; ^1G_4 \rightarrow ^3H_6$ transition at ~ 483 nm

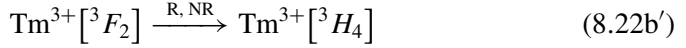
In the work just described, Hebert *et al.* also obtained cw blue output at 483 nm. However, only single-wavelength pumping at 628 nm was needed in order to populate the upper laser level owing to a photon-avalanche mechanism at work in the laser crystal. In particular, the ESA step $^3F_4 \rightarrow ^1G_4$ was resonant with the cw pump laser while two cross-relaxation mechanisms were proposed to help populate the $^3F_4$ intermediate level from the ground state. Both mechanisms share the initial cross-relaxation step



The first mechanism, however, proceeds by



while the second proceeds by

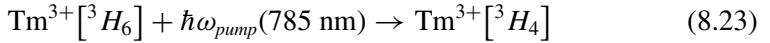


plus the additional cross-relaxation step

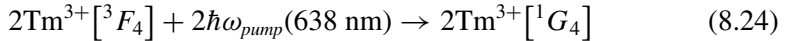


Thus the second mechanism delivers three Tm^{3+} ions to the metastable ${}^3F_4$ state for every sacrificed ${}^1G_4$ excitation while the first yields only two such ions. Using the same laser crystal and same mirror coatings as were used in obtaining 450-nm output, 30 mW of 483-nm output was obtained at a sample temperature of 26 K. As with the shorter-wavelength output, self-pulsing was observed at the longer wavelength. The 483-nm oscillation persisted up to a temperature of 110 K, but switching of the pump wavelength to 648 nm yielded oscillation up to 160 K, albeit at reduced output power.

The cross-relaxation step Equation (8.22c') was also implicated in a 486-nm Tm^{3+} upconversion-laser experiment involving a YAG host material (Scott *et al.*, 1993). In this case, two-wavelength pumping yielded population build up in the ${}^1G_4$ upper laser level according to the sequence



followed by Equation (8.22c') and finally



Using a monolithic laser cavity doped at 3 at% Tm^{3+} , 70 μW of quasi-continuous pulsed output was measured at 12 K.

Thrash and Johnson (1994) also obtained room-temperature output at 483 nm with Tm^{3+} , Yb^{3+} double-doped BaY_2F_8 samples. Like the 456-nm output, intermittent pulsing was observed. After attaching the sample to a liquid nitrogen cold finger, however, these workers evidently obtained true cw oscillation at 483 nm.

8.2.3.3 Tm^{3+} ; other visible wavelengths

In another early demonstration of a true visible upconversion laser operating at room temperature, Antipenko *et al.* (1990) obtained pulsed oscillation on the 649-nm ${}^1G_4 \rightarrow {}^3F_4$ transition in a BaYb_2F_8 crystal doped with 0.2 at% Tm^{3+} . These workers were the first to exploit the multiple-transfer mechanism

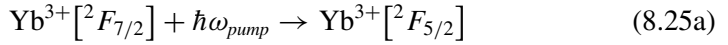
Equations (8.20), in this case using a pulsed 1.05- μm Nd laser as a pump source to drive the $\text{Yb}^{3+} {}^2F_{7/2} \rightarrow {}^2F_{5/2}$ transition. Thrash and Johnson (1994) likewise observed room-temperature cw oscillation at 649 nm; the role played by the 649-nm intracavity radiation in helping populate the ${}^1D_2$ state has already been discussed. As with the 456-nm output, intermittent pulsed output on the ${}^1D_2 \rightarrow {}^3H_5$ transition at 512 nm was obtained with the help of 649-nm ESA. Cw diode pumping yielded oscillation at all three wavelengths with the sample at room temperature. Room-temperature oscillation on yet two more transitions of Tm^{3+} was obtained in this work: Again with the benefit of the multiple-transfer mechanism Equations (8.20), true cw output was observed for the ${}^1G_4 \rightarrow {}^3H_5$ at 799 nm. Taking advantage of the existing ${}^1D_2$ population and a fourth $\text{Yb} \rightarrow \text{Tm}$ radiationless transfer event, population was also built in the very high lying ${}^1I_6$ state, which yielded oscillation at 457 nm on the ${}^1I_6 \rightarrow {}^3F_4$ transition when the sample was cooled by a liquid nitrogen cold finger.

8.2.4 Pr^{3+} upconversion lasers

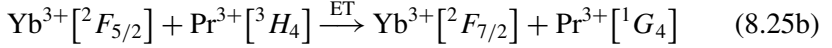
Unlike erbium and thulium, very few upconversion laser experiments have been performed using Pr^{3+} ions in a bulk gain medium. The very first of these was undertaken by Koch *et al.* (1990) and was described in Section 8.1. In this work – one of the first to exploit the photon-avalanche mechanism in an upconversion laser (Figure 8.8) – optimum output of 230 mW at 80 K was obtained on the ${}^3P_0 \rightarrow {}^3F_2$ transition at 644 nm following single-wavelength cw pumping at 677 nm. The laser cavity consisted of a 1.22-mm platelet of $\text{Pr}^{3+}(7\%):\text{LaCl}_3$ mounted in a cryostat fitted with uncoated access windows. One of these served as the output coupler, while an external high-reflectivity concave mirror served as the end reflector. Note that only the Fresnel reflection from the cryostat window provided optical feedback in this experiment, a testament to the unusually high stimulated-emission cross-section of the $\text{Pr}^{3+} {}^3P_0 \rightarrow {}^3F_2$ transition. The threshold pump power was just 100 mW. Even more remarkable was that oscillation persisted up to a temperature of 210 K, a good indication that room-temperature operation was well within reach.

In fact, cw room-temperature operation on this transition was demonstrated for the first time six years later by Sandrock *et al.* (1996). Using a $\text{Pr}^{3+}(1\%), \text{Yb}^{3+}(10\%):\text{YLF}$ laser crystal 3.2 mm in length, 75 mW of output at 640 nm was obtained for 9.3 W of pump power from a cw Ti:S laser tuned to ~ 830 nm. The laser resonator consisted of separate concave mirrors in a confocal configuration with the laser crystal at the resonator waist; the threshold pump power was 3 W. That single-wavelength infrared pumping of this system was effective in yielding population inversion was a result of a different type of photon-avalanche mechanism first proposed a year before (Xie and Gosnell, 1995); definitive experimental evidence for

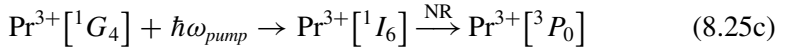
this proposal was presented by Sandrock *et al.* (1996) and by Gosnell (1997). Specifically, the upconversion sequence begins with the ground-state-absorption step



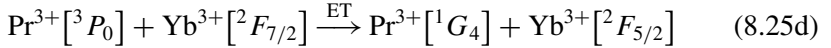
which is only weakly driven due to the short pump wavelength relative to the peak of the Yb^{3+} absorption. Next, energy transfer according



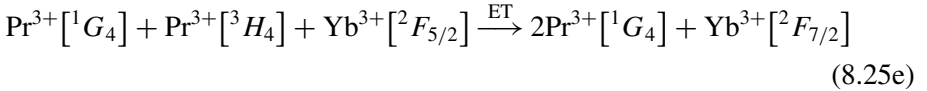
establishes a population in the ${}^1G_4$ state. The ESA step



then establishes a small seed population in the upper laser level. The photon avalanche is driven by the *mixed-species* cross-relaxation step

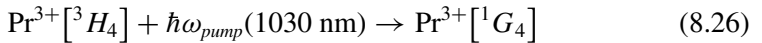


which allows for the additional energy-transfer step



thus sustaining of the avalanche through the ESA step Equation (8.25c). Clearly it is of great advantage in this experiment to use as high an Yb^{3+} concentration as possible in order to enhance the efficiency of this last step.

Still a third pumping mechanism for a Pr^{3+} -based upconversion laser, also operating at room temperature, was demonstrated by Binun *et al.* (1996). In this experiment, simple two-photon pumping of the upper laser level resulted from the ground-state-absorption step



This first step was accomplished by intracavity pumping within a cw Yb:YAG laser. With the help of a cw Ti:S tuned to 842 nm, the ESA step Equation (8.25c) then filled the upper laser level. Output coupling of 0.3% at 640 nm yielded ~ 2.3 mW of output at this red wavelength. Binun *et al.* also demonstrated orange output on the ${}^3P_0 \rightarrow {}^3H_6$ transition at 607 nm, in this case obtaining 3.5 mW of cw output.

8.2.5 Nd^{3+} upconversion lasers

Still fewer upconversion laser experiments have been conducted by using Nd^{3+} as the active ion. Despite this apparent lack of attention, however, the ion is

especially interesting as it has yielded the shortest wavelength upconversion lasers demonstrated to date.

The entirety of Nd^{3+} upconversion research in bulk materials has been conducted by R. M. Macfarlane and colleagues at IBM. Initially, two different two-photon sequential absorption pumping mechanisms were explored (Macfarlane *et al.*, 1988), as shown in Figure 8.16. Using the first of these pumping mechanisms (Figure 8.16(a)), 12 mW of cw output power at 380 nm ($^4D_{3/2} \rightarrow ^4I_{11/2}$) was obtained from a monolithic laser cavity (Figure 8.9(a)) formed of $\text{Nd}^{3+}(1\%):\text{LaF}_3$ at a temperature of 20 K. The total pump power in this case was ~ 390 mW. By contrast, the second pumping scheme (Figure 8.16(b)) yielded at the same temperature only 3.5 mW for a total pump power of ~ 550 mW. While the two-photon mechanisms depicted in Figure 8.16 were favored by the authors, they also reported a possible radiationless process that may have contributed to upconversion pumping in this system. Specifically, the population reservoir residing in the long-lived $^4F_{3/2}$ state combined with the population in the ground-state-absorption-pumped $^4G_{5/2}$ state

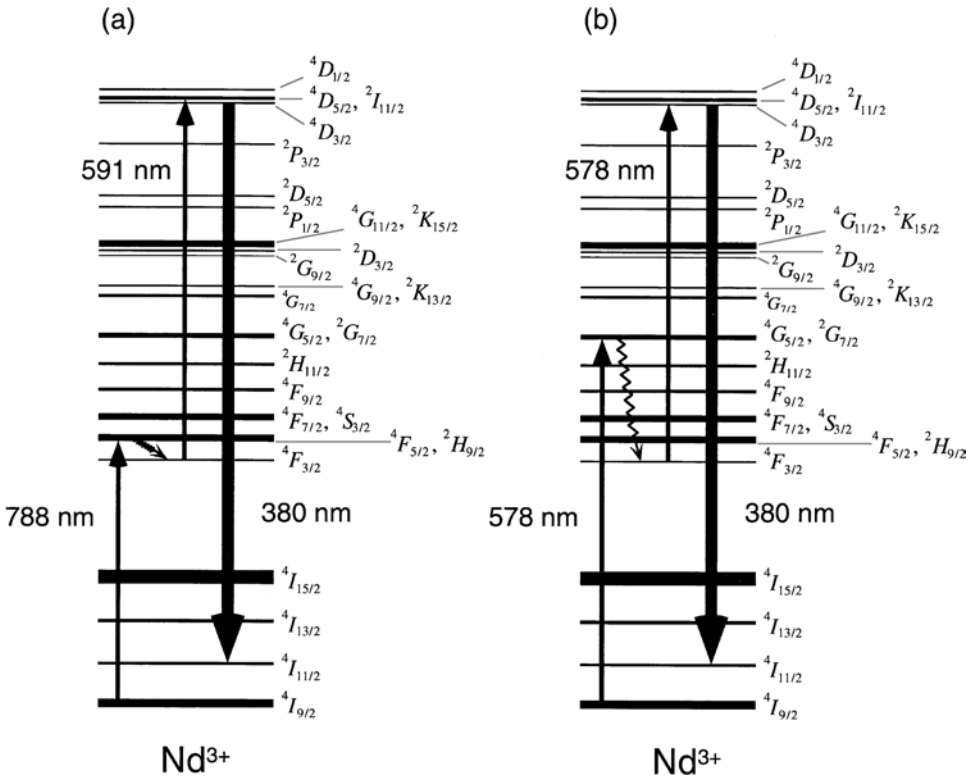
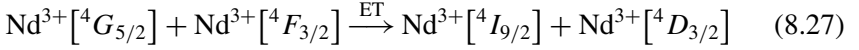


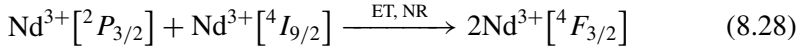
Figure 8.16: Violet upconversion laser based on sequential two-photon pumping of Nd^{3+} (Macfarlane *et al.*, 1988): (a) two-wavelength mechanism; (b) single-wavelength mechanism.

made possible the phonon-assisted transfer step



However, the short 40-ns lifetime of the $^4G_{5/2}$ state along with the expectation that cw pumping would only weakly populate this level argued against a large contribution from this upconversion channel.

In contrast to the LaF_3 host material, the lifetime of the $^4D_{3/2}$ state in YLF is so short (1 μs at cryogenic temperatures) that the next lower $^2P_{3/2}$ level becomes populated by nonradiative decay. Thus Lenth and Macfarlane (1990) in further experiments with the Nd^{3+} ion obtained blue output on the 413-nm $^2P_{3/2} \rightarrow ^4I_{11/2}$ transition. However, the 603-nm pump wavelength used in this experiment was not resonant with any ground-state transition; hence the authors asserted that a photon avalanche was responsible for upconversion pumping in this host. The cross-relaxation step was



from which ESA on the $^4F_{3/2} \rightarrow ^4D_{3/2}$ transition at 603 nm repopulated the upper laser level.

8.3 UPCONVERSION FIBER LASERS

An upconversion laser in the format of a single-mode optical fiber offers this crucial advantage: Because fiber cores are typically only a few microns in diameter, the ability to create large population densities over very long distances yields potentially very high round-trip gains. Further, the ability to maintain a high pump intensity over long distances ensures high efficiency for the nonlinear processes at the heart of all upconversion mechanisms. A principal disadvantage of the fiber geometry is that almost exclusively glassy materials must be used in fabricating the fiber, which implies strong inhomogeneous broadening of metastable levels as well as shortened level lifetimes, the latter effect being a result of the higher maximum phonon frequencies and therefore enhanced nonradiative relaxation exhibited by fluoride and silica glasses. Another disadvantage is that pump lasers of high beam quality are required in order to yield efficient coupling into narrow fiber cores. This makes optical pumping of fiber upconversion systems with diode lasers more problematic than is the case with bulk upconversion lasers insofar as high-power diode sources are often constructed of multiple emitters (see Chapter 3). Nevertheless, the advantages of the fiber format appear to have outweighed the disadvantages: far greater success has been obtained with fiber-based devices than with their bulk counterparts in demonstrations of room-temperature operation, as Table 8.2 reveals. Indeed, according to Tables 8.1

Table 8.2. Upconversion fiber laser experiments

Ion	Codopant	Host	Pump mechanism	Pump source	Pump wavelength	Operating temperature	Transition, wavelength	Max output power or energy	Reference
Er ³⁺		ZBLAN	MP	cw Ti:S laser	801 nm	298 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 546 nm	23 mW	Whitley <i>et al.</i> , 1991
Er ³⁺		ZBLANP	MP	cw Ti:S laser	800 nm	298 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 548 nm	10 mW	Hirao <i>et al.</i> , 1992
Er ³⁺		ZBLAN	MP	cw Ti:S laser	970 nm	298 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 546 nm	50 mW	Allain <i>et al.</i> , 1992
Er ³⁺		ZHBL- AYNLIP	MP	cw diode laser	801 nm	298 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 543 nm	3 mW	Massicott <i>et al.</i> , 1993
Er ³⁺		ZBLAN	MP	cw InGaAs laser	971 nm	298 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 546 nm	12 mW	Piehler and Craven, 1994
Er ³⁺		SiO ₂	MP	cw Nd:YAG laser	1.32 μ m	298 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 546 nm (ASE)	unreported	Nicácio <i>et al.</i> , 1994
Er ³⁺		ZBLA	MP	cw Ti:S laser	790 nm	77 K	$^4S_{3/2} \rightarrow ^4S_{13/2}$ 850 nm	500 μ W	Saissy <i>et al.</i> , 1995
Er ³⁺						298 K	$^4S_{3/2} \rightarrow ^4S_{13/2}$ 850 nm	1.3 mW	
Er ³⁺		ZBLAN	MP	cw Ti:sapphire laser	800 nm	298 K	$^4S_{3/2} \rightarrow ^4I_{15/2}$ 546 nm	850 μ W	Boj <i>et al.</i> , 1995
Ho ³⁺		ZBLAN	MP	cw Kr ⁺ ion laser	647 nm	298 K	$^5S_2 \rightarrow ^5I_8$ 540–553 nm	10 mW @ 550 nm	Allain <i>et al.</i> , 1990a
Ho ³⁺		ZBLAN	MP	cw dye laser	640–653 nm	300 K	$^5S_2 \rightarrow ^5I_8$ 550 nm	12 mW	Funk <i>et al.</i> , 1993
Ho ³⁺		ZBLAN	MP	cw dye laser	643 nm	298 K	$^5S_2 \rightarrow ^5I_8$ 550 nm	38 mW	Funk <i>et al.</i> , 1994, 1994a
Ho ³⁺		ZBLAN	MP	cw dye laser	643 nm	298 K	$^5S_2 \rightarrow ^5I_8$ 539–550 nm	38 mW	Funk <i>et al.</i> , 1996
Ho ³⁺		ZBLAN	MP	cw diode laser	643 nm	298 K	$^5S_2 \rightarrow ^5I_8$ 549 nm	1.2 mW	Funk <i>et al.</i> , 1997
Tm ³⁺		ZBLAN	MP	cw Kr ⁺ ion laser	647 + 676 nm	77 K	$^1G_4 \rightarrow ^3H_6$ 480 nm	400 μ W	Allain <i>et al.</i> , 1990a
						77 K	$^1D_2 \rightarrow ^3F_4$ 455 nm	single pulse at pump turn on	

Tm^{3+}	ZBLAN	MP	cw Nd:YAG laser	1.12 μm	298 K	$^1G_4 \rightarrow ^3H_6$ 480 nm	57 mW	Grubb <i>et al.</i> , 1992
Tm^{3+}	Yb^{3+}	MP				$^1G_4 \rightarrow ^3F_4$ 650 nm	unreported	
Tm^{3+}	Yb^{3+}	MP + ET	unreported	960 nm	298 K	$^1G_4 \rightarrow ^3F_4$ 649 nm	unreported	Jarman <i>et al.</i> , 1993
Tm^{3+}	ZBLAN	MP	cw Nd:YAG + dye lasers	1064 + 645 nm	298 K	$^1D_2 \rightarrow ^3F_4$ 455 nm	3 mW	Le Flohic <i>et al.</i> , 1994
Tm^{3+}	ZBLAN	MP	cw Nd:YAG + dye lasers	1120 + 680 nm	298 K	$^1G_4 \rightarrow ^3H_6$ 480 nm	15 mW	Tohmon <i>et al.</i> , 1995
Tm^{3+}	ZBLAN	MP	cw InGaAs-AlGaAs laser	1130 nm	298 K	$^1G_4 \rightarrow ^3H_6$ 482 nm	106 mW	Sanders <i>et al.</i> , 1995
Tm^{3+}	ZBLAN	MP	cw InP laser	1135 nm	298 K	$^1G_4 \rightarrow ^3H_6$ 482 nm	5 mW	Booth <i>et al.</i> , 1996
Tm^{3+}	ZBLAN	PA	cw Nd:YLF laser	1053 nm	298 K	$^3H_4 \rightarrow ^3H_6$??? 810 nm	130 mW (cw mode locked)	Yang <i>et al.</i> , 1996
Tm^{3+}	ZBLAN	MP	cw diode + dye lasers	1210 + 649 nm	298 K	$^1G_4 \rightarrow ^3H_6$ 482 nm	20 mW	Tohmon <i>et al.</i> , 1997
Tm^{3+}	Yb^{3+}	ET	cw Nd:silica laser	1065 nm	298 K	$^1G_4 \rightarrow ^3H_6$ 482 nm	106 mW	Zellmer <i>et al.</i> , 1997
Tm^{3+}	ZBLAN	MP	cw Nd:YAG laser	1123 nm	298 K	$^1G_4 \rightarrow ^3H_6$ 481 nm	230 mW	Paschotta <i>et al.</i> , 1997a
Pr^{3+}	ZBLAN	ET	cw Ti:S laser	844 nm	298 K	$^3P_0 \rightarrow ^3F_2$ 635 nm	20 mW	Allain <i>et al.</i> , 1991
Pr^{3+}	ZBLAN	MP	cw Ti:S laser	1010 + 835 nm	298 K	$^3P_0 \rightarrow ^3F_2$ 635 nm	185 mW	Smart <i>et al.</i> , 1991
						$^3P_0 \rightarrow ^3H_6$ 605 nm	30 mW	
						$^3P_1, ^1I_6 \rightarrow ^3H_5$ 520 nm	1 mW	
						$^3P_0 \rightarrow ^3H_4$ 491 nm	1 mW	

Table 8.2. (*cont.*)

Ion	Codopant	Host	Pump mechanism	Pump source	Pump wavelength	Operating temperature	Transition, wavelength	Max output power or energy	Reference
Pr^{3+}	Yb^{3+}	ZBLAN	MP + ET	cw InGaAs + AlGaAs lasers	1016 + 833 nm	298 K	$^3P_0 \rightarrow ^3F_2$ 635 nm $^3P_1, ^1I_6 \rightarrow ^3H_5$ 521 nm $^3P_0 \rightarrow ^3F_2$ 635 nm $^3P_1, ^1I_6 \rightarrow ^3H_5$ 521 nm	6.2 mW 700 μW 3.9 mW 370 μW	Piehler <i>et al.</i> , 1993
Pr^{3+}		ZBLAN	MP	cw Ti:S lasers	1017 + 835 nm	298 K	$^3P_0 \rightarrow ^3H_4$ 492 nm	9 mW	Zhao and Poole, 1994
Pr^{3+}		ZBLAN	MP	cw Ti:S lasers	1017 + 835 nm	298 K	$^3P_0 \rightarrow ^3H_4$ 492 nm	22 mW	Zhao <i>et al.</i> , 1995
Pr^{3+}		ZBLAN	MP	cw Ti:S lasers	1017 + 835 nm	298 K	$^3P_0 \rightarrow ^3H_4$ 492 nm	3 mW	Zhao, 1995
Pr^{3+}	Yb^{3+}	ZBLAN	ET	cw Ti:S laser	860 nm	298 K	$^3P_0 \rightarrow ^3F_2$ 635 nm	10 mW	Boj <i>et al.</i> , 1995
Pr^{3+}	Yb^{3+}	ZBLAN	PA	cw Ti:S laser	860 nm	298 K	$^3P_0 \rightarrow ^3F_2$ 635 nm $^3P_0 \rightarrow ^3H_6$ 605 nm $^3P_1 \rightarrow ^3H_5$ 521 nm $^3P_0 \rightarrow ^3H_4$ 491 nm	300 mW 44 mW 20 mW 4 mW	Xie and Gosnell, 1995
Pr^{3+}	Nd^{3+}	ZBLANP	MP + ET	unreported	796 nm	298 K	$^3P_0 \rightarrow ^3F_2$ 635 nm $^3P_0 \rightarrow ^3H_4$ 488 nm $^3P_0 \rightarrow ^3F_2$ 635 nm $^3P_0 \rightarrow ^3H_6$ 602 nm	unreported	Goh <i>et al.</i> , 1995
Pr^{3+}	Yb^{3+}	ZBLAN	ET	cw diode laser	860 nm	298 K		3.9 mW 200 μW	Baney <i>et al.</i> , 1995

Pr^{3+}	ZBLAN	MP	cw diode lasers	1017 + 835 nm	298 K	$^3P_0 \rightarrow ^3H_4$ 492 nm	1.2 mW	Zhao and Fleming, 1996
Pr^{3+}	ZBLAN	MP	cw diode lasers	1017 + 831 nm	298 K	$^3P_0 \rightarrow ^3H_4$ 492 nm	1 mW	Baney <i>et al.</i> , 1996
Pr^{3+}	Yb^{3+} ZBLAN	ET	cw diode laser	856 nm	298 K	$^3P_1 \rightarrow ^3H_5$ 520 nm $^3P_0 \rightarrow ^3H_4$ 492 nm	700 μW 700 μW	Baney <i>et al.</i> , 1996a
Pr^{3+}	ZBLAN	MP	cw Yb:silica + Ti:S lasers	1020 + 840 nm	298 K	$^3P_0 \rightarrow ^3F_2$ 635 nm $^3P_1 \rightarrow ^3H_5$ 520 nm $^3P_0 \rightarrow ^3H_4$ 491 nm	54 mW 18 mW 7 mW	Pask <i>et al.</i> , 1997
Pr^{3+}	Yb^{3+} ZBLAN	PA	cw Ti:S laser	850 nm	298 K	$^3P_0 \rightarrow ^3F_2$ 635 nm	1020 mW	Sandrock <i>et al.</i> , 1997
Pr^{3+}	Yb^{3+} ZBLAN	PA	cw Ti:S laser	840 nm	298 K	$^3P_0 \rightarrow ^3F_2$ 635 nm $^3P_1 \rightarrow ^3H_5$ 520 nm	440 mW 100 mW	Zellmer <i>et al.</i> , 1998
Nd^{3+}	ZBLAN	MP	cw dye laser	590 nm	298 K	$^4D_{3/2} \rightarrow ^4I_{11/2}$ 381 nm	74 μW	Funk <i>et al.</i> , 1994
Nd^{3+}	ZBLAN	MP	cw dye laser	590 nm	298 K	$^3P_{3/2} \rightarrow ^4I_{11/2}$ 412 nm	500 μW	Funk <i>et al.</i> , 1995

MP: multiphonon
PA: photon avalanche
ET: energy transfer
Z: ZrF₄; B: BaF₂; L: LaF₃; A: AlF₃; N: NaF; P: PbF₂; H: HaF₄; Y: YF₃; Li: LiF

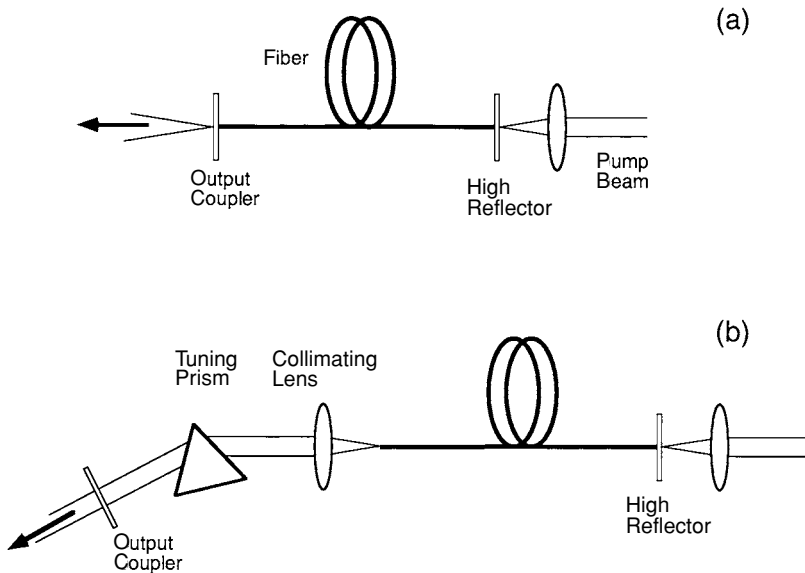


Figure 8.17: Basic optical configurations for fiber upconversion lasers. (a) Dielectric mirrors are butt-coupled to the fiber ends with pump light injected through the high reflector. Hence, this mirror is designed to yield high transmission at the pump wavelength and high reflectivity at the output wavelength. The output coupler on the opposite end may be designed to yield good reflectivity at the pump wavelength so as to optimize pump absorption. Index matching fluid may be applied between the mirrors and the fiber ends in order to minimize loss. Dielectric mirrors deposited directly onto the fiber ends are sometimes employed in order to yield still lower loss. (b) A tunable upconversion fiber laser, where the laser output wavelength is selected by an intracavity prism.

and 8.2, reports of fiber experiments published since 1993 outnumber those on bulk lasers by well over 2 to 1. P. W. France (1991) has compiled an anthology of articles that further discuss the optics and physical properties of rare-earth-based fiber lasers and amplifiers (France and Brierley 1990).

Another advantage of fiber lasers over bulk lasers is the simplicity of the optical design. A generic upconversion fiber laser experiment is shown in Figure 8.17(a). The most common means for introducing feedback in fiber laser cavities is to press-fit flat dielectric mirrors against the fiber ends, a procedure often termed “butt-coupling” in the literature. Index matching fluid is commonly applied to the fiber–mirror interface in order to minimize optical losses. A rarely used alternative approach is to deposit dielectric mirrors directly onto the fiber ends; in this way, optical losses are held to an absolute minimum. In either case, most of the difficulty in aligning a fiber laser experiment then becomes that of precise focusing of the pump beam onto the fiber core. When diode laser pumping is used, an anamorphic prism pair may be added to the optical set up in order to symmetrize the profile of the diode laser’s focused spot. Occasionally, the intracavity beam is collimated as

it exits the fiber and a tuning prism is interposed between the collimating optic and a separately adjustable output coupler, as shown in Figure 8.17(b).

The dramatic upsurge in upconversion fiber laser research was a direct result of the development of high-quality ZBLAN glass optical fibers. While the main impetus for the enormous investment that both public and private research institutions have made in this material was its potential applications in ultra-low-loss fiber telecommunications systems, its value in yielding lower rates of nonradiative relaxation compared with silica immediately recommended its application to the upconversion laser problem. As a glance at Table 8.2 indicates, ZBLAN currently dominates upconversion fiber laser research. Further information on fluoride glasses and fibers may be found in the book by Aggarwal and Lu (1991). France and Brierly (1991) have discussed the spectroscopic properties of rare-earth ions in fluorozirconate glass.

As only a single report of upconversion output from a fiber laser involved a working temperature lower than 300 K, the remainder of this chapter will assume that the lasers discussed are at room temperature unless otherwise specified.

8.3.1 Er^{3+} fiber lasers; $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$ transition at 556 nm

Except for a few experiments concerning upconversion-pumped infrared lasers (Millar *et al.*, 1990, Saissy *et al.*, 1995), the entirety of Er^{3+} upconversion fiber lasers are based on the $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$ transition at 556 nm. The first such experiment was conducted by Whitley *et al.* (1991) of British Telecom Laboratories, who used doubly-resonant two-photon sequential pumping of a ZBLAN fiber doped with 0.05 wt% Er^{3+} in order to obtain 23 mW true cw output in the green with 780 mW of absorbed Ti:S pump-laser power at 801 nm. The laser comprised a 2.4-m length of fiber with butt-coupled cavity mirrors; the output coupler reflectivity was 65%. Figure 8.18 illustrates the two-photon pumping scheme used in this experiment and also indicates the ESA step $^4\text{I}_{13/2} \rightarrow ^2\text{H}_{9/2}$ coincident with the output wavelength. This is the very same ESA transition that was deemed responsible for self-Q-switched output in many of the Er^{3+} bulk laser experiments described previously. In this case, however, no self-pulsing was observed, a result one can reasonably conclude derived from the broader and therefore more difficult to saturate transitions characteristic of glassy host materials. A slope efficiency of 11% against the absorbed pump power was observed in this experiment, at least at pump powers below ~ 250 mW. Unfortunately, at higher pump power the green output saturated apparently as a result of competition with oscillation on the high gain $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{13/2}$ transition at 850 nm.

Allain *et al.* (1992) recognized that gain on this near-infrared transition could be suppressed by pumping at 970 nm instead of 800 nm. In particular the ESA transition

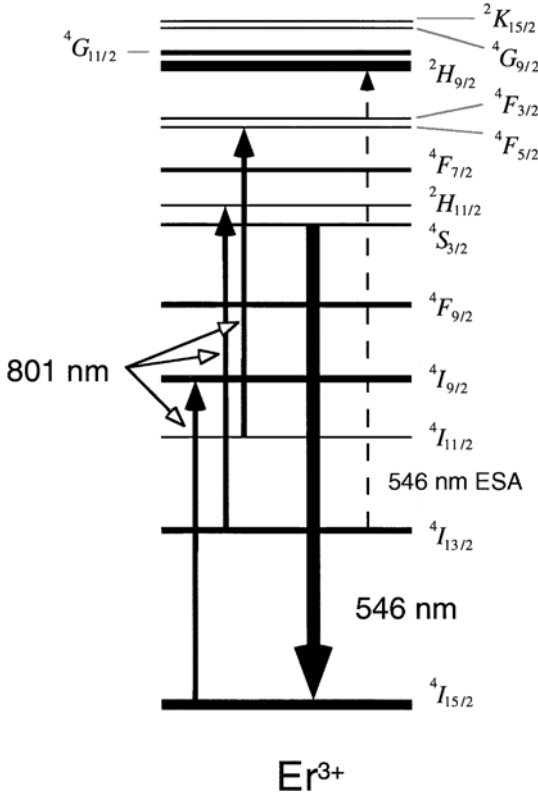


Figure 8.18: Green upconversion laser based on single-wavelength sequential two-photon pumping of Er^{3+} . Two possible upconversion pathways are shown, while the dashed arrow indicates excited-state absorption that compromises the laser performance (Whitley *et al.*, 1991).

$^4I_{13/2} \rightarrow ^2H_{11/2}$ at 800 nm is no longer active and thus population is stored in the $^4I_{13/2}$ state, in turn eliminating the inversion with respect to the $^4S_{3/2}$ state. At the same time, doubly-resonant sequential two-photon upconversion pumping of the upper laser level occurs according to

$$\text{Er}^{3+}[^4I_{15/2}] + \hbar\omega_{\text{pump}}(970 \text{ nm}) \longrightarrow \text{Er}^{3+}[^4I_{11/2}] \quad (8.29a)$$

$$\text{Er}^{3+}[^4I_{11/2}] + \hbar\omega_{\text{pump}}(970 \text{ nm}) \longrightarrow \text{Er}^{3+}[^4F_{7/2}] \xrightarrow{\text{NR}} \text{Er}^{3+}[^4S_{3/2}] \quad (8.29b)$$

In order to further suppress the gain on the 850-nm transition, however, Allain *et al.* added an intracavity prism adjusted so as to optimize feedback for the green wavelength (Figure 8.17(b)). Finally, these workers established simultaneous oscillation on the 1.5- μm $^4I_{13/2} \rightarrow ^4I_{15/2}$ transition, which reduced signal ESA on the green $^4I_{13/2} \rightarrow ^2H_{9/2}$ transition as well as helped quickly repopulate the ground state

with ions otherwise lost to the upconversion pathway. Unlike the case of 800-nm pumping, no saturation of the green output was observed; 50 mW of output power was measured for a 1.2-m fiber and 35% output coupling. In this experiment, the slope efficiency was 15%, with output power varying linearly with incident pump power up to 900 mW.

Combining the use of tuning prisms and 800-nm pumping, Hirao *et al.* (1992) obtained 15 mW of output at 548 nm for 700 mW of pump power incident on the pump input optic. The output power in this experiment likewise did not saturate; the measured slope efficiency with respect to the incident pump power was 7%.

Encouraged by the results just described, which involved purely Ti:S pumping, two groups have separately investigated both 800- and 970-nm diode-laser pumping of Er^{3+} -doped fluorozirconate fibers. In the first of these, the British Telecom group continued to work with 801-nm pumping of a ZHBLAYNLiP fiber doped at 0.05 wt% Er^{3+} . Using a large numerical aperture of 0.4 and 35% output coupling, an output power of 3 mW in the green was measured for 40 mW of pump power launched into a 3-m-long fiber. The threshold pump power in this experiment was only ~ 10 mW, a result of the large numerical aperture and low loss of the fiber. Because of the low pump powers used, the saturation effect previously described by these workers was not observed. A year later Piehler and Craven (1994) demonstrated four times greater output power in a 971-nm pumped ZBLAN fiber doped at 0.1 wt%. With output coupling of 29%, an NA-0.31 fiber 1.5 m in length, and an incident pump power of 150 mW from an InGaAs laser diode, 11.7 mW of output at 544 nm was measured.

With an eye toward diode-laser pumping, which demands low pump thresholds owing to the lower output powers and poorer beam quality of diode-laser sources, Boj *et al.* (1995) investigated Ti:S laser pumping of ZBLAN fibers with dielectric mirrors deposited directly onto the fiber end faces. A 38-mW threshold against launched 800-nm pump power was measured for a 120-cm fiber doped at 0.1 wt% and possessing end-mirror reflectivities in the green of 94%. (For convenience, both fiber ends were coated simultaneously.) A slope efficiency of 2% was measured for this fiber up to an output power of ~ 0.8 mW, both values being low consistent with the high end-mirror reflectivities at the signal wavelength. These workers also experimented with different fiber lengths in the hope of optimizing the laser cavity with respect to the ever present ESA on the green $^4I_{13/2} \rightarrow ^2H_{9/2}$ transition. A 20-cm cavity with 86% reflectivity mirrors yielded a significantly lower threshold than a 60-cm cavity with 94% reflectivity mirrors (38 versus 57 mW), although the longer fiber yielded $\sim 30\%$ greater output power at the maximum incident pump power of 150 mW. Perhaps the most significant discovery in this work was that multi-layer oxide coatings could be successfully evaporated onto

the mechanically and thermally dissimilar fluoride fibers without evidence of surface defects. Further, these coatings supported pump intensities up to 4 MW/cm² without damage.

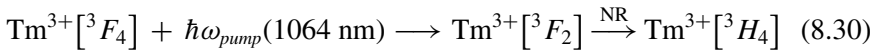
Other Er³⁺-based fiber experiments include upconversion pumping at 800 nm in order to yield 850-nm output (Millar *et al.*, 1990, Saissy *et al.*, 1995) and observation of amplified spontaneous emission at 546 nm in a single-mode erbium-doped germanosilicate fiber pumped at 1.32 μ m (Nicácio *et al.*, 1994).

8.3.2 Tm³⁺ fiber lasers

8.3.2.1 ¹D₂ $\rightarrow$ ³F₄ transition at 455 nm

The only upconversion fiber laser experiment conducted at other than room temperature was also the first: Allain *et al.* (1990) used a 170-cm length of thulium-doped ZBLAN fiber immersed in a liquid nitrogen bath to obtain blue output at both 455 and 480 nm. The fiber was doped at 0.125 wt% Tm³⁺ while optical feedback was provided by dielectric mirrors butt-coupled to the fiber ends, both of which were held above the cryogenic bath. A cw krypton-ion laser with its tuning prism removed served as the pump source; in this mode, the ion laser operated simultaneously on two red wavelengths, namely 676.4 and 647.1 nm. Hence two-photon sequential absorption populated the ¹D₂ upper laser level of the 455-nm transition, as shown in Figure 8.6. However, despite cw pumping, only a short output pulse at 455 nm was observed owing to the long 6.0-ms lifetime of the ³F₄ lower laser level. This blue transition was thus self-terminating in this experiment.

Four years later, however, room-temperature cw oscillation on the 455-nm transition was obtained by Le Flohic *et al.* (1994). The trick in this case was to combine 645-nm dye-laser pumping, which drove the two-photon pathway shown in Figure 8.19, with infrared pumping at 1064 nm. In this way, the energy storing ³F₄ lower laser level was depopulated through the ESA step



Hence the effective lifetime of the ³F₄ state was reduced while advantage was taken of the energy stored by an ion delivered to this state in returning it to the upconversion pathway. Best results were demonstrated with a 1.5-m fiber doped at 0.1 wt%; 3 mW of blue output was measured for 600 mW and 230 mW of red and infrared pump power, respectively, incident on the fiber's input end. Other ideas for depopulating the lower laser level have been described. These include codoping with either Eu³⁺ (Tohmon *et al.*, 1993) or Tb³⁺ (Percival *et al.*, 1993), (which quench the Tm³⁺ population through radiationless energy transfer) and inducing simultaneous oscillation on the ³F₄ $\rightarrow$ ³H₆ transition at 1.8 μ m (Percival *et al.*, 1992).

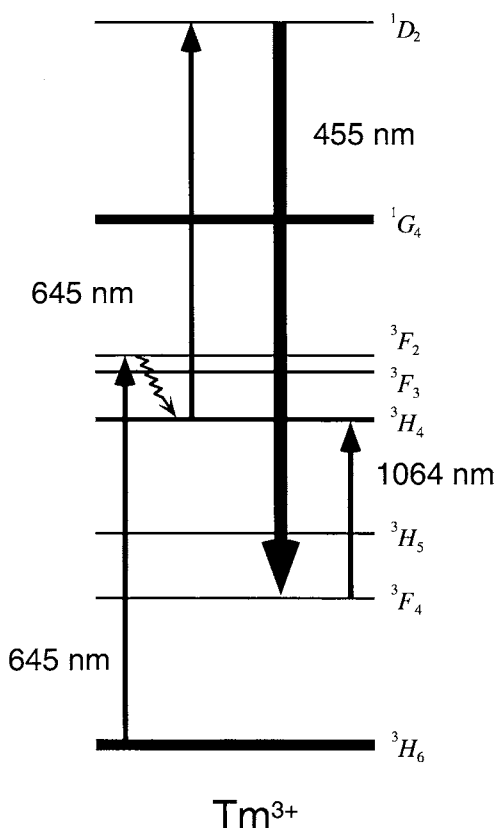


Figure 8.19: Continuous-wave 455-nm Tm^{3+} upconversion laser based on the indicated two-photon sequential pumping pathway. Addition of 1064-nm pumping helps deplete the lower laser level, thereby sustaining continuous oscillation in this otherwise self-terminating system (Le Flohic *et al.*, 1994).

8.3.2.2 $^1G_4 \rightarrow ^3H_6$ transition at 480 nm

One of the most influential publications on upconversion fiber lasers was authored by Steve Grubb and colleagues (Grubb *et al.*, 1992), then of the Amoco Technology Company. Using a cw Nd:YAG laser with nonstandard cavity mirrors designed to yield simultaneous output on the long wavelength 1112-, 1116-, and 1123-nm transitions, 57 mW of blue output at 480 nm was obtained on the $^1G_4 \rightarrow ^3H_6$ transition of Tm^{3+} in a ZBLAN fiber. The slope efficiency was 18% with respect to the launched pump power; Figure 8.20 depicts the three-photon sequential pump mechanism by which population inversion was created in this system.

This device was the first single-wavelength pumped cw upconversion laser operating in the blue at room temperature; that so much output power was obtained at such high efficiency quickly attracted the attention of numerous researchers. In

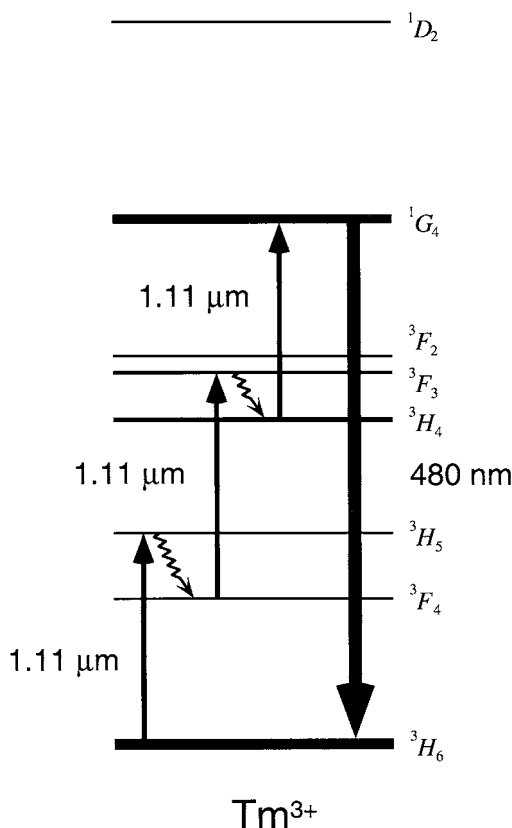


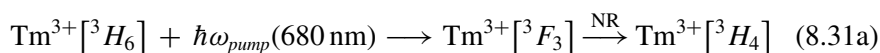
Figure 8.20: Blue upconversion laser based on single-wavelength, sequential three-photon pumping of Tm^{3+} (Grubb *et al.*, 1992).

the Amoco experiment, a 2-m length of NA 0.21 ZBLAN fiber doped at 0.1 wt% was formed into a laser cavity by butt-coupling of dielectric mirrors to the fiber ends (Figure 8.17(a)). The output coupler reflectivity was 90%, yielding a threshold launched pump power of 46 mW. However, only 45% of the pump power was absorbed by the fiber, leading Grubb *et al.* to extrapolate a slope efficiency of 32% with respect to the absorbed pump power. As a diode-pumped 1.12- μm Nd:YAG laser had already been demonstrated by the time of this experiment, clear prospects existed for the development of an all-solid-state blue upconversion laser operating at room temperature. Duclos and Urquhart (1995) have published a thorough rate-equation analysis of this experiment.

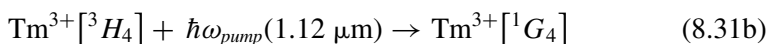
Following the basic design of the Amoco group, Tohmon *et al.* (1995) added as a second pump source a cw dye laser operating at $\sim 680\ \text{nm}$ in the hope of decreasing the pump threshold and increasing the slope efficiency. The motivation for the extra complication was the belief that a more compact device could be constructed if the bulky Nd:YAG laser – diode pumped or not – could be directly replaced by a 1.12- μm diode laser. On the other hand, high output power could not be expected from

diode lasers operating at this wavelength owing to materials constraints. Thus the second pump wavelength would help reduce the dependence on high performance from the infrared source; 0.68- μm diode lasers could eventually serve in this role.

The idea was to introduce a two-photon pathway to the upper laser level based on the ground-state transition



followed by the ESA transition (as shown in Figure 8.20)



In this way, the first two steps of the sequential three-photon infrared pathway were reinforced. Using a doped ZBLAN fiber nearly identical in design to the Amoco fiber, Tohmon *et al.* observed a factor of 4 increase in output power for a 680-nm input power of just 60 mW (Figure 8.21) for the same incident 1.12- μm pump

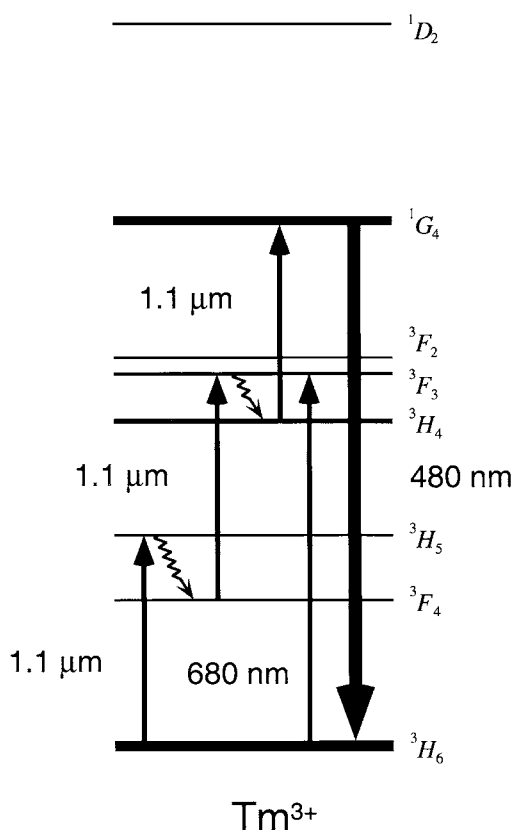


Figure 8.21: Blue upconversion laser based on single-wavelength, sequential three-photon pumping of Tm^{3+} reinforced by a two-photon pathway created through the addition of 680-nm pumping (Tohmon *et al.*, 1995).

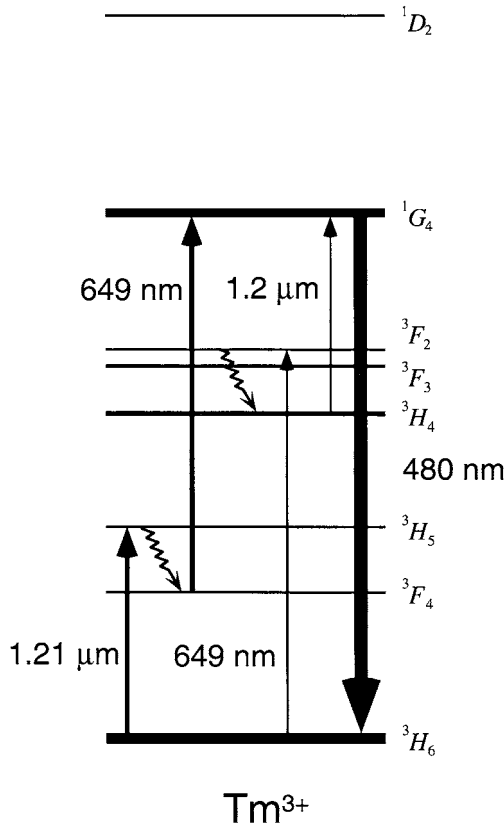


Figure 8.22: Blue upconversion laser based on two-wavelength, sequential two-photon pumping of Tm^{3+} . The alternative pathway, indicated by the thin arrows, also contributes to optical pumping (Tohmon *et al.*, 1997).

power. However, a commensurate reduction in the threshold 1.12- μm power was not observed owing to the onset of oscillation on the 0.8- μm $^3H_4 \rightarrow ^3H_6$ transition.

Two years following their initial work on two-wavelength pumping of $\text{Tm}:\text{ZBLAN}$, Tohmon *et al.* (1997) investigated a similar scheme that employed a 1.21- μm diode laser and a 649-nm dye laser, the latter laser being ultimately replaceable by a laser diode. The pumping scheme is shown in Figure 8.22. Note that unlike in their previous scheme, the short wavelength drives an ESA transition instead of a ground-state-absorption transition, and vice versa for the infrared wavelength. This approach helped avoid simultaneous oscillation at 0.8 μm ; the operating wavelength of the diode laser was thus chosen so as to optimize absorption on the $^3H_6 \rightarrow ^3H_5$ transition. Nevertheless, there existed enough overlap of the two pump wavelengths with the same transitions exploited in their previous work (thin arrows in Figure 8.22) that this latter pathway also contributed to the 480-nm gain. Using a 1.3-m fiber doped at 0.1 wt%, a maximum output power of 2.7 mW was

measured for 4% output coupling, 120 mW of 649-nm input power, and 30 mW of 1.21- μm input power. It is clear by inspection of Tohmon *et al.*'s results that total threshold pump powers of perhaps 20 mW were possible, while their extensive rate-equation analysis of the experiment predicted blue output powers of 20 mW from an optimized laser constrained only by available diode laser pump powers at the needed wavelengths.

The need, however, for multiple-wavelength pumping due to the low power of 1.1–1.2- μm diode lasers was greatly relaxed in a 1995 experiment performed at Spectra Diode Labs. Returning to single-wavelength pumping, truly excellent results were obtained by Sanders *et al.* (1995) in the first all-solid-state Tm:ZBLAN experiment. Using two polarization-combined single-emitter diode lasers each capable of 600-mW output in the range of 1100–1140 nm, 106 mW of blue output was obtained with a 2.5-m ZBLAN fiber doped at 0.1 wt%. The output-coupler reflectivity was 80%, which yielded a threshold incident pump power of 80 mW and a remarkable slope efficiency of 30% with respect to the as-launched pump power. As of 1995, this was the highest-power blue upconversion fiber laser yet demonstrated.

One issue not well addressed in ZBLAN fiber laser research is the role of fiber losses in determining threshold pump powers and output powers. This question appears to have been first investigated by Booth *et al.* (1996), who compared a 38-cm laser doped at 0.25 wt% with a 106-cm laser doped at 0.1 wt%. Using an as-launched pump power of 55 mW from a 1.135- μm diode laser, the shorter laser yielded 480-nm output power of 4.5 mW and a slope efficiency with respect to launched power of 13.8% whereas the longer laser yielded an output power of only 3 mW and a slope efficiency of 10.8%. These authors also demonstrated increased output power and slope efficiency with the addition of 1.22- μm pumping.

More than a doubling of blue output power over the previous maximum was demonstrated by Paschotta *et al.* (1997a) working at the University of Southampton. Following the suggestion of Grubb *et al.* (1992), this group used a diode-pumped Nd:YAG laser operating at 1123 nm in order to produce 230 mW at 481 nm from a 0.1-wt% Tm:ZBLAN fiber with a “modified” but unspecified composition. Using a 2.2-m length of this fiber and 27% output coupling, a slope efficiency of 19% was measured with respect to the incident pump power up to an output power of 120 mW. Unfortunately, this slope efficiency was not maintained at higher output powers as a result of a broadband absorptive loss in the blue that developed in the fiber at output powers greater than 100 mW. This loss, however, could be reversed by reducing the output power to below 100 mW for time periods on the order of an hour. The really bad news was that following high-power operation and allowing the laser sit idle overnight, oscillation could not be restarted even with a high reflectivity output coupler.

This “photodarkening” problem in Tm:ZBLAN has unfortunately proved to be the Achilles heel of an otherwise extraordinarily promising approach to all-solid-state generation of coherent light in the blue. In fact, several other reports of induced blue loss in Tm:ZBLAN had been previously reported and even studied in detail. (Barber *et al.*, 1995, Laperle *et al.*, 1995, Booth *et al.*, 1996, 1996a, Laperle *et al.*, 1997). For example, Barber *et al.* (1995) investigated three large-NA ZBLAN fibers doped at 0.1, 0.3, and 1 wt% Tm³⁺ by exposing them to low powers (<100 mW) of 1.14- μm laser light (no blue oscillation was permitted). These workers found that while no photodarkening occurred with the lightly doped sample, the two more heavily doped samples exhibited an induced absorption in the visible and infrared roughly commensurate in magnitude with the thulium concentration. The spectrum of this absorption is shown in Figure 8.23. At least at the low pump powers used in this work, the effect was reversible, and was further found to reverse more quickly at elevated temperature.

That same year, Laperle *et al.* (1995) reported a systematic investigation of infrared-induced photodarkening in Tm:ZBLAN fibers doped at levels in the range

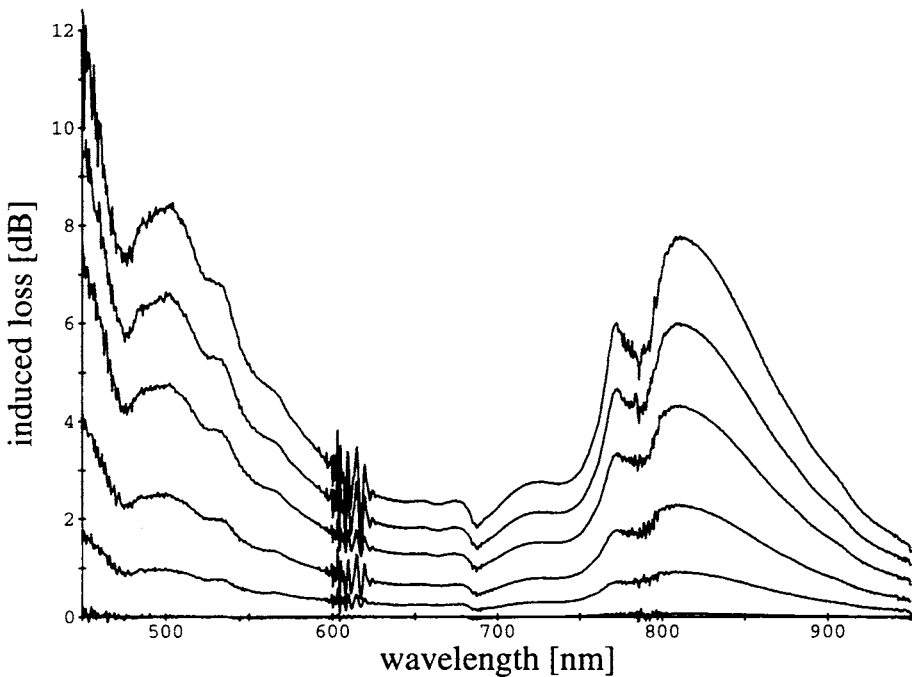


Figure 8.23: Absorption spectrum of infrared-induced color centers in Tm³⁺-doped ZBLAN. The various curves show the induced absorption for a 60-s exposure and various values of the 1.12- μm pump power ranging from 22 mW to 77 mW. After Barber *et al.* (1995), reprinted by permission.

of 0.05–1.17 wt%. Using pump powers at 1.12 μm up to 840 mW, these researchers observed an induced absorption spectrum similar to that seen by Barber *et al.* and further quantified the time development, pump-power dependence, and concentration dependence of the effect. In particular, the rate of photodarkening increased as the fourth power of the pump intensity and also increased dramatically with the thulium concentration. Laperle *et al.* attributed the darkening effect to the creation of color centers in the host glass resulting from ultraviolet fluorescence of Tm^{3+} ions on the $^1D_2 \rightarrow ^3H_6$ transition. Moreover, the precise composition of the fiber appeared to have a dramatic effect on the darkening rate: because small amounts of glass constituents such as lead or hafnium are often used as refractive index modifiers, fibers with different numerical apertures will possess varying amounts of these minority species. Thus Laperle *et al.* found that two fibers with identical thulium concentrations but different numerical apertures exhibited greatly differing darkening rates.

Analogous high-power measurements were performed a year later by Booth *et al.* (1996a) using fibers produced by the same manufacturer as Barber *et al.* and doped at 0.1, 0.12, and 0.25 wt%. Allowing the fibers to oscillate at 480 nm, similar induced-absorption spectra in the visible range were produced in all three fibers. Booth *et al.* also noticed a temporary bleaching effect for the induced absorption due to the presence of blue light. In a few hundred hours of dark time, however, the induced absorption would return. Measurement of the pump thresholds for lasers of varying concentration – and fiber lengths prepared in inverse proportion to the concentration) – revealed much greater increases in threshold power for more highly doped samples, thus implying that cooperative phenomena involving two or more thulium ions must be involved in the photodarkening process. This apparently led Booth *et al.* to likewise posit the creation of color centers in the glass resulting from ultraviolet fluorescence originating in high lying states of Tm^{3+} . Heating of the fiber to 100 °C was found to completely remove these color centers.

The photobleaching effect alluded to above has been further studied by Laperle *et al.* (1997) who argued that short-wavelength one-photon photoionization of the infrared-induced color center defects yielded trapped electrons and holes in the glass matrix that would then slowly recombine. These workers also argued that nonexponential and incomplete recovery of the color-center population following the photobleaching step implied the existence of at least two types of infrared induced defects. As of this writing, a thorough account of the photodarkening mechanisms at work in $\text{Tm}:\text{ZBLAN}$ has not been reported.

Returning to our discussion of upconversion experiments, Zellmer *et al.* (1997) have introduced a further variation of the $\text{Tm}:\text{ZBLAN}$ laser design in order to

construct an all-solid-state device. First, a 1065-nm pump source was constructed by diode pumping of a Nd^{3+} -doped double-clad silica fiber. Such double-clad fibers consist of a large-diameter multi-mode waveguide within which is embedded a single-mode waveguide containing the laser-active ions. In this way, pump radiation with poorer beam quality may be injected into and guided by the fiber yet still ultimately absorbed by the embedded single-mode core. Using a 12-W laser-diode bar as a pump source, Zellmer *et al.*'s Nd laser generated 3 W of output at 1065 nm with superb beam quality. However, in order to take fuller advantage of this high infrared pump power in a thulium upconversion laser, 0.5 wt% Yb^{3+} was included with 0.15 wt% Tm^{3+} in the ZBLAN fiber composition. Thus three steps of energy transfer from Yb^{3+} to Tm^{3+} analogous to the sequential absorption process shown in Figure 8.20 helped populate the upper laser level. Using a 1-m fiber with optimized output coupling of 30%, 106 mW of 482-nm output power was measured for a launched pump power of ~ 1.4 W. However, some saturation of the output power with respect to the input power was observed in this experiment; a slope efficiency of 6.6% was found below ~ 70 mW of output whereas a slope efficiency of 2.6 % was measured at higher outputs.

Excellent theoretical work has been performed in modeling of 480-nm Tm:ZBLAN upconversion lasers pumped in the 1.1–1.2- μm range. Aside from the work of Duclos and Urquhart (1995) mentioned earlier, Paschotta *et al.* (1997) have published a thorough rate-equation analysis and also discussed the role of energy transfer phenomena in thulium-doped fibers. Using realistic losses, a model concentration of 0.3 wt%, optimized output coupling and fiber length, and a numerical aperture of 0.3, Paschotta *et al.* predicted 102-mW of 480-nm output for a launched infrared power of 200 mW.

8.3.2.3 Other Tm^{3+} upconversion experiments

Although the bulk of attention given to Tm^{3+} -based upconversion lasers has focused on the 480-nm transition, a few ancillary experiments at other wavelengths deserve mention. Thrash and Johnson (1994) have used the Yb^{3+} – Tm^{3+} transfer idea in a study of crystalline BaY_2F_8 fibers prepared by the technique of laser-heated pedestal growth. The idea was to take simultaneous advantage of the large absorption and stimulated-emission cross-sections characteristic of crystalline host media and the waveguiding properties of optical fibers. Preliminary experiments with a 0.75-mm length of fiber 10–20 μm in diameter yielded cw oscillation at 649 nm at room temperature. Dennis *et al.* (1994) have demonstrated an 810-nm Tm:ZBLAN fiber laser upconversion pumped at 1064 nm by a Nd:YAG laser. Using a 35-cm fiber, output power of nearly 1.2 W was generated on the $^3H_4 \rightarrow ^3H_6$ transition at a

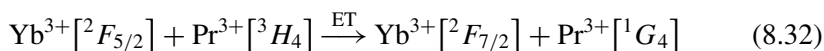
slope efficiency of 37% with respect to the launched pump power. An actively mode-locked and all-solid-state version of this laser was demonstrated two years later by Yang *et al.* (1996). At an output power of 130 mW, an 80-MHz mode-locked train of 380-ps pulses was produced in this experiment.

8.3.3 Pr³⁺ fiber lasers

The rare-earth ion that has received the greatest attention in the context of upconversion fiber lasers is praseodymium. Three simple reasons may be stated for this popularity: (1) The ion is able to produce red, orange, green, and blue output wavelengths from the same upper laser level. (2) The system may be pumped with a single wavelength. (3) ZBLAN fibers prepared with Pr³⁺ do not exhibit photodarkening effects such as those that plague thulium-doped fibers. However, reports of two-wavelength pumping experiments number nearly the same as single-wavelength experiments. As the number of pump wavelengths used in Pr³⁺-based upconversion lasers experiments is perhaps these devices' greatest distinguishing feature and since the same upconversion pathway applies to all output wavelengths, we depart in this section from the organization used up till now and separately discuss experiments using single- and two-wavelength pumping. Readers interested in particular Pr³⁺ output wavelengths may use Table 8.2 as an aid to finding relevant references.

8.3.3.1 Pr³⁺ fiber lasers pumped at one wavelength

Demonstrating yet again their talent for publishing upconversion “firsts”, Allain *et al.* (1991) reported an output power of 24 mW at 635 nm obtained from the high-gain $^3P_0 \rightarrow ^3F_2$ red transition of Pr³⁺ in an Yb³⁺-codoped ZBLAN fiber, the first upconversion fiber laser to use the Pr³⁺ ion. The optical pumping scheme is depicted in Figure 8.24 and proceeds as follows. First, Ti:S pumping at 849 nm, a wavelength that falls in the high-frequency wing of the Yb³⁺ $^2F_{7/2} \rightarrow ^2F_{5/2}$ transition, creates a population of excited donor ions. Although the Yb³⁺ ion is only weakly pumped, the large concentration of this species (2.0 wt%) yielded 40–50% absorption of the pump radiation over the ~1-m lengths of fiber used in this experiment. Next, radiationless energy transfer to Pr³⁺ occurred according to



and finally pump radiation drove ESA on the broad and closely spaced $^1G_4 \rightarrow ^3P_{0,1}, ^1I_6$ transitions. Thermalization among these three manifolds thus yielded substantial population in the $^3P_0$ and $^3P_1$ manifolds. With a Pr³⁺ concentration of

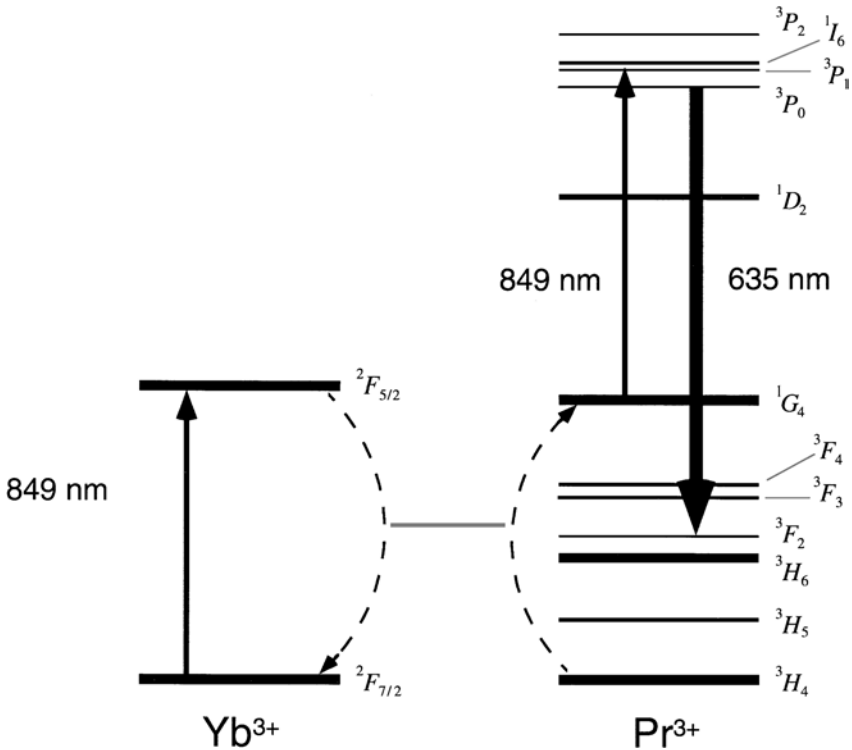


Figure 8.24: Single-wavelength-pumped red upconversion laser based on Pr^{3+} . Energy transfer from infrared pumped Yb^{3+} codopant ions is followed by excited-state absorption (Allain *et al.*, 1991).

0.1 wt%, however, the total absorption of pump radiation rose to over 90% as the pump power was increased, a result Allain *et al.* took to be a signature of the ESA step just described. In fact, the latter observation was a good indication that the ESA absorption cross-section was significantly larger than the 849-nm GSA cross-section of Yb^{3+} . Using output coupling of 40%, optimum output power was obtained with a 75-cm length of fiber, the shortest of three fibers investigated, thus indicating that intracavity losses compromised the performance of this device. Nevertheless, a slope efficiency of 10% with respect to the launched pump power was obtained. In a brief report of follow-up work performed with apparently the same fiber, Boj *et al.* (1995) reported 10 mW of output in the red from cavities possessing dielectric cavity mirrors deposited directly onto the fiber endfaces. Although lower output coupling was used in this experiment relative to the one just described, thus making direct comparison of the two experiments more difficult, it appears that fiber losses rather than loss at the end mirrors most affected the laser performance.

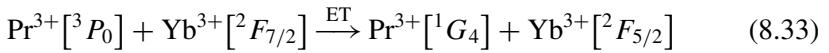
Invoking the same pump mechanism, analogous experiments performed by Baney *et al.* (1995) of the Hewlett-Packard Laboratories produced the usual red output as well as orange output on the $^3P_0 \rightarrow ^3H_6$ transition. These workers used a reduced Yb^{3+} concentration of 1 wt%, but also employed diode laser pumping, thus demonstrating all-solid-state operation. As in the work of Boj *et al.* just mentioned, dielectric mirrors were deposited directly onto the ends of doped ZBLAN fibers ~ 1 m in length. However, perhaps the most important difference in this more recent work was the use of an NA-0.39 fiber, which likely helped increase the gain and reduce the threshold pump power. With 96% output coupling, 3.9 mW of 635-nm output power was measured for 69 mW of 860-nm pump power, while at 10% output coupling ~ 1.8 mW was obtained. At the 602-nm orange output wavelength, 200 μW of output was obtained at an unspecified output coupling.

A year later, the Hewlett-Packard group (Baney *et al.*, 1996a) demonstrated simultaneous output in the green ($^3P_1 \rightarrow ^3H_5$ transition at 520 nm) and in the blue ($^3P_0 \rightarrow ^3H_6$ transition at 490 nm) using the same fiber composition as in their earlier work. Depositing as before dielectric mirrors directly onto the fiber endfaces, ~ 700 μW of output power was obtained at each of the two wavelengths. Although the two output transitions ostensibly originate in different upper laser levels, because these levels are thermally coupled to one another, something of a balance must be struck between the relative gain and loss at the two wavelengths. Thus output coupling in the blue was set to 10% while output coupling in the green was set to 20%. Even so, the threshold pump power at 520 nm was significantly lower than at 490 nm, namely 55 mW versus 87 mW launched power as obtained from a tunable laser-diode-based master-oscillator-power-amplifier system operating at 856 nm. The authors state in their conclusions that simultaneous blue, green, and red output might be obtained from a Pr,Yb:ZBLAN fiber device with the help of carefully designed cavity mirrors.

Likewise employing a Pr,Yb:ZBLAN fiber, dramatically larger red, orange, green, and blue output powers were demonstrated in a single-wavelength pumping experiment (860-nm) performed at Los Alamos National Laboratory by Xie and Gosnell (1995). In particular, 300 mW of red output was obtained from a 60-cm length of fiber that for optical feedback relied solely upon the 4% Fresnel reflection from the cleaved fiber end. Just as remarkable, the red slope efficiency with respect to the launched pump power in this laser was 52%. Both of these figures were the largest ever reported for a room-temperature visible upconversion laser up to that time. Maximum output powers of 44 mW, 20 mW, and 4 mW were obtained for cavities individually designed for the orange, green, and blue wavelengths, respectively; these results were obtained with fiber lengths and output couplings of 60 cm and 5% at 617 nm, 42 cm and 3% at 520 nm, and 26 cm and 3% at 491 nm. As the blue laser transition terminated on the ground state, the short

fiber length chosen for the blue laser helped minimize the round-trip intracavity loss. Another important result demonstrated in this work was a large pump tuning range of 780–880 nm, a band easily matched by high-power diode lasers currently available.

That these experiments yielded such a large enhancement of performance compared with those of Allain *et al.* and Baney *et al.* was most likely a consequence of larger dopant concentrations. Specifically, a Yb^{3+} concentration of 3 wt% and a Pr^{3+} concentration of 0.2 wt% were used. At the same time, Xie and Gosnell surmised that at these increased concentrations a new upconversion pathway became available to the system, namely a photon-avalanche process involving cross-relaxation between Pr^{3+} ions in the upper laser level and ground state Yb^{3+} ions:



At this point the energy-transfer process Equation (8.32) stepped in to yield a second Pr^{3+} ion in the $^1G_4$ state, after which strong ESA on the $\text{Pr}^{3+} \ ^1G_4 \rightarrow \ ^3P_{0,1}, \ ^1I_6$ transitions populated the upper laser levels just as before. An energy-level diagram that summarizes this upconversion pumping mechanism is shown in Figure 8.25.

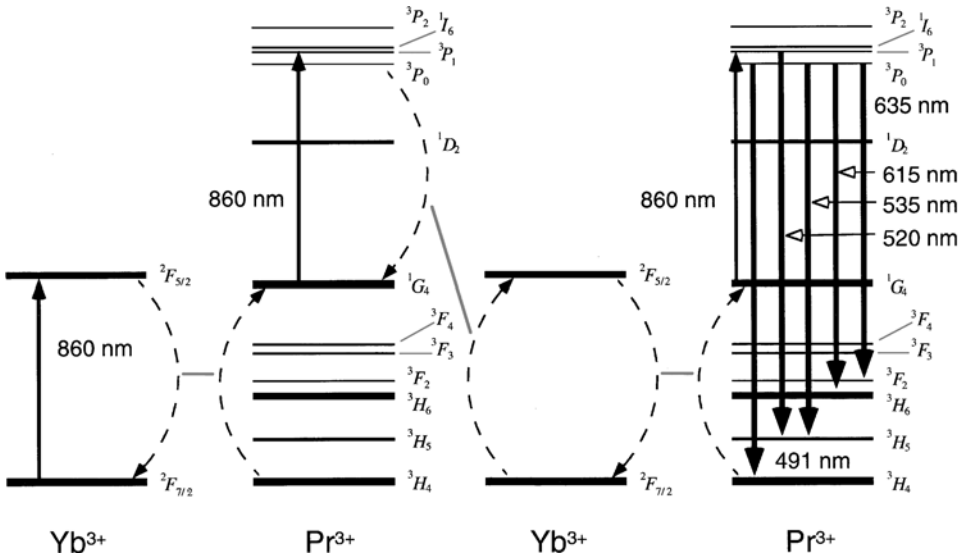


Figure 8.25: Single-wavelength photon-avalanche pump mechanism for multi-wavelength upconversion output from Pr^{3+} . A seed population in the upper laser levels is created by the energy-transfer and ESA steps shown in the leftmost pair of diagrams. The cross-relaxation step shown in the middle pair of diagrams enables a second Yb^{3+} – Pr^{3+} energy transfer, as shown in rightmost pair of diagrams. ESA by the two Pr^{3+} ions in the $^1G_4$ state completes the avalanche cycle (Xie and Gosnell, 1995).

Using dopant concentrations identical to those used by Xie and Gosnell, Sandrock *et al.* (1997) more than tripled the 635-nm output power obtained by the former workers by focusing the output of two high-power cw Ti:S lasers into the core of a 51-cm fiber. With optical feedback provided by a single 635-nm high reflector butted against one of the fiber ends and 4% Fresnel reflection from the opposite end, a maximum output power of 1.02 W in the red was obtained with a pair of Ti:S pump lasers tuned to 826 and 852 nm. This output power is the highest yet reported for a cw visible upconversion laser. The combined incident pump power was 5.51 W. The use of two different pump wavelengths in this experiment evidently helped to avoid optical saturation of the Pr^{3+} ESA transition insofar as different terminal states were accessed. A slope efficiency with respect to the incident pump power of 19% was measured and no saturation of the output power was observed all the way up to the maximum applied pump power. Sandrock *et al.* also described a photon-avalanche pump mechanism identical in its essentials to that described two years earlier by Xie and Gosnell and further offered experimental evidence for the effect.

In a variation of this experiment, Zellmer *et al.* (1998) designed a double-clad fiber for the purpose of demonstrating high output power at 635 nm in a system pumped by a low brightness source. The motivation for this experiment was the recognition that high-power all-solid-state upconversion lasers would require as pump sources high-power diode laser arrays with less than ideal beam quality. The fiber used in this experiment therefore possessed a doped single-mode waveguide with an NA of 0.15 embedded within an undoped 25- μm outer waveguide of NA 0.35. In this way, very high pump powers of reduced intensity can be injected and guided by the multi-mode outer waveguide yet ultimately absorbed by the single-mode core over a sufficient length of the fiber. Such a design has the additional benefit of reducing the risk of optical damage of the fiber input face. But a critical fact to be kept in mind with such an approach is that given the nonlinear character of the upconversion process – especially in the case of photon-avalanche pumping – one must be sure to maintain high pump intensity in the doped inner core.

Creating a surrogate diode laser by mode scrambling of a Ti:S laser with a multi-mode silica fiber, Zellmer *et al.* injected up to 3 W of 840-nm pump light into the multi-mode core of their double-clad fiber. With a fiber length of 3 m, dopant concentrations of 0.2 wt% Pr^{3+} and 3 wt% Yb^{3+} , and output coupling of 30%, 440 mW of output power was obtained at 635 nm. The measured slope efficiency was 17% with respect to the launched pump power, but since only 30% of the pump power was absorbed by the core, a slope efficiency of 56% is implied with respect to the absorbed power. This experiment was the first to exploit double cladding in an upconversion fiber laser and represents the current state of the art for single-wavelength pumping of Pr^{3+} -based upconversion systems.

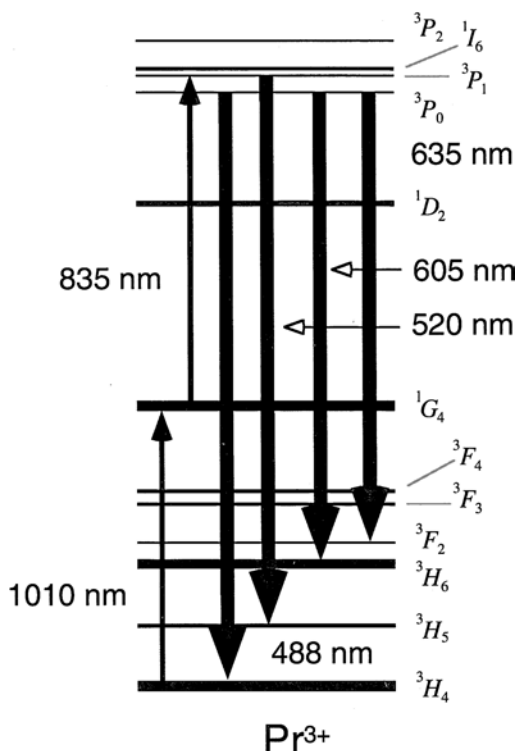


Figure 8.27: Dual-wavelength sequential two-photon pump mechanism for multi-wavelength output from a Pr^{3+} upconversion laser (Smart *et al.*, 1991).

scheme: Using two Ti:S lasers tuned to 1010 and 835 nm, these workers demonstrated cw output at 635 nm, 605 nm, 520 nm, and 488 nm in a ZBLAN fiber doped at 0.056 wt% Pr^{3+} . At the red wavelength, a 10-m length of fiber with a single high-reflectivity end mirror formed the optical cavity – as with several of the Pr^{3+} red fiber lasers already discussed, Fresnel reflection from the opposite fiber end served as the output coupler. The long fiber length was motivated in part by the small absorption cross-section of the $^3H_4 \rightarrow ^1G_4$ ground-state-absorption transition. Thus with 2 W of incident pump power at 1010 nm and 700 mW of incident pump power at 835 nm, a total output of 185 mW was obtained. Given the 30–40% coupling efficiency of the pump radiation, these figures correspond to an overall efficiency of $\sim 20\%$. However, significantly improved results are expected given the 0.3 dB/m background fiber loss measured in this early experiment. Switching to a shorter fiber in order to help reduce background losses, a 605-nm laser was constructed using a 1.2-m fiber and output coupling of 60%. Setting the 1010-nm pump power to 1 W and varying the 835-nm pump power between 0 and 400 mW, a threshold pump power of ~ 30 mW and a maximum orange output power of 30 mW was measured. At low pump powers, the slope efficiency with respect to the 835-nm power was 7%,

but above ~ 250 mW noticeable saturation of the output occurred apparently as a result of optical saturation of the $^1G_4 \rightarrow ^3P_{0,1}$, $^1I_6$ ESA transitions. At the blue and green wavelengths, pairs of 99% reflectivity mirrors selective for one wavelength or the other were butted to the fiber ends in order to form the optical cavity. Approximately 1 mW of output power was measured at each wavelength; total threshold pump powers of several hundred milliwatts were measured in both cases.

One might well ask which of single- or two-wavelength pumping yields superior performance in a Pr:ZBLAN upconversion laser. A partial answer to this question appears in an interesting paper published by Piehler *et al.* (1993). Using a ZBLAN fiber doped at 0.2 wt% Pr^{3+} and 0.4 wt% Yb^{3+} , a Ti:S laser was used in conjunction with an 833-nm diode laser to measure 521-nm output power as a function of the Ti:S wavelength. In other words, by tuning the Ti:S laser over the range of 900–1020 nm, a comparison could be made between indirect pumping of the $^1G_4$ level (via energy transfer from Yb^{3+}) or direct pumping of this state via ground-state absorption. (Note, however, that at the longer wavelengths good overlap still existed with the Yb^{3+} absorption spectrum; at 1015 nm for example, the absorption coefficients for the two species are about equal.) At fixed Ti:S pump power, essentially identical output powers were measured at Ti:S wavelengths of 975 nm and 1015 nm, the former wavelength being coincident with the peak Yb^{3+} absorption cross-section for the $^2F_{7/2} \rightarrow ^2F_{5/2}$ transition and the latter wavelength being coincident with the peak $^3H_4 \rightarrow ^1G_4$ Pr^{3+} cross-section. These measurements were made with a fiber length of 70 cm, a distance over which absorption by Yb^{3+} at 975 nm would have been nearly 100% but over which incomplete absorption by Pr^{3+} at 1015 nm would have occurred. In short, a shorter length of fiber pumped at 975 and 833 nm might have yielded better results in light of reduced fiber losses. It is also possible that improved performance in a codoped fiber would result from increased Yb^{3+} concentration. Clearly at the Yb^{3+} concentration used in this experiment, photon-avalanche pumping did not occur.

However, in substituting a 985-nm or 1016-nm laser diode for the Ti:S laser, best results were obtained for the longer pump wavelength despite lower pump power. This no doubt resulted from the reduced Yb^{3+} absorption at 985 nm compared with 975 nm. Piehler *et al.* quoted a maximum 521-nm output of 0.7 mW for 112 mW of pump power from an 833-nm laser diode combined with 80 mW of pump power from a 1016-nm laser diode. The fiber length was 1 m while identical output and end mirrors were used; at the signal wavelength, the reflectivity of these butt-coupled mirrors was 98%. A maximum output power of 6.2 mW was obtained at 635 nm under the same conditions, except that an output coupler of 77% reflectivity was used that also reflected 97% of the unabsorbed pump light back into the fiber.

Returning to investigations of singly-doped Pr:ZBLAN fiber lasers, Zhao and Poole (1994) working at the University of Sidney have focused attention on

the $^3P_0 \rightarrow ^3H_4$ blue laser transition. Using two Ti:S lasers tuned to 835 and 1017 nm, 9 mW of 492-nm output power was measured from an 85-cm fiber with 10% output coupling. The Pr^{3+} concentration in this NA-0.21 fiber was 0.05 wt%. A mere 65 mW of launched pump power at 835-nm and only 100 mW of launched 1017-nm power was used to obtain this result.

More refined results obtained by this group were reported the following year (Zhao *et al.*, 1995). Using a 78-cm NA-0.24 fiber and output coupling of 30%, 22 mW of blue output power was obtained at the same concentration as used in their previous work. In this case, just 42 mW of launched pump power at 835 nm was used; the threshold 1017-nm launched pump power at this 835-nm power was only ~ 35 mW. These pump powers represented a 20% reduction in total threshold pump power over that of Zhao and Poole (1994) even with the higher output coupling. A numerical model of the upconversion laser performance agreed well with these results; in particular, of central importance in the laser design was the question of loss at the signal wavelength resulting from ground-state absorption. The choice of fiber length thus represented the optimum compromise between pump absorption and signal ground-state absorption. However, only $\sim 50\%$ of the pump power was absorbed by the optimum-length fiber. Given that the output coupler in this experiment was only weakly reflecting at the pump wavelengths, higher output power and lower threshold pump power could be expected for an output coupler of high reflectivity at the pump wavelengths. This expectation was later realized in numerical simulations and confirming experiments performed by Zhao (1995). Specifically, Zhao demonstrated that a 40% reduction in threshold and a doubling of output power could be obtained by back reflection of the residual pump radiation.

Collecting all of their previous advances together, the Sidney group (Zhao and Fleming, 1996) next built an all-solid-state 492-nm Pr:ZBLAN upconversion laser pumped by a pair of semiconductor diode lasers possessing silica-fiber pigtailed. The use of bulk focusing optics was avoided in this experiment by fusion splicing the pump-laser pigtailed to a wavelength-division multiplexer in order to combine the two pump beams. The output end of the fiber multiplexer was then butt-coupled to the ZBLAN fiber, which possessed dielectric cavity mirrors deposited directly onto the fiber endfaces. For this experiment, an output power in the blue of 1.2 mW was obtained from a 68.5-cm fiber of numerical aperture 0.39, doping level of 0.1 wt%, and output coupling of 10% (this mirror coating was designed to give high reflectivity at the pump wavelengths). The pump powers were 45 mW at 1017 nm and 28 mW at 835; the total threshold pump power was just 40 mW. Anticipating the development of single-emitter laser diodes producing higher output powers at the needed wavelengths, Zhao and Fleming predicted the possibility of 10 mW blue power from an all-solid-state system.

Similar performance in the blue was reported just a few months later by Baney *et al.* (1996), likewise using as pump sources a pair of laser diodes. With pump output powers of 200 mW at 831 nm and 100 mW at 1017 nm, 1.2 mW of 492-nm output power was produced from a fiber apparently identical in design to that used in the experiment of Zhao and Fleming (1996). In their case, however, Baney *et al.* used a fiber length of 48 cm and output coupling of 7%; the cavity mirrors were deposited directly onto the fiber endfaces. No special effort was made to back reflect the pump beams, although a 35% reflection of the 835-nm pump light was obtained off the output coupler. Baney *et al.* took a special interest in the spectral and noise properties of the blue laser output, finding three sharp features in the output spectrum spanning ~ 1 nm. Measuring the noise spectrum of the blue output with a fast photodiode, a peak at 286 kHz was found that corresponded to the frequency of relaxation oscillations in the laser cavity as induced by environmental fluctuations.

Applying yet another twist to the problem of two-wavelength pumping of a Pr:ZBLAN fiber, Pask *et al.* (1997) used 840-nm Ti:S pumping of an Yb:silica-fiber laser operating at 1020 nm. In addition to the latter wavelength, the output of this fiber laser consisted of unabsorbed 840-nm pump light. Thus imaging of the Yb:silica output onto the input face of a Pr:ZBLAN fiber would two-photon pump the system. Various lengths of the Yb laser yielded different ratios of 840- versus 1020-nm light; optimum pumping of Pr^{3+} was obtained with a 6-m Yb laser for which output consisted of 115 mW of 1020-nm light and 180 mW of unabsorbed 840-nm light. In turn, various lengths of the ZBLAN fiber, doped at 0.048 wt% Pr^{3+} , and various butt-coupled cavity mirrors selective for red, green, and blue output wavelengths were investigated. At 635 nm, a 5.5-m cavity with the now familiar Fresnel output coupling yielded 54 mW of output power at a slope efficiency of 23% with respect to the incident laser power. Total threshold pump power was 120 mW, a figure that increased to 160 mW in a 9-m Pr:ZBLAN laser cavity of otherwise equivalent design. The latter observation indicated that fiber losses significantly larger than the theoretical minimum for the ZBLAN glass composition still affected the performance of upconversion laser devices even as late as 1997; in this experiment, the measured loss was 0.03 dB/m at 635 nm. Nevertheless, a factor of 10 decrease in background loss had been obtained compared with the fiber used by this same group in 1991.

At the output wavelength of 520 nm, best performance was obtained with 8% output coupling and a fiber of comparable length to that used in the 635-nm laser. Specifically, 18 mW of green output was measured at a slope efficiency of 8.5%. In this experiment, background fiber loss at the signal wavelength was measured to be ~ 0.5 dB/m, which significantly affected the performance at this wavelength. At

491 nm, loss due to ground-state absorption constrained the fiber to a more modest 2-m length; output coupling of 6% yielded a respectable 7 mW of blue output.

Owing to the single pathway, two-photon pumping mechanism and the apparent absence of energy transfer mechanisms either contributing to or detracting from the population of the upper laser level, theoretical studies of the Pr:ZBLAN up-conversion fiber laser have been successfully performed. In particular, the reader is referred to the work of Tropper *et al.* (1994), Zhao (1997), and Zhao and Fleming (1997, 1998). The last paper should be of particular interest to those interested in the affect of numerical aperture on the performance of upconversion fiber lasers.

8.3.4 Ho³⁺ fiber lasers, $^5S_2 \rightarrow ^5I_8$ transition at ~550 nm

As described in the introduction to this chapter, the first room-temperature upconversion fiber laser – and indeed one of the first room-temperature upconversion lasers of any format – was based on the Ho³⁺-doped ZBLAN system investigated by Allain *et al.* (1990a). The double-pathway, doubly-resonant, two-photon pumping mechanism used in this experiment is depicted in Figure 8.7. Employing a 1-m length of fiber doped at 0.12 wt% Ho³⁺, an output power of 10 mW was produced following optical pumping with a krypton-ion laser running on the 647.1-nm line, a wavelength available from commercial semiconductor diode lasers. The maximum incident pump power was 300 mW; however, owing to only 30% coupling of the pump laser light into the fiber core, a slope efficiency of 20% with respect to the launched power was quoted by the authors. Adding an intracavity dispersive prism (Figure 8.17(b)), Allain *et al.* demonstrated tunable operation of their Ho:ZBLAN laser over the full range of 540–553 nm. Two years after this ground-breaking experiment, Atkins *et al.* (1992) developed a rate-equation model of the Ho:ZBLAN upconversion fiber laser and showed good success modeling the performance data of Allain *et al.*

Since 1993, the torch of Ho³⁺-based upconversion laser development has been carried exclusively by D. S. Funk, J. G. Eden, and colleagues at the University of Illinois. In this group's first effort (Funk *et al.*, 1993), 12.4 mW of output at 549 nm was measured for a 22-cm fiber doped at 0.1 wt% and pumped with a tunable dye laser at 646.9 nm. This pump wavelength was selected as optimum following investigation of the laser and fluorescence excitation spectra obtained by tuning of the dye laser. In particular, these spectra largely exhibited the Stark-level structure of the $^5I_8 \rightarrow ^5F_5$ ground-state-absorption transition, which the authors believed to demonstrate that ESA originating in the upper laser level had minimal impact on the laser performance; this despite the observation of fluorescence features originating in higher lying states. The authors also found that output coupling of 1%, 3%, and

5% yielded nearly identical threshold pump powers but increasing slope efficiency with increased coupling; there was no sign of output power saturation up to the full incident pump power of 330 mW. With respect to the launched pump power, a maximum slope efficiency of 14% was determined. However, the fiber used in these experiments possessed an unusually large core diameter of 11 μm and thus operated in multiple transverse modes at both the pump and output wavelengths. Improved performance would be expected for a fiber allowing single-transverse-mode propagation at one or both of the two wavelengths.

In further work with same fiber design (Funk *et al.*, 1994a, 1996), the Illinois group investigated more systematically the effect of output coupling and fiber length on the output characteristics. In fact, a maximum output of 38 mW was measured for 270 mW of pump power absorbed by a 21-cm fiber at the maximum employed output coupling of 30%; in this optimum case, the slope efficiency with respect to the absorbed pump power was 24%. As in their previous work, there was no sign of output saturation with increasing pump power, although for larger values of the output coupling (7.5%, 12%, and 30%) the typical systematic increase in threshold absorbed pump power (up to ~ 140 mW at 30%) was observed. Switching to an 86-cm fiber, higher thresholds and lower slope efficiencies were measured, evidently as a result of increased round-trip loss in the fiber. However, a new feature not firmly demonstrated in any previous work with Ho:ZBLAN was the observation of self-pulsing of the output. Reminiscent of the green output behavior of bulk Er^{3+} upconversion lasers, 1–2- μs pulses at a repetition rate of ~ 100 kHz occurred in ~ 5 -ms bursts with a regular separation between bursts of ~ 10 ms. Further, the pulsing frequency increased as the pump power increased consistent with the time required for population build up in the upper laser level. The authors thus concluded that self-Q-switching occurred resulting from saturated absorption of ground-state absorption on the inverse laser transition $^5I_8 \rightarrow ^5S_2$.

In their 1997 publication on the Ho:ZBLAN fiber laser, the Illinois group switched to a large-NA single-mode fiber and 30-mW diode-laser pumping at 643 nm (Funk *et al.*, 1997). The principal objective in this work was to demonstrate as low a pump threshold as possible given the relatively low output power of the pump source. Again using a doping level of 0.1 wt% and a 22-cm fiber length, a maximum output power of 1.2 mW in the green was measured at an incident pump power of 24 mW and 24% output coupling. The threshold launched pump power was a remarkable 3.5 mW, and fell to just 1.9 mW with 1.6% output coupling, both figures the lowest ever reported for an upconversion laser. Unlike previous findings, however, true cw oscillation occurred, at least if the incident pump power were held well above threshold. In concluding remarks, the authors justifiably state that this all-solid-state device is a viable substitute for the 2-mW 543-nm HeNe laser.

In later work, the Illinois group used the same pumping mechanism and fiber design in order to construct a 412-nm violet laser operating on the $\text{Nd}^{3+2} P_{3/2} \rightarrow {}^4 I_{11/2}$ transition. Population of the upper laser level evidently resulted from nonradiative relaxation of the laser-pumped ${}^4 D_{3/2}$ level, as shown in Figure 8.28. In this experiment, a 39-cm length of fiber with output coupling of 0.45 yielded 470 μW of output power for 320 mW of 590-nm incident pump power. The slope efficiency of this device was 0.5% with respect to the incident pump power and by no means had the output power been optimized with respect to output coupling. Contrary to the results obtained at 381 nm, upon onset of oscillation true cw output was obtained after an $\sim 200\text{-}\mu\text{s}$ initial period of severe relaxation oscillations or perhaps self-Q-switching. Even so, rapid fluctuations $\sim 50\%$ in amplitude were observed in the output after settling of the initial transients. Despite the short output wavelengths of study in the two publications just discussed, it is interesting that no mention is made of photodarkening phenomena analogous to that observed for Tm-doped ZBLAN, this despite the near-equal energies of the Nd^{3+} and Tm^{3+} excited states that may lead to photodarkening.

8.4 PROSPECTS

At the time of this writing, the center of attention in upconversion laser research remains with doped ZBLAN fibers. The great advantage that a guided wave offers in terms of high pump intensities and the resulting large gain coefficients is the primary factor that has made demonstrations of room-temperature upconversion lasers commonplace. Indeed, in only one of the experiments discussed in the previous section was the laser fiber subject to cooling of any kind. One can thus easily predict that much in the way of evolutionary advances remains to be accomplished so that upconversion lasers may be pushed closer to the marketplace. The purpose of this section, however, is to report on an incomplete sampling of the upconversion literature that may indicate more long-range and speculative directions for the field.

Perhaps of most immediate concern are the prospects for developing upconversion lasers based on new rare-earth species. In this regard, room-temperature near-infrared operation of a bulk $\text{Ho}^{3+}/\text{Yb}^{3+}$ laser upconversion pumped with a Ti:S laser has been demonstrated (Möbert *et al.*, 1998). To our knowledge this is the first example of a bulk holmium-based upconversion laser; studies of visible upconversion fluorescence from bulk room-temperature Ho^{3+} (Mosses *et al.*, 1998, Zhang *et al.*, 1998) suggest that the production of visible laser output at room temperature is imminent. Another possible new upconversion system has been investigated by Gharavi and McPherson (1994). In spectroscopic measurements, these workers observed ultraviolet emission from Gd^{3+} ions in a low-phonon crystal codoped

with Er^{3+} . In particular, long-lived fluorescence at 281 and 313 nm was observed following 552-nm pumping of the $^4S_{3/2}$ state of Er^{3+} . Insofar as strong green emission originating in this state is possible with upconversion pumping at infrared wavelengths, there exists the possibility of demonstrating an infrared-pumped hard ultraviolet laser. The possibility of a Tb^{3+} upconversion laser has been investigated by Noginov *et al.* (1996). Terbium fluorescence lines ranging in wavelength from 635 to 375 nm were observed in an Yb-codoped bulk garnet crystal at room temperature. With primarily an interest in visible phosphor applications, a wide range of Yb-codoped rare-earth crystals has been studied by Page *et al.* (1998).

New kinds of host media are also under investigation. What appears to be of keenest interest are fluoroindate glasses, namely, glass compositions analogous to ZBLAN such as $38\text{InF}_3\text{-}19\text{BaF}_2\text{-}19\text{ZnF}_2\text{-}10\text{PbF}_2\text{-}10\text{-SrF}_2\text{-}2\text{AlF}_3\text{-}2\text{GdF}_3$. The idea here is that owing to the reduced maximum phonon frequencies in these types of materials, nonradiative relaxation rates are smaller than in fluorozirconate glasses so that intermediate states in the upconversion pumping chain are more effective in storing energy. Strong upconversion fluorescence at ultraviolet, blue, and green wavelengths has been observed in neodymium-, thulium-, and erbium-doped fluoroindate glasses (Catunda *et al.*, 1996, de Araújo *et al.*, 1996, Kishimoto and Hirao, 1996, Maciel *et al.*, 1997, Menezes *et al.*, 1997, 1997a). Other interesting compositions include fluoroaluminozirconate glasses (Shikida *et al.*, 1994), which have yielded – as compared with fluorozirconate glasses – increased green upconversion luminescence in Ho,Yb codoped material and fluoroaluminate glasses (Tanabe *et al.*, 1993). All of these materials are in principle amenable to fabrication in the form of optical fibers. An even more exotic idea has been investigated by Koeppen *et al.* (1996), who observed intense infrared-pumped green upconversion emission from dissolved Er^{3+} ions in a liquid solution formed of POCl_3 and SnCl_4 . Numerical analysis based on their fluorescence measurements led these workers to predict a 45-mW threshold pump power for an upconversion laser comprising a hollow-core fiber filled with this solution.

Yet another variation on upconversion laser design attempts to marry the benefits of bulk crystalline lasers, namely long lifetimes and large absorption and stimulated emission cross-sections, with those of glass optical fibers. The basic approach is to deposit rare-earth-doped thin films onto suitable substrates after which waveguide structures are etched into the films. For example, in a LaF_3 film deposited onto CaF_2 by molecular beam epitaxy, Er^{3+} upconversion luminescence at 538 nm has been observed (Uda *et al.*, 1997). Along similar lines, liquid-phase epitaxy has been used to prepare thin films of Pr^{3+} -doped gadolinium-gallium-garnet. Low-temperature upconversion luminescence spectra exhibited unusually strong emission in the blue. A completely different and especially inexpensive approach to the upconversion problem is to use a polymer host medium. This concept has been developed by

Brodin *et al.* (1995), who spin coated poly(ethylene oxide) containing ErF_3 onto a steel plate. Using a 650-nm dye laser as a pump source, these workers observed strong green and weak blue upconversion fluorescence from this sample.

Departing entirely from the use of rare-earth ions, G. S. He and colleagues at SUNY Buffalo have investigated two-photon upconversion in various dye-doped materials and actually demonstrated upconversion lasers based on their results (He *et al.*, 1996, 1996a, 1998). However, a fundamental difference exists in this approach in that rather than sequential two-photon pumping through a resonant intermediate level the upper laser level is populated by a two-photon nonlinear transition. Typical of their work is a 1996 paper in which a polymer fiber is doped with a novel dye compound selected for its high two-photon absorption cross-section. Upon pulsed pumping with a Nd:YAG laser, 1.25 μJ of output is produced at 610 nm with ~ 0.88 mJ of 1060-nm pump radiation.

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Part 3

Blue-green semiconductor lasers

9

Introduction to blue-green semiconductor lasers

9.1 OVERVIEW

From its inception in 1962, the semiconductor laser based on GaAs and related III–V compounds has evolved into an extraordinary and versatile optoelectronic device. Today we find this ubiquitous, relatively low-cost, high-efficiency coherent light source used in a wide spectrum of applications ranging from long-haul optical communications systems to high-density optical data storage. The evolution of these devices to their present state of sophistication – with wall-plug efficiencies of 50% and greater in the conversion of DC electric current to coherent photons (as one illustrative characteristic) – did not happen overnight, however. Nearly two decades of research and development were spent before the edge-emitting GaAs-based diode laser emerged as a mature optoelectronic technology suited for applications and integration. Once the basic guiding scientific principles were established in the design of these devices, making specific use of optical and electronic confinement in heterostructures, a major challenge remained at the basic materials science level. This challenge concerned the role of crystal defects, both point and extended-state in nature, which were found to lead to degradation and early burnout of the lasers. However, advances in epitaxial techniques in the III–V arsenide and phosphide systems, together with improvements in the device fabrication techniques, led to the development of low threshold current density quantum well (QW) heterostructure lasers, which are now contributing to the revolution in optical network technology.

The history of semiconductor light emitters at the short visible wavelengths stands in sharp contrast. To be sure, wide bandgap semiconductors had been recognized as prime candidates for efficient compact blue-green light sources in the early 1960s. At that time, fundamental optical studies exposed a rich phenomenology of light–matter interaction near the bandgap in semiconductors, especially in terms of elementary excitations such as excitons and polaritons. Yet approximately three decades passed before the early promise of blue-green LEDs and diode lasers finally

began to see a technical fulfillment in the early 1990s. New epitaxial techniques provided the capability for the synthesis of wide-bandgap QW heterostructures and also played a profound role in cracking problems of doping in bulk ZnSe- and GaN-based semiconductors, which had frustrated researchers in many laboratories for a long time.

While the first diode laser demonstrations in the blue-green were made in ZnSe-based QW heterostructures in 1991 (Haase *et al.*, 1991, Jeon *et al.*, 1991), a rapid sequence of developments in GaN compounds brought LEDs based on these materials to technological and commercial viability in the mid-1990s (Nakamura *et al.*, 1993). Since the first pulsed violet InGaN QW diode lasers were demonstrated by Nakamura *et al.* (1996) and Akasaki *et al.* (1996), this laser technology has made significant strides, especially at the Nichia Chemical Company, and commercially-available devices have begun to appear. As one consequence, it now appears likely that the 405–415-nm InGaN laser will form the basis of the next generation of optical disk technology, which will supersede the current DVD format. There have been feasibility demonstrations using the 405-nm laser in a rewritable dual-layer phase-change standard 12-cm optical disk, corresponding to a capacity of 27 Gbytes per side with a user data transfer rate of 33 Mbps (Akiyama *et al.*, 2001). This compares with approximately 5 Gbytes per side for the present DVD using a 630-nm red laser. The 405–414-nm wavelength range is best suited to achieving high performance in the InGaN QW system for reasons of optical gain as described later. It appears difficult to reach the longer blue and green wavelengths with this system, at least for now.

The success of the nitride devices, including advances in the development of high-power microwave field-effect transistors (FETs), has been a major surprise, because the underlying heterostructure material has been typically “loaded” with defects. This would normally be considered unacceptable for electronic device grade semiconductor material. The first InGaN QW diode lasers were demonstrated in material where a dislocation density on the order of 10^{10} cm^{-2} was present. Such a high concentration of extended state defects would be unthinkable in a cw GaAs-type diode laser, under typical current densities $J < 1 \text{ kA cm}^{-2}$. We will refer to the “bullet-proof” nature of the nitrides and their apparent defiance of conventional wisdom in the sections below. Even so, for long-lived diode lasers, innovative growth techniques have been required, particularly the use of lateral epitaxial regrowth to reduce the density of threading dislocations.

By contrast, the ZnSe-based II–VI QW diode lasers have been shown to be quite vulnerable to the presence of defects, as is the case with “conventional” III–V compound semiconductor lasers. Although the dislocation-related defect density has been reduced in the as-grown heterostructure material to below 10^3 cm^{-2} and this has led to the extension of the cw green laser (500–520 nm) lifetime to nearly 1000 h, it is still an open question whether further reductions in the defect density

are required in order to realize a technologically viable device (as it appears unlikely that InGaN lasers will be able to reach into the green). In spite of these apparent handicaps, and because of the extraordinary heteroepitaxial quality of the wide-gap II–VI QW devices, a great deal of very useful basic optical and optoelectronic science has been conducted in these systems. Accordingly, we will devote some space in this treatise to the II–VI materials and devices since their research has yielded important basic science insights. These insights are likely to be useful for future nitride diode lasers, including the ongoing push towards the near-ultraviolet, as well as the efforts to reduce the high threshold current density of the present InGaN QW violet diode lasers.

In this chapter we provide an introduction to the basic physical properties of wide-gap semiconductors relevant for optoelectronic materials. Chapter 10 is devoted to the discussion of contemporary device science blue diode lasers, including some examination of the physics of stimulated emission in wide-gap semiconductor light emitters. Chapter 11 examines the current efforts to produce blue and violet vertical cavity surface emitting devices.

9.2 OVERVIEW OF PHYSICAL PROPERTIES OF WIDE-BANDGAP SEMICONDUCTORS

Let us outline some of the generic physical properties of wide-gap semiconductors, which will illustrate and underscore some of the frustrations associated with developing practical optoelectronic devices based on GaN- and ZnSe-based semiconductors. We emphasize that both groups of semiconductors, perhaps better termed “semiinsulators”, share physical features which are profoundly distinct from, say, GaAs. Wide-gap semiconductors present special challenges particularly in terms of their electrical (p -type transport) properties. On the other hand, their optical properties offer new opportunities for LEDs, even reaching to the fundamental study of light-matter interaction.

9.2.1 Lattice matching

With a view towards heteroepitaxy, Figure 9.1 shows the bandgaps of a number of III–V and II–VI semiconductors as a function of their lattice constants. Whereas a number of possibilities exist for close lattice matching for heteroepitaxy of the zincblende cubic II–VIs, the situation is much more complex and still not fully resolved for the wurtzite Group-III nitrides, specifically in terms of the substrate. The left-hand side of Figure 9.1 displays their basal plane lattice constant a , together with that for SiC and ZnO. Note that the bandgap of the AlGaInN system covers a huge range, from approximately 1.9 eV to about 6.2 eV.

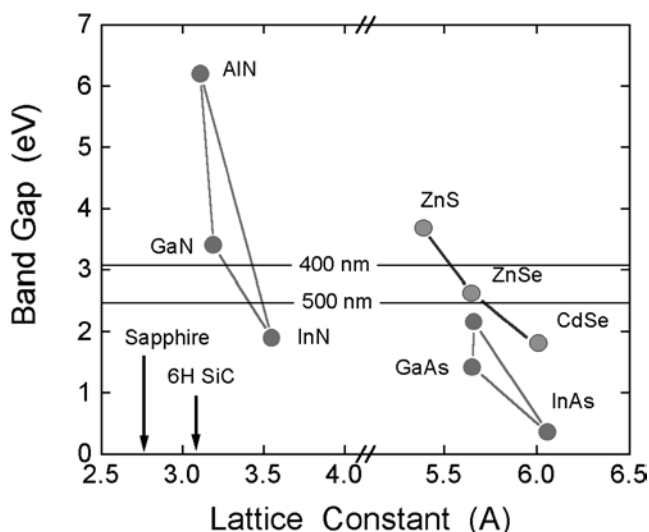


Figure 9.1: Energy bandgap versus lattice constant for heteroepitaxy of selected III–V and II–VI semiconductors.

Whereas very nearly lattice-matched GaAs substrates could be routinely used for high-quality ZnSe epitaxy, this luxury is not enjoyed by GaN and its related alloys (InGaN and AlGaIn). Compatible substrates are still being sought for growth of nitride layers, and this is a topic of current material science interest. A satisfactory solution for commercial high-brightness LEDs (e.g. those produced by Nichia Co. of Japan, Cree Lighting and Lumileds Lighting in the USA) and for the first cw demonstrations of violet lasers was the growth of a thick (up to several microns) GaN buffer layer atop of a vastly lattice mismatched sapphire substrate (13.5% mismatch). The use of either GaN or AlN nucleation layers in MOCVD (OMVPE) (metal-organic chemical vapor deposition (organo metallic vapor phase epitaxy)) epitaxy is viewed as being essential for the subsequent layered growth. Nonetheless, high densities of threading dislocations permeate the active light-emitting regions. As an alternative, blue LEDs grown on SiC substrates were introduced some time ago (by Cree Lighting). The first violet diode laser demonstration of an InGaIn QW device grown on a SiC substrate likewise occurred a while ago (Bulman *et al.*, 1997). SiC is also attractive as a possible alternative substrate for the nitride laser because of properties that impact practical device fabrication and operation: it has a good thermal conductivity and is readily cleaved to create laser facets. Other “substrate solutions” include efforts to create a natural single-crystal GaN substrate, either by increasing the buffer layer thickness to hundreds of microns by hydride vapor phase epitaxy (Molnar *et al.*, 1997) or by the growth a sufficiently high-quality and large-area GaN bulk crystal substrates (Porowski *et al.*, 1997).

Some of the combinations of materials that could be used to fabricate separate confinement QW diode laser heterostructures are highlighted in Figure 9.1, identified by the compounds connected by solid lines. The GaInN/GaN/AlGaIn and ZnCdSe/ZnSSe/ZnMgSSe QW heterostructures form the basis of the device designs that led to the first demonstrations of the cw violet and green-blue diode lasers, respectively. The typical layered structures for such lasers will be described in Chapter 10 where the design of both the electronic and optical confinement for the edge-emitting devices is outlined. In the case of the nitride lasers, new approaches have been introduced, where the techniques of lateral epitaxial overgrowth have created “artificial” substrates with much lower defect density than in the heterostructures grown directly on GaN/sapphire templates. As for the II–VI lasers, the beryllium-containing compounds (BeSe and BeTe) represented a development which offered the possibility of designing these lasers for the deeper blue, while strengthening the material due to an increase in its covalency (Waag *et al.*, 1996). It is worthwhile to note that large lattice mismatch strains that are the source of dislocations have not, to first order, hampered the evolution of the nitride LEDs and diode lasers so far, contrary to much of the conventional wisdom.

9.2.2 Epitaxial lateral overgrowth (ELOG)

While InGaIn QW diode lasers are able to operate even with extremely high densities of dislocations, their device lifetime as well as electrical transport properties and optical losses are such that technologically viable device performance is not obtained. An effective growth strategy has been developed to reduce the density of the extended defects arising from the use of lattice mismatched substrates, which has furthered both fundamental materials studies and technology development. The method of ELOG, introduced by Usui, Davis and others (Usui *et al.*, 1997, Nam *et al.*, 1997), involves interrupting the initial growth of the GaN layer to deposit and pattern a dielectric film before resuming growth. As shown in Figure 9.2, a stripe-patterned amorphous dielectric film (SiO_2) defines windows of exposed GaN for the crystalline regrowth. Proper orientation of the stripes relative to crystallographic axes of the underlying GaN promotes rapid lateral growth over the dielectric and planarization of the growing GaN layer. The benefit of this intriguing process is truncation at the dielectric of the threading dislocations that originate at the sapphire–GaN interface and the nearly total absence of these extended defects in the laterally grown GaN that is directly above the dielectric stripes. Above the dielectric, the two laterally propagating growth fronts from the opposite sides of a stripe coalesce to form an interfacial layer (the “suture” in Figure 9.2), the structural quality of which depends on the degree of tilt

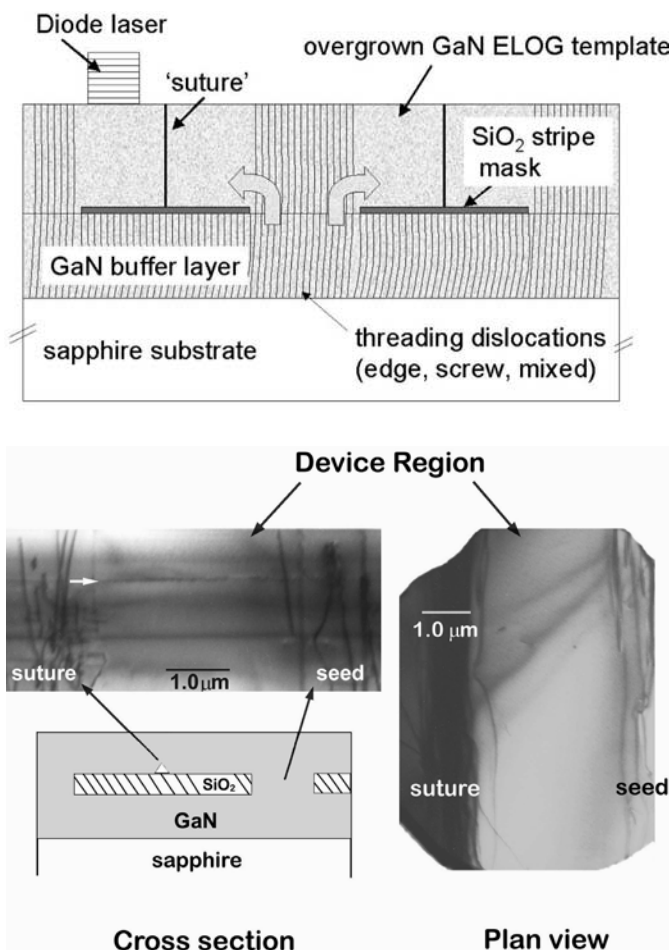


Figure 9.2: Schematic of ELOG (upper panel). The lateral growth fronts, emanating from spaces between the SiO_2 stripes, coalesce near the center of each mesa to form a ‘suture’ region that commonly still contains defects. The electron micrograph images show how the ‘wing’ regions between the seed areas and the sutures have significantly fewer dislocations. (From Romano (2000).)

misalignment between the coalescing fronts. The region above the dielectric between the coalescence interface and the edge of the window defines the “wing” of the ELOG material where the density of threading dislocations is reduced by several orders of magnitude, on either sapphire or SiC substrates, perhaps to values as low as 10^5 cm^{-2} and below. A dramatic illustration of the impact of reduced dislocation density by ELOG is the greatly reduced reverse leakage current obtained in GaN p – n junctions. In connection with ultraviolet AlGaIn photodetectors, a reduction of the reverse-bias leakage current by up to six orders of magnitude has been reported (Parish *et al.*, 1999).

Table 9.1. Comparison of selected material parameters for GaAs, GaN, and ZnSe

	Covalent Energy (eV)	Polar Energy (eV)	Young's modulus (GPa)	Thermal cond. W/cm K	T growth (°C)	Optical Phonon (meV)	Frohlich Coupling (meV) ^a	Exciton Binding (meV)
GaAs	5.55	1.88	120	0.45	−650	36	5	−10
ZnSe	5.55	3.80	80	0.19	−300	30	36	−40
GaN	8.85	3.92	150	1.3	>900	90	?	22 ^b

^a $\Gamma = \Gamma_0 + \Gamma_{LO}/[\exp(h\omega_{LO}/kT) - 1]$.

^b A-exciton for bulk GaN.

As discussed in Chapter 10, the use of ELOG has improved the performance of laser diodes. Groups at Nichia, Sony and others have demonstrated lasers with threshold current densities as low as $J \sim 3 \text{ kA/cm}^2$, fabricated from the portions of the ELOG wafer where dislocations are minimized. The characteristics of lasers fabricated from such ELOG wafers have been systematically investigated, specifically in terms of variations in threshold current density for diodes fabricated above the wing, window, and coalescence regions. The use of ELOG is expected to stabilize and extend the longevity of InGaN/GaN QW lasers, which is important for practical applications. Yet, the threshold current density even in the best lasers remains high, raising questions about the microscopic mechanisms involved in the formation of optical gain and stimulated emission in the InGaN QW system.

9.2.3 Basic physical parameters

Some of the key physical parameters of GaN and ZnSe are compared with GaAs in Table 9.1. Note the large polar energy for the chemical bond for both GaN and ZnSe, as well as the large covalent energy for the former. Thus, for example, in terms of its mechanical properties, GaN is a stiff and a “hard” material with a high thermal conductivity. The strength of the underlying chemical bonds leads the material to be able withstand high current densities (tens of kiloamperes per square centimeter) even in the presence of a very large number of threading dislocations and related microstructural defects. The epitaxial growth temperatures for the nitride semiconductors are considerably higher than those used for the growth of GaAs. However, since the optimal growth temperatures for AlGaN, GaN, and InGaN can cover a range of as much as 200 °C (in descending order), the MOCVD (or OVMPE) epitaxial growth of quality AlGaInN heterostructures is a significant challenge. By way of contrast, ZnSe requires a low growth temperature, a factor relevant to subsequent device processing, and is much more susceptible to dynamical dislocation generating processes that lead to device degradation.

Table 9.1 also shows the impact of the strong electron–hole Coulomb interaction (quantified here as the exciton binding energy) which leads to large enhancements in the interband optical cross-sections for GaN and ZnSe. This “built-in” property, generic for both the II–VI compounds and the III–V nitrides, has already been studied in much detail in ZnSe QWs, including diode lasers, demonstrating the impact of such a many-body enhancement to the optical gain and stimulated emission. We will return to this subject in Chapter 10. Table 9.1 also includes a measure of the strength of the coupling of electrons to optical phonons, a parameter of importance in electronic scattering and hot electron relaxation. The carrier mobility in wide-gap semiconductors at room temperature (and above) is strongly dominated by the LO–phonon scattering, leading specifically to low values of the hole mobility. Hot carrier energy relaxation, such as occurs when an electron or a hole is captured in an InGaN or ZnCdSe QW is likewise dominated by the LO–phonon interaction. While the exact values for the Frohlich coupling constant are largely unavailable for GaN, these are expected to exceed those in ZnSe which are very large in their own right as seen from the table. Of course, at high carrier injection such as encountered in a diode laser, partial screening of the Frohlich interaction is expected. The actual details of the screening process are so far unexplored in the nitride diode lasers.

9.3 DOPING IN WIDE-GAP SEMICONDUCTORS

We will not treat the vital but vast topic of *p*-doping in wide-bandgap semiconductors in this review in significant depth. The subject, while still not entirely understood, surely requires its own treatise, specifically in terms of the challenges and level of understanding that exists today for the *p*-type doping. The reader is referred to representative review articles, which contain more detailed discussion on the practice of doping in the nitrides (Gotz and Johnson, 1999) and wide-gap II–VI materials (Van de Walle, 1997). The international conference series devoted to each group of semiconductors have proceedings where the *p*-doping still figures prominently as a separate subject. In the next section we link up the subject with the issues of ohmic contact formation, which is another specific challenge in the blue/green diode lasers.

The microscopic issue concerning *p*-doping in the AlGaInN system is the nature of the local microscopic environment for the dopants and their correlation with the intrinsic and extrinsic defect microstructure. As a precursor to the development of GaN-based light emitters, a key breakthrough was the achievement of *p*-type doping of GaN using Mg and low-energy electron beam irradiation (LEEBI) “activation” treatment of this acceptor by Amano *et al.* (1989). Nakamura and co-workers at Nichia Chemical Company built on this fundamental breakthrough to achieve even higher *p*-doping (perhaps up to 10^{18} cm^{-3} range) while demonstrating

low resistivity in GaN epilayers (Nakamura *et al.*, 1992). Their approach relied on uniform activation of Mg throughout thick (micrometer scale) *p*-GaN layers by using high-temperature thermal annealing under nitrogen ambient. Subsequently, Nakamura and coworkers have employed short-period *p*-GaN/AlGaIn superlattices to achieve high *p*-doping levels in the 10^{18} cm^{-3} range; enhanced doping efficiency in such heterostructures with a lateral resistivity as low as $0.2 \Omega\text{-cm}$ has been reported (Kozodoy *et al.*, 1999, Nakamura *et al.*, 1998). Short-period superlattices, in terms of enhanced doping, improved contacts, and acting as “dislocation filters” are now included in violet laser designs.

Complicating the microscopic description of the impurity states in GaN:Mg is the scarce experimental information about the local chemical structure associated with the doping sites, with the added issue concerning the impact of the complex material microstructure on their electrical activation. While the actual practical details of doping are frequently still proprietary at this writing, it is generally acknowledged that typical Mg concentrations on the order of 10^{20} cm^{-3} are required for *p*-*n* junction devices. Hence less than 1% of the Mg dopant provides free holes, on a statistical basis.

Advanced theoretical models have been used to address the general question about the electronic structure associated with impurities in GaN but the connection to experiment remains tenuous at this time. We now understand that acceptors in GaN form acceptor–hydrogen complexes during epitaxial growth that remove the acceptor energy level from the bandgap, a mechanism previously established for acceptors in Si and GaAs. In MOCVD growth there are numerous sources of hydrogen (e.g., the primary source for nitrogen, NH_3 , as well as several metal-organic compounds). Interstitial hydrogen is incorporated in the GaN during growth and forms Mg–H complexes during cooling. Subsequently, these complexes can be thermally dissociated, with the hydrogen migrating out of the material or to other stable, electrically inactive sites, with creation of *p*-type conductivity. Using a hydrogen-free ambient during the high-temperature anneal prevents hydrogen indiffusion and repassivation from competing with thermal dissociation. Fundamental understanding of the local electronic structure of these complexes has been advanced through computational studies of Neugebauer and Van de Walle at Xerox (Van de Walle and Johnson, 1999), and the existence of the Mg–H complex was experimentally demonstrated by combined vibrational mode spectroscopy and Hall-effect measurements by Götz *et al.* (1996) at Xerox. On the other hand, there has been no experimental determination of the physical descriptors for the kinetics of the hydrogenation process such as the thermal dissociation energy or the diffusivities of monatomic, interstitial hydrogen in its different charge states.

Achieving the high hole concentrations at room temperature required for laser devices is further frustrated by the high activation energy for thermal ionization

of Mg acceptors in GaN, measured by several investigators to be approximately ~ 200 meV at low densities. In the one-electron approximation, the binding energy depends on the dielectric constant and effective mass of the material. The nitride system possesses low dielectric constants ($\epsilon \sim 9.5$ for GaN versus 13 for GaAs) and large effective masses (e.g., the heavy hole mass in GaN is $\sim 0.75m_0$), which result in large binding energies and only partial ionization of the available acceptors at room temperature under thermal equilibrium. The problem can be further exacerbated by the presence of significant concentrations of *n*-type background contaminants that partially compensate the *p*-type dopants. The low *p*-type doping (typically 10^{17} cm^{-3}) leads to high contact resistance with almost any metal of choice. Ongoing research is focusing on means to increase the *p*-doping concentration and identify a shallower new *p*-dopant to improve the operating voltage and power efficiency of present lasers and LEDs. Increasing the bandgap for future ultraviolet light emitters by employing AlGaN at elevated Al concentrations ($>30\%$) raises another fundamental issue due to possible coupling of the acceptor states with phonons. In an analogous case studied in the II–VI semiconductor alloys, such “polaron” formation was found to result in very deep acceptor levels, creating a fundamental barrier for *p*-doping.

While the specific issue associated with the strongly compensating effect of atomic hydrogen in the doping process appears to have been settled, the role of dislocation-type defects is less clear. Underlining the impact of the defect microstructure are experimental observations that dopants may concentrate in spatial regions of high dislocation density, see for example Bertram *et al.* (1999). To put the challenges into perspective, device quality *p–n* junctions for a high-performance InGaN blue-lightemitting source have been realized probably in only about a couple of dozen laboratories worldwide at this writing, due at least in part to the difficulty in implementing *p*-doping in practise. Finally, an open future question for the wider-gap AlGaN at high Al concentrations, relevant to the development of ultraviolet LEDs and lasers, concerns the possible deep-level formation induced by hole–phonon interaction, similar to the “self-trapping” phenomena (“AX center”) encountered in the case of *p*-ZnMgSSe mentioned below.

In contrast, the case of the nitrogen acceptor in *p*-ZnSe, a primary dopant, is relatively well understood and serves as a potentially useful reference for the nitride case as well. We mention briefly that issues concerning the ZnSe-based II–VI compounds relative to the LEDs are mostly centered on the microscopics of *p*-doping of the larger bandgap quaternaries such as ZnMgSSe ($E_g > 3$ eV), including the coupling of the electronic states with the lattice (phonons) that can lead to the formation of deep (acceptor) states (Han *et al.*, 1994). In this instance the hole–phonon coupling creates an AX center (analogous to DX centers in GaAs) relatively abruptly with increasing Mg concentration. The *p*-doping has been

successfully achieved only in molecular beam epitaxy, where atomic nitrogen is supplied from an *in-situ* RF plasma source (dissociating N_2) so as to achieve free-hole concentrations up to and beyond $5 \times 10^{17} \text{ cm}^{-3}$. For reasons that have been generally attributed to the compensating action of hydrogen, the use of nitrogen (or any other *p*-doping scheme) in the context of MOCVD growth has been singularly unsuccessful. On the other hand, *n*-type doping with free-carrier levels greater than $1 \times 10^{18} \text{ cm}^{-3}$ has been readily achieved within both MBE and MOCVD with chlorine and iodine from zinc chloride and zinc iodide sources as the *n*-type dopants for ZnMgSSe and BeZnMgSe based materials, respectively.

9.4 OHMIC CONTACTS FOR *p*-TYPE WIDE-GAP SEMICONDUCTORS

Following the breakthroughs in the *p*-doping of the ZnSe -based material around 1989 and the *p*-doping advances in the nitrides shortly thereafter, the next challenge to be addressed was the fabrication of low-resistance ohmic contacts to *p*-type materials suitable for the high-injection case encountered in a diode laser. At the simplest level, the problem exists due to the lack of metals with suitably large work functions, which are needed to reduce the (potentially very large) potential energy barrier for hole injection. While reasonably-low-resistance contacts could be formed more easily on *n*-type materials, with specific resistances of $10^{-4} \Omega \text{ cm}^2$ and better, the poor contacts to *p*-type ZnSe and *p*- GaN caused the first LEDs and diode lasers to operate at unacceptably high voltages. For example, the first pulsed ZnCdSe QW diode lasers in 1991 had a voltage threshold exceeding 20 V, as did also the first pulsed InGaN QW lasers in 1996. To obtain a low-resistance contact between a metal and a wide-bandgap semiconductor (*p*- or *n*-type) presents a fundamental dilemma of electrically bridging two materials with vastly different electronic band structures. The difficulty of the problem scales qualitatively with the bandgap of the semiconductor. In the case of GaN and ZnSe as well as their alloys, the relative ease of achieving heavy *n*-type doping, together with the position of the surface Fermi level, presents a rather low barrier for electron injection with many common metals ($\ll 1 \text{ eV}$).

Here we sketch the present state of affairs for the *p*-contacts to the *p*- AlInGaN system. While the best InGaN QW diode lasers have been operated at threshold voltages near 4 V, there appears to be no clear cut approach to low contact resistance for *p*- GaN based on an understanding of the underlying microscopic physics. By way of contrast, we include a proven approach to creating *p*- ZnSe contacts in this section: a “textbook” case of using bandstructure engineering to achieve “electronic state impedance matching” at the interface. This in turn led to the demonstration in 1993 of the cw SQW (square quantum well) ZnCdSe green

diode laser at voltages as low as 4 V. The II–VI case provides useful guidelines for designing contact schemes for the *p*-nitrides, where such bandstructure engineering concepts are likely to be necessary in the future to reduce the finite Schottky barriers and voltage drops across the contacts, especially in the AlGaIn system in the ultraviolet.

9.4.1 Ohmic contacts to *p*-AlGaInN

The discovery of Mg as a *p*-dopant in GaN, subject to subsequent annealing for hole conductivity activation, was not only a seminal development that paved a way for the fabrication of *p*–*n* junctions but also brought to sharper focus the issue of ohmic contacts to *p*-type material in the blue LEDs and, especially, the diode lasers. A somewhat bewildering host of contacting schemes have been reported in the literature, including metals and their combinations that involve Au, Ni, Ti, Pd, Pt, W, WSi, Ni/Au, Pt/Au, Cr/Au, Pd/Au, Au/Mg/Au, Pd/Pt/Au, Ni/Cr/Au, Ni/Pt/Au, Pt/Ni/Au, Ni/Au–Zn, Ni/Mg/Ni/Si, etc. In addition to its purely electronic function, a metal such as Pd might be helpful as a gettering agent for the extraction of hydrogen from *p*-GaN, some of which remains imbedded in the epilayers as part of the doping process, even after annealing. Because much of the information related to contact fabrication is proprietary, it is difficult to gauge the state of the art in an open review such as this treatise. It is clear, however, that the acceptable *p*-contact resistance in a typical commercial blue LED is probably on the order of $R_c \sim 10^{-2} \Omega \text{ cm}^2$: this is most often implemented with Ni/Au-based contacts. The contacts require a thermal annealing step, most commonly in a nitrogen ambient. This range of contact resistance, while a working solution for an LED, is quite unacceptable for a high-performance device such as the diode laser, compared for example with $R_c \sim 10^{-5} \Omega \text{ cm}^2$ and below found in a long wavelength III–V laser. From the best-reported results for the InGaIn MQW diode laser, one can surmise that a contact resistance on the order of $10^{-3} \Omega \text{ cm}^2$ has been now achieved routinely, and with some effort the values in practical devices probably reaching into the range of $R_c \sim 10^{-4} \Omega \text{ cm}^2$. These values are still high and contribute significantly to the electrical and thermal budget of the blue laser.

It is difficult to evaluate the relatively few published reports on contact resistance to *p*-GaN, because of the sensitivity of contact resistance to both the quality of the underlying *p*-GaN material and to the detailed history of the process used to fabricate the contacts (recall also that *p*-GaN requires its own postgrowth thermal activation in the first place). Invariably, some form of thermal annealing (typically at temperatures around 500 °C) is associated with the *p*-contact formation, sometimes in conjunction with the postgrowth activation of the free holes, sometimes in addition to this step. *p*-type contact schemes based on Ni/Au and on Pd/Pt/Au

are presently the most popular choices because they are easy to fabricate and have been extensively used in blue LEDs. The Ni/Au contact provided the starting point for the diode lasers, judged, for example, by the current–voltage characteristics of InGaN MQW laser diodes by Nakamura *et al.* (1998). However, even in this case, the microscopic characteristics of the contact remain largely unknown and subject to empirical design/process parameters. To illustrate the point, there are several reports in the literature of a low-contact-resistance scheme with Ni/Au, where the thermal annealing in oxygen-rich ambients might have resulted in the formation of NiO (e.g. Ho *et al.* (1999), Sheu *et al.* (1999)). Figure 9.3 shows the experimentally extracted contact resistance of Ni(10 nm)/Au(5 nm) to *p*-GaN in different ambient annealing conditions from the work of Ho and colleagues. We note that NiO is a wide-gap, usually *p*-type semiconductor so that an “accidental” bandstructure engineered GaN/NiO/Ni–Au contact scheme might be generated in this case. However, other alternatives have been suggested, including one where the incorporation of O₂ into the activation ambient lowers the sheet resistivity of *p*-GaN by enhancement of outdiffusion of hydrogen from the *p*-GaN layer (Hull *et al.*, 2000). In any event, it is clear that the subject requires continued basic research from which future generations of nitride diode lasers will benefit. It needs to be emphasized that, in addition to low contact resistance, a practical contact used in a laser diode must also be able to sustain high current densities (well above 1 kA cm⁻²) without degradation.

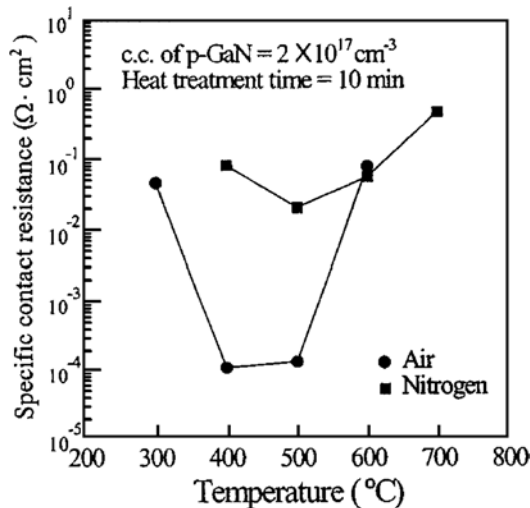


Figure 9.3: The effect of thermal annealing on the contact resistance of Ni(10 nm)/Au(5 nm) to *p*-GaN under two different ambient conditions. (From Ho *et al.* (1999).)

9.4.2 New approaches to *p*-contacts

We note two innovative approaches to contacts on *p*-nitrides. The first uses short-period superlattices which can show high hole concentrations ($>10^{18} \text{ cm}^{-3}$). For example, Zhou *et al.* (2000) used a *p*-AlGaIn/GaN superlattice to obtain low contact resistance. Kumakura *et al.* (2000) extended the concept to InGaIn/GaN superlattices, recognizing further that the smaller bandgap of InGaIn compared with GaN should reduce the metal–semiconductor contact barrier and hence the contact resistance. These authors also showed that the *p*-InGaIn contact was less susceptible to dry etching damage than a comparable *p*-GaN case, a feature of importance in device fabrication.

An entirely different approach has been demonstrated, in which a (Esaki) tunnel junction has been employed to provide the effective contact to *p*-GaN. A tunnel junction is composed of a heavily-doped p^{++}/n^{++} diode where interband tunneling across the very narrow depletion layer provides a low resistance that is constant for small bias voltages across the junction. The use of the tunnel junction in conventional III–V semiconductors can be found in certain specific edge-emitting and vertical-cavity lasers (Wierer *et al.*, 1999). For example, this approach has been employed to minimize the portion of *p*-type material in an InP-based VCSEL (vertical cavity surface emitting laser) (Ortsiefer *et al.*, 2000) since the InP-based *p*-layer has relatively high resistivity, which leads to serious heating problems. In case of the nitrides, it is not clear *a priori* whether the *p*-type doping allows for a workable tunnel junction. However, following demonstrations of Mg-doping of InGaIn layers with very high carrier concentrations ($p \sim 1 \times 10^{19} \text{ cm}^{-3}$ at 300 K), a workable tunnel junction based on the successful growth of a heavily-doped p^{++}/n^{++} thin InGaIn/GaN junction has been achieved (Jeon *et al.*, 2001, Takeuchi *et al.*, 2001). In the nitride case a tunnel junction becomes especially useful as a contacting scheme with good lateral current spreading, given that the conductivity of *n*-type GaN is nearly 100 times higher than that of *p*-GaN. To achieve hole injection with the tunnel junction embedded in the GaN-based LED structure, Takeuchi and colleagues grew 30 nm GaN:Si $^{++}$ and 15 nm InGaIn:Mg $^{++}$ atop a GaIn–InGaIn QW *p*–*n* LED heterostructure. In particular, the tunnel junction was grown directly on the *p*-GaN layer that normally completes the LED device structure and resides on top of the *p*–*n* junction light emitter device. As shown in the schematic Figure 9.4 of the entire device, the top contact can now be made to the n^{+} -GaN:Si layer with a low contact resistance. As shown in Figure 9.5, the forward voltage of the entire LED, including the voltage drop across the reverse-biased tunnel junction, was measured to be $\sim 4.1 \text{ V}$ at 50 A cm^{-2} injection current density, while that of a standard LED with a conventional contact structure is 3.5 V. The light output of the diode with the tunnel junction was likewise comparable to that of the standard device. The

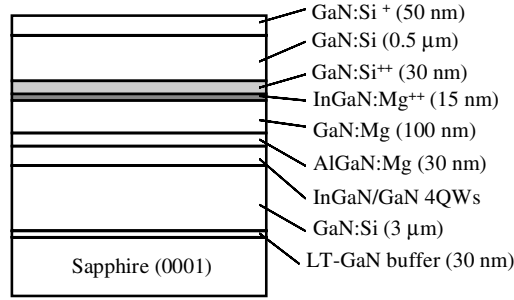


Figure 9.4: Schematic of a blue LED structure incorporating a p^{++}/n^{++} InGaIn:Mg/GaN:Si tunnel junction.

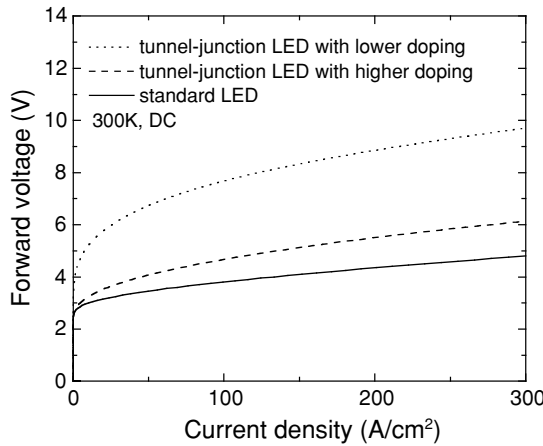


Figure 9.5: I – V characteristics of the LED with the lower doping levels at the tunnel junction, the LED with the higher doping levels at the tunnel junction, and our standard LED with p -contact. (From Takeuchi *et al.* (2001).)

tunnel junction eliminates the need for a highly-resistive p -AlGaIn cladding layer in short-wavelength laser diodes and the semitransparent electrode required for current spreading in conventional GaN-based LEDs. We will return to this subject when discussing vertical cavity devices in Chapter 11.

9.4.3 Ohmic contacts to p -ZnSe: bandstructure engineering

A solution to the acute problem of contacts to p -ZnSe was achieved some time ago by a bandstructure engineering approach that involves a short-period p -ZnSe/ZnTe superlattice, where individual layer thicknesses are on the order of a monolayer (Fan *et al.*, 1992, Hiei *et al.*, 1993). Unlike ZnSe, the smaller-bandgap ZnTe can be doped to obtain hole concentrations up to 10^{19} cm^{-3} (by nitrogen) within the

molecular beam epitaxy growth of ZnTe (Fan *et al.*, 1993). Since a low-resistance contact to such highly-doped *p*-ZnTe epilayers can be achieved by Pd and Pt, one is led to consider the use of ZnTe as an intermediate “electrical buffer” for contacting to *p*-ZnSe. However, the large valence band offset ($\Delta E_v \approx 1$ eV) between ZnTe and ZnSe layers forms a barrier to the hole injection at a *p*-ZnTe/*p*-ZnSe interface (ZnTe/ZnSe is a Type II QW). A possible solution for removing the expected energy barrier in the valence band (a potential energy “spike” due to the electrostatic charge redistribution) is to introduce a graded bandgap *p*-ZnSe_{1-y}Te_y layer in which the Te concentration varies from $y = 0$ to $y = 1$ across such a contact layer. In practice, the Zn(Se,Te) alloy was created by a “pseudograded” *p*-type, strained ZnTe/ZnSe ultrathin layer, with the ZnTe and ZnSe layer thicknesses in each cell varying to approximate a graded bandgap material. Results of conductivity measurements of *p*-ZnSe epilayers for three different contacting schemes consisting of: (i) Au/*p*-ZnTe/*p*-Zn(Se,Te)/*p*-ZnSe, housing the graded pseudoalloy, (ii) direct Au/*p*-ZnSe, and (iii) an Au/*p*-ZnTe/*p*-ZnSe heterostructure are compared in Figure 9.6. As seen in Figure 9.6(a), the graded contact is quite ohmic, and shows a contact resistance of $3 \times 10^{-4} \Omega \text{ cm}^2$. The inset shows that the *I*-*V* characteristics maintain the same slope even at a few millivolts from the origin. Figure 9.6(b) shows the characteristics of the contact formed by gold deposited onto an as-grown ZnSe:N epilayer. The *I*-*V* characteristic corresponds to two back-to-back Schottky diodes; the observed turn-on voltage is the reverse bias

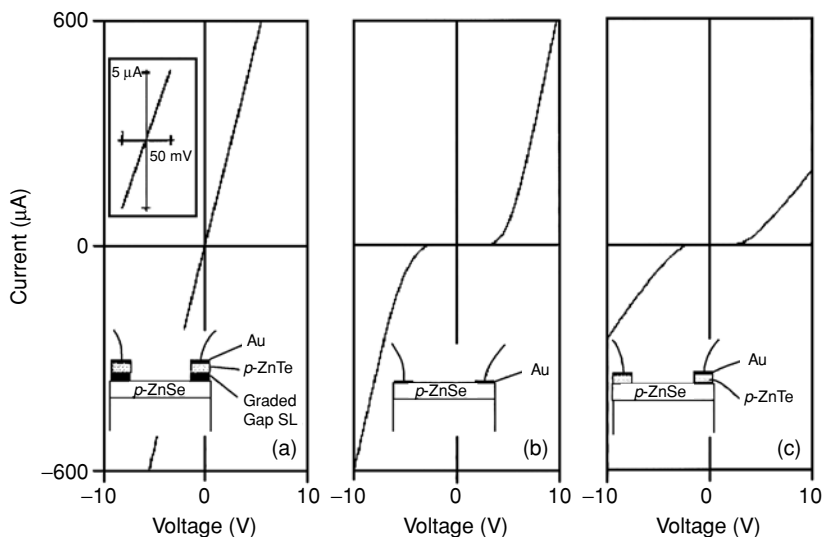


Figure 9.6: *I*-*V* characteristics at room temperature for contact arrangements on a *p*-ZnSe epilayer whose schematics are shown in the inset: (a) Au/Zn(Te,Se) graded gap layer/*p*-ZnSe, (b) Au/*p*-ZnSe, and (c) Au/*p*-ZnTe/*p*-ZnSe. (After Fan *et al.* (1992).)

breakdown voltage. Increasing the doping level is expected to reduce the “turn-on” voltage of the contact. Figure 9.6(c) shows the I - V characteristic when p -ZnTe is used to inject holes into the ZnSe epilayer in the absence of the graded region.

9.5 SUMMARY

In this chapter we have given an overview of selected physical attributes and approaches to heterostructure synthesis which are of specific importance in the design and material choice for the short-wavelength nitride-based light emitters. These have included issues of p -doping, contacts to p -type material, and the approaches to dislocation/defect reduction. While there has been much progress in all of these areas, further research on these subjects continues with the aim of improving the performance and expanding the applicability of the violet/blue diode lasers. In the next chapter we focus on the device science of these lasers, specifically on the edge-emitting InGaN MQW emitter.

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10

Device design, performance, and physics of optical gain of the InGaN QW violet diode lasers

10.1 OVERVIEW OF BLUE AND GREEN DIODE LASER DEVICE ISSUES

In this chapter we focus chiefly on the device science and engineering features of the violet edge-emitting InGaN MQW diode lasers. The extraordinary progress made with these devices since 1999, spearheaded by Nakamura and coworkers, seems to assure them an important place in future optoelectronics technology (Nakamura, 1999). By 2001, approximately half a dozen research groups reported achieving lifetimes of hundreds to a thousand hours for cw room-temperature operation, although the extrapolated lifetime of 15 000 hours at Nichia remained unequalled. Among the other groups we mention those at the laboratories of Sony, Toyoda Gosei, NEC, and Sharp in Japan, Samsung in Korea, and Xerox PARC, Cree Lighting, and Agilent Technologies in the USA.

A number of the core issues that intertwine the design, performance, and the physics of operation of the nitride lasers will be discussed in this chapter. We will focus on representative heterostructures that encompass the requirements of joint electronic and optical confinement, comment on some fabrication techniques, and highlight continuing challenges. The last include questions concerning the high threshold current density and the continued efforts to create artificial substrate templates for reducing the misfit (threading) dislocation density for improved device performance and lifetime. At a more fundamental level, there is evidence that the InGaN alloy which forms the optically-active QW medium has characteristic compositional disorder that impacts the gain spectrum of the laser. This feature, which increases in seriousness with the indium concentration, may restrict the operation of the devices at practical threshold current densities to the violet, leaving the longer blue and green regions to await future developments, perhaps involving complementary material approaches.

While the prospects for developing the green ($\lambda \sim 500$ nm) ZnCdSe QW diode laser into a practical technology are hampered by the serious, unresolved degradation problems, a brief examination of its basic device science offers a useful point of comparison. This is so, in particular, because the threshold current densities that have been achieved in the ZnCdSe QW lasers (< 200 A cm⁻²) (Katayama *et al.*, 1998), are more than an order of magnitude lower than those achieved in the best InGaN QW lasers today (~ 3 kA/cm²). Since at the level of simple effective mass theory, the electronic structure and optical properties of the II–VIs and nitride III–Vs are rather comparable, the contrast in the threshold current density suggests that significant improvement in the operation of the nitride diode lasers should be possible in the future. Moreover, as illustrated below, the physics associated with optical gain and stimulated emission that is generic for wide-gap semiconductors is quite sharply drawn out in the II–VI lasers and serves as a pedagogical treatment for other wide-gap semiconductor systems. The rich many-electron physics as related to optical gain offers specific enhancement factors to the maximum achievable optical gain at a given injection level so that the subject is of practical importance to the laser designer as well.

Conventional GaAs-based III–V compound semiconductor diode lasers have evolved over three decades, during which time their device performance and durability have steadily improved due to the combination of progress in material growth and device fabrication techniques. A variety of design and fabrication approaches have been developed to produce today's sophisticated III–V laser devices for optical telecommunication, and some of these techniques have contributed usefully to the development of wide-bandgap semiconductor diode lasers. On the other hand, because the nitrides have very different chemical and physical properties, processing tools and steps specific to the fabrication of nitride devices have been developed independently in many laboratories.

10.2 THE InGaN MQW VIOLET DIODE LASER: DESIGN AND PERFORMANCE

10.2.1 Layered design and epitaxial growth

The prototypical violet edge-emitting nitride diode lasers are constructed around an InGaN/GaN/AlGaIn MQW separate confinement heterostructure (SCH), most frequently grown on a (nonconducting) sapphire substrate by MOCVD (OMVPE). As already noted in Chapter 9, the large lattice mismatch and issues of crystalline orientation create a number of problems for the laser devices, in terms of eventual large-volume manufacturing and performance. There have been successful attempts to realize an InGaIn MQW laser on SiC substrate at Cree Lighting which provides a considerably smaller lattice mismatch ($\sim 2\%$) and allows for conventional cleaving

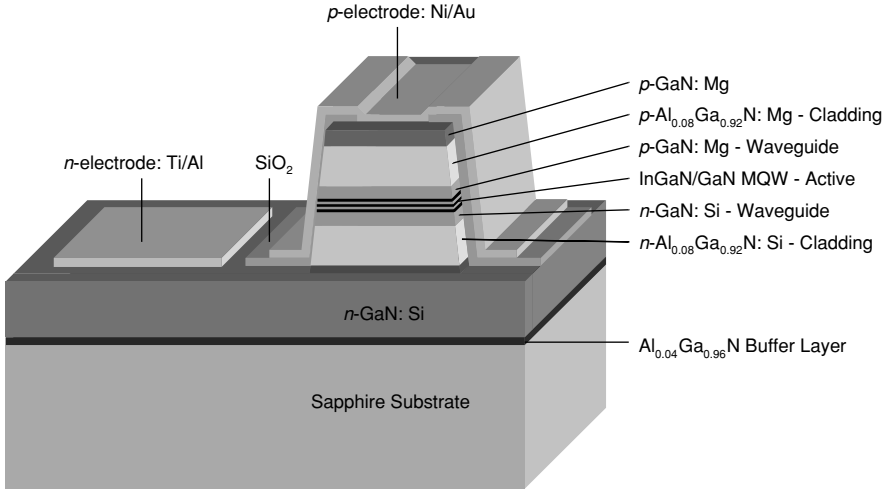


Figure 10.1: Schematic of layer structure for a SCH MQW violet laser AlGaInN heterostructure.

techniques to be used for fabricating optical resonator facets. For a review of the crystal growth on the Group-III nitrides by the MOCVD approach, the reader is referred to an article by DenBaars and Keller (1999).

The details of growth of the state-of-the-art material for InGaN QW diode lasers involve specific strategies to realize an electronically and optically optimized structure, with emphasis on electronic/photonic confinement, large radiative recombination current, and low optical losses. In Figure 10.1, the layering details (exclusive of the substrate template details) are shown for a typical InGaN SCH MQW diode laser, which combines the generic features of a separate confinement edge-emitting diode laser, within the constraints imposed by the growth of the AlGaIn, GaN, and InGaIn cladding, waveguide, and QW active layers, respectively. We note that a wide range of temperatures is required when growing the three different materials, anywhere from about $T = 800^\circ\text{C}$ for InGaIn to nearly $T = 1100^\circ\text{C}$ for AlGaIn. Furthermore, as noted in Chapter 9, the p -type doping requires both a high concentration of the Mg dopant ($\sim 10^{20}\text{ cm}^{-3}$) as well as (usually) a postgrowth annealing step for activating the free-hole conduction. Similarly, the fabrication of low-resistance p -contacts is still a nontrivial challenge.

The heteroepitaxy of a typical diode laser structure, such as in Figure 10.1, might find a low-temperature-grown GaN nucleation layer ($\sim 250\text{ \AA}$), a possible second $\text{Al}_{0.10}\text{Ga}_{0.10}\text{N}$ buffer layer ($>1000\text{ \AA}$), an $n\text{-GaN:Si}$ ($\sim 2.8\text{ }\mu\text{m}$) contact/buffer layer, an $\text{Al}_{0.10}\text{Ga}_{0.10}\text{N:Si}$ lower cladding layer ($>5000\text{ \AA}$), a GaN:Si lower waveguide layer ($\sim 1000\text{ \AA}$), an $\text{In}_{0.15}\text{Ga}_{0.85}\text{N/GaN}$ MQW region, having approximately $20\text{--}50\text{ \AA}$ thick wells and $75\text{--}100\text{ \AA}$ thick barriers, an $\text{Al}_{0.2}\text{Ga}_{0.8}\text{N:Mg}$ “blocking layer” ($\sim 250\text{ \AA}$) (not shown explicitly), a GaN:Mg upper waveguide layer

(~ 1000 Å), an $\text{Al}_{0.10}\text{Ga}_{0.90}\text{N}:\text{Mg}$ upper cladding layer (~ 5000 Å), and a $p\text{-GaN}:\text{Mg}$ contact layer (~ 1400 Å). The role of the blocking layer is in part to prevent electron overflow (leakage current) but also involves other considerations, including prevention of the InGaN decomposition under the high-temperature growth of the structure above the MQW section. The range of In concentrations for the lowest-threshold diode lasers has so far been rather limited ($x \sim 0.10\text{--}0.15$), with typical wavelength of emission near $\lambda = 410$ nm. The optimum number of quantum wells has been determined empirically to be in the range of 3–5. An example of a different kind of pragmatic challenge, nearly unheard of in any other semiconductor lasers, is the ease with which cracks form in the AlGaIn-containing heterostructures. The cracks are induced by the stress introduced into the AlGaIn layers due to lattice mismatch and, especially, due to the differences in the coefficients of thermal expansion with respect to the thick GaN layers. Finally, as already mentioned in Chapter 9, the initial buffer layers can be synthesized essentially on a different substrate template, prepared by epitaxial lateral overgrowth that significantly reduces the density of threading dislocations (of screw, mixed, and edge type) in the active region.

Many of the details in optimizing the SCH design and the active QW region have been determined empirically, with only a limited amount of the type of modeling which is routinely applied to the infrared communication lasers. Accurate information about the QW confinement of the InGaIn/GaN heterojunction is not available from spectroscopic work, owing largely to the very soft bandedge of InGaIn that originates from compositional anomalies in this ternary. We will discuss the impact of this problem on the optical gain later. The common anion supposition implies that the electronic confinement in the valence band is weak, perhaps on the order of 50 meV or less, while the conduction band offset might be expected to be on the order of 250–300 meV for the typical average In composition ($x \sim 0.10\text{--}0.15$) in the laser devices. However, both the In compositional fluctuations and the presence of large (fluctuating) strain and piezoelectric fields makes it very difficult to obtain an operationally meaningful measure of the QW confinement. There seems to be an empirical agreement that QW thicknesses on the order of 40 Å are optimal for the diode laser performance, with 3–5 wells defining the MQW active region. Unlike the cases of GaAs- and ZnSe-based lasers, only a limited amount of systematic work exists concerning the designs of the SCH MQW nitride lasers and the phenomenon of current overshoot. Yet the reports from various laboratories suggest that a significant leakage current competes with the radiative recombination current in both the diode lasers as well as LEDs at high injection, indicating that the finite QW confinement is a factor. We note that, while the electron–optical phonon (Frohlich) coupling is very large in GaN, and hence would be expected to promote very efficient carrier relaxation in the InGaIn QW, there are no accurate experimental data about the rates of electron–hole capture to the quantum well in the nitride heterostructures either.

All of the first InGaN QW diode lasers were devices grown on thick GaN buffer layers on the (0001) *c*-face of sapphire. This meant that dislocation densities in the range of 10^8 – 10^{10} cm⁻² were threading into the active region (Lester *et al.*, 1995, Ponce *et al.*, 1994). Later in this chapter we show examples of high-spatial-resolution microscopies that strongly suggest how these types of extended-state defects in the nitrides do not lead to overwhelming nonradiative recombination rates. Such behavior is in remarkable contrast to all other semiconductor light emitters. Equally surprising is the fact that these defects do not doom a high-current-density device such as the diode laser to very rapid degradation and early death, which, again, would be expected based on past experience with the GaAs and ZnSe diode lasers. In fact, Nakamura (1999) has reported a cw InGaN MQW diode laser operating for several hundred hours with a threshold current density of 3.6 kA cm⁻² in this type of high-defect-density material, a feat unlikely to be equalled by any other electronic material. An oversimplified but useful answer concerning the question about the remarkable tolerance of GaN and its related compounds to high injection and temperature can be found in the large covalent bond energy that makes this family of semiconductors exceptionally stable (see Table 9.1).

For truly long-lived blue and ultraviolet diode lasers (lifetime > 10⁴ h), this high defect density is likely to be unacceptable. Furthermore, the thermal budget even in today's very best InGaN diode lasers is still very high, due in part to the high-threshold-current density in the edge-emitting devices. Innovative approaches are now beginning to be implemented to address these issues. A significant reduction in the defect density has been achieved by the introduction of the ELOG technique which was introduced and described in Chapter 9. Recall how such spatially patterned growth is implemented by employing a micron-scale apertured SiO₂ mask on the GaN buffer layer. The epitaxial growth nucleates on the exposed GaN within the stripes of the SiO₂ template and, under a sufficiently high temperature ($T > 1050$ °C) and constituent flow in the MOCVD reactor, acquires a large lateral growth rate, once the aperture area is filled with GaN. From first work by Usui *et al.* (1997) and Nam *et al.* (1997), it became clear how the threading dislocations that emanate from the GaN–sapphire interface extend largely only just above the window area in the SiO₂ mask. By the standards of the nitrides, there can be very few (<10⁶ cm⁻²) dislocations in the overgrown areas above the SiO₂ mask, specifically in the so-called “wing area” (see below). These tend to be of the edge dislocation type, parallel to the (0001) plane after a 90° bend in the regrown region. After a thick (say >20 μm GaN) layer has been overgrown, this new “substrate” is ready for the subsequent deposition of an optoelectronic or electronic device heterostructure. Sakai *et al.* (1998), among others, have made detailed studies of the dislocation propagation during the ELOG process.

Shortly after reports of success with the ELOG approach, Nakamura *et al.* (1998) implemented the technique for their InGaN MQW laser diodes. A significant

improvement in the room-temperature cw device lifetime to beyond 100 h was achieved, apparently as a direct consequence of the reduced dislocation density. The Nichia group applied an SiO₂-free ELOG technique (where SiO₂ is used as an etch mask to define the GaN window region and then etched away prior to the continued epitaxy), which was originally developed by Zheleva *et al.* (1998), to further extend the device lifetime longer than 1000 h under high power (~50 mW) cw operation (Nakamura *et al.*, 1999). It is worth noting that the ELOG techniques, while being very desirable for the advancement of the violet lasers, are likely to be essential for nitride-based short-wavelength photodiode detectors. The necessity of reducing the reverse bias leakage current (“dark current”), which is presently dominated by contributions from the dislocations and related extended state and point defects, makes it imperative to use a truly high-grade electronic material. In this instance, the eventual requirements for the AlGaIn heterostructures may not be very different from those that are routinely required from other semiconductors in electronic and optoelectronic applications.

We now give contemporary examples of the impact of the ELOG-based substrates on the violet diode lasers. Tojyo *et al.* (2001) have studied the correlation between photoluminescence efficiency, surface profile of cleaved laser facets, and laser performance on ELOG-based substrates. The relationship between the dislocation density (measured as etch pit density) and photoluminescence efficiency of the approximately 5-μm thick GaN substrate is shown in Figure 10.2, whereas

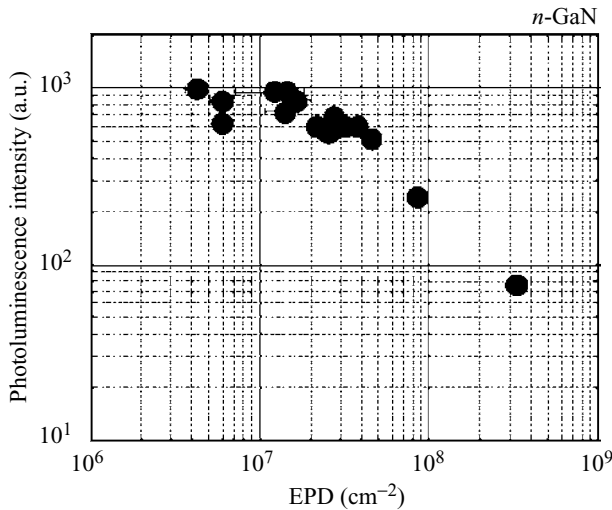


Figure 10.2: Correlation between photoluminescence intensity and etch pit density (EPD, dislocation density) in GaN substrate, demonstrating the impact of ELOG techniques. (After Tojyo *et al.* (2001).)

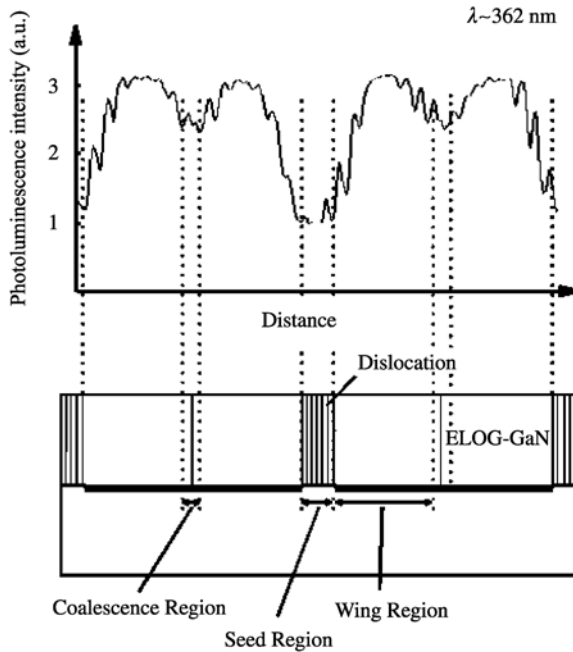


Figure 10.3: Spatially-resolved photoluminescence intensity in ELOG-GaN. (After Tojyo *et al.* (2001).)

Figure 10.3 shows the spatially resolved photoluminescence emission, indicating that the “best-quality” material is found under the wing region. Only the mixed dislocation type was found in this region, at concentrations less than 10^5 cm^{-2} . To prepare the optical resonator required for the complete diode laser structure, the sapphire substrate was thinned to about $100 \text{ } \mu\text{m}$ and the laser diode facets were subsequently fabricated by cleaving (as an alternative to their fabrication by dry etching). Interestingly and usefully, the surface flatness and morphology were superior for those portions of the cleaved facets that coincided with the wing region of the ELOG substrate. Figure 10.4 shows the results of atomic force microscopy (AFM) profiling where the smoothness of the laser facet can reach values better than 1 nm , in contrast with roughness measured on cleaved laser devices grown directly on sapphire (without the ELOG process) beyond 10 nm . The latter value for roughness adds scattering losses of several percent to the optical resonator and would be considered unacceptable in a high-performance diode laser device. In the case of the ELOG devices by Tojyo and others, lifetime tests under 30 mW cw output power indicate good performance beyond 1000 h .

The group at Nichia has systematically studied various ELOG approaches and their correlation with diode laser device degradation and lifetime (Nagahama *et al.*, 2000). Figure 10.5 shows a schematic for comparing three different

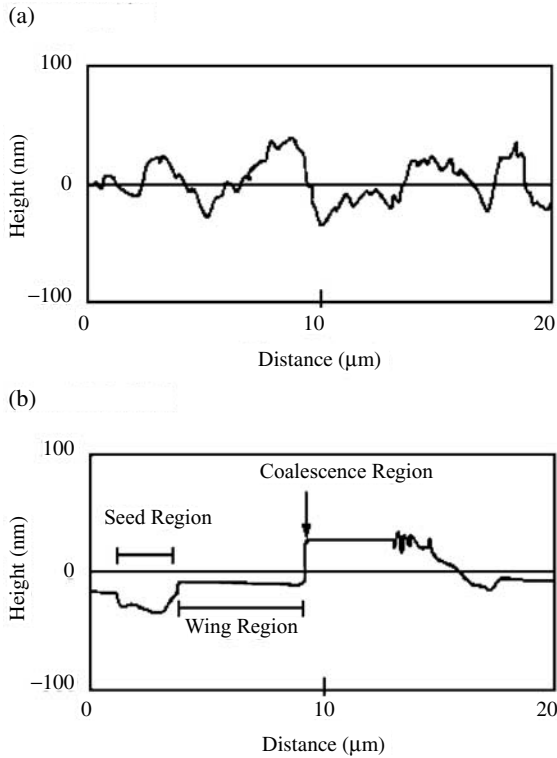


Figure 10.4: Effect of a GaN ELOG substrate/template on facet roughness of a cleaved diode laser: (a) on sapphire; (b) on ELOG-GaN. (After Tojyo *et al.* (2001).)

substrate/template cases in which the ELOG idea is implemented at levels of successively increasing complexity and physical thickness. The first case corresponds to the usual ELOG already outlined. The second involves the use of hydride vapor phase epitaxy (HVPE) (Molnar *et al.*, 1997) to synthesize a very thick (200 μm) GaN atop the ELOG undercarriage; subsequently most of the substrate and HVPE-grown thick GaN are removed by polishing leaving an about-150-μm-thick free-standing GaN substrate. In this case, the HVPE layer is assumed to direct residual dislocations towards the edge of the wafer. In the third instance, the substrate/template growth continues atop the HVPE GaN with another ELOG step which is then ready for the heteroepitaxy of the active device. In this case also, the backside of the device is polished to remove the substrate, including some of the HVPE GaN, to form a free-standing device film so that cleaving techniques can be used to form a laser cavity with facets in the $\{1\bar{1}00\}$ direction. Cathodoluminescence images show how the dislocations are reduced with the increased complexity of the ELOG-based substrate/templates, specifically in terms of their density in GaN above the SiO₂ mask window region. With the laser devices grown on the three

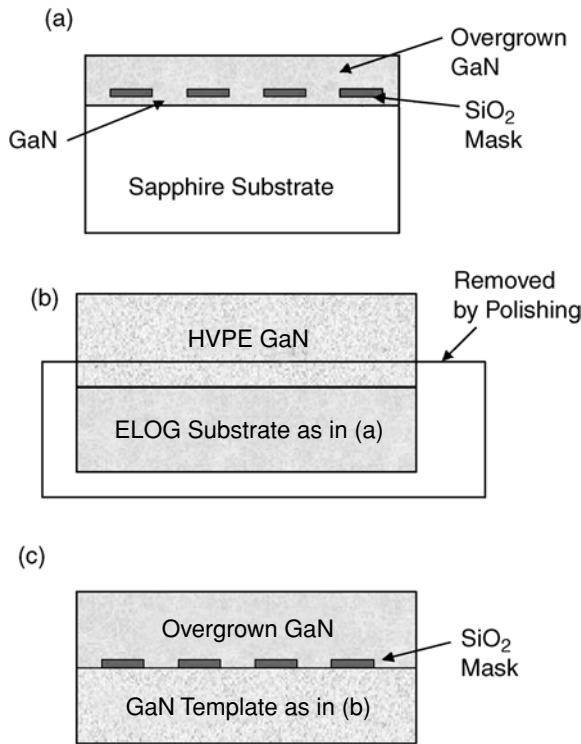


Figure 10.5: Schematic of ELOG substrate/templates for InGaN MQW diode laser, at varying levels of complexity: (a) the basic ELOG approach; (b) thick HVPE overgrowth and the removal of the initial ELOG template; (c) repeating the ELOG step on the template of (b). (After Nagahama *et al.* (2000).)

substrate/templates, the lifetimes at 30 mW average power were reported to be 700, 300, and 15 000 h, respectively at a device case temperature of 60 °C (Nagahama *et al.*, 2000). This comparison dramatically illustrates how important it is to reduce dislocations in order to achieve truly long-lived violet diode lasers. Even so, the ELOG intensive approach still leaves a presumed dislocation density (say, $>10^4 \text{ cm}^{-2}$) which would be considered totally unrealistic for a viable conventional III–V diode laser.

Given the substantial heat that is generated in the present diode lasers (typically in excess of 500 mW), the presence of the sapphire substrate presents another hindrance in the development and flexible application of the InGaN MQW violet laser. The thermal conductivity of sapphire at room temperature is about $50 \text{ W m}^{-1} \text{ K}^{-1}$, compared with about $130 \text{ W m}^{-1} \text{ K}^{-1}$ for GaN and $490 \text{ W m}^{-1} \text{ K}^{-1}$ for SiC. Moreover, the nonconducting sapphire prevents the present edge-emitting diode laser structures from being flip-chip mounted for heatsinking since both the *n*- and *p*-type contacts reside on the same side of the device (see next section).

Hence, the introduction of the HVPE growth technique for synthesis of very thick ($>100\text{ }\mu\text{m}$) GaN buffer layers offers a solution to the management of both the thermal budget and conductivity problems. Nakamura and coworkers first adapted the approach to separate InGaN QW heterostructures with their thick GaN buffer, to transfer laser devices onto another “artificial” host substrate. Other techniques have been developed for separating the nitride heterostructure from its sapphire substrate, opening another avenue for device design. We will return to this subject in Chapter 11 in connection with vertical cavity optical structures.

10.2.2 Diode laser fabrication and performance

In this section we first give a schematic guideline to InGaN MQW diode laser device fabrication, while noting that the details of diode laser processing are proprietary at industrial laboratories. Figure 10.1 showed an artist’s view of the device structure, less the substrate/template details, that typify the present nitride blue laser grown on a sapphire substrate. Note how the electron injection involves lateral transport through the bottom n -type GaN layers.

Briefly, the SCH MQW InGaN/AlGaIn heterostructure material is fabricated into index-guided mesa structures, usually by employing dry-etching techniques. Electron cyclotron resonance plasma etchings chemically-assisted ion beam etching, and standard reactive-ion etching, all of which use chlorine chemistry, are known to produce acceptable etch quality for the vertical walls of the lateral (ridge) waveguide as well as the resonator end facets. Figure 10.6(a) shows an SEM image of an approximately $5\text{-}\mu\text{m}$ wide etched ridge (mesa), demonstrating quality processing (Song *et al.*, 1998). The etched n -GaIn surface onto which metallization is applied for the n -type contact likewise has an acceptable electronic quality following properly applied dry etching. As already noted, in the case of the HVPE grown thick buffer layers, the optical resonator can be formed also by cleaving of free standing $\sim 100\text{-}\mu\text{m}$ films, following the (laborious) removal of the substrate/template. Figure 10.7 illustrates schematically the principal steps in the process flow for the fabrication of diode laser devices. The steps are largely routine in terms of the general strategy and implementation in the fabrication of other (that is, II–VI) wide-bandgap semiconductor diode lasers. For discussion of the contacts, we refer the reader to Chapter 9.

There has been a wide variation in the few reported values for cavity losses in the InGaN MQW diode lasers, in part due to the uneven pace of device development between various laboratories. Extracting the loss coefficient from threshold current density variation as a function of resonator length and/or facet reflectivity suggests, roughly, a typical value of $20\text{--}50\text{ cm}^{-1}$ for the optical losses. Some of this should

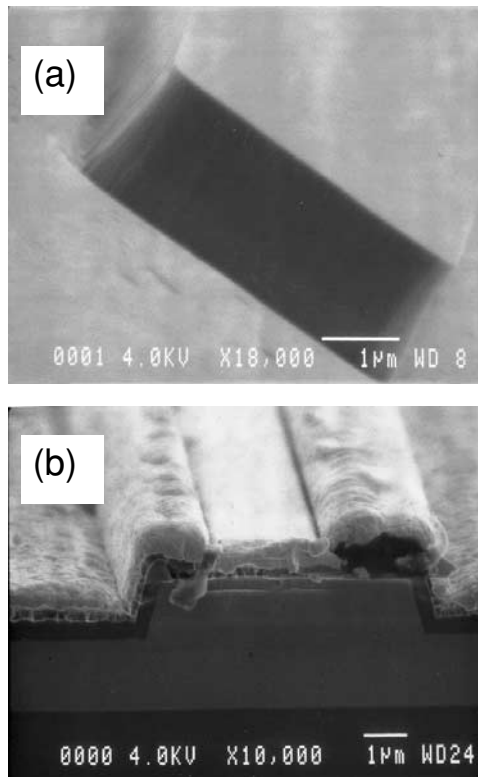


Figure 10.6: (a) Illustration of the use of electron cyclotron resonance plasma etching to define the ridge waveguide and the resonator end facets; (b) SEM image of a processed InGaN diode laser with cleaved facets; grown on SiC substrate (Song *et al.*, 1998).

be apportioned to the quality of the end mirror facets. In Figure 10.6(b), we show an SEM image of a completed laser device where the cavity facets have been formed by cleaving a structure grown on a SiC substrate, leading to cw operation (Song *et al.*, 1998).

We now turn to the device performance characteristics of the InGaN MQW violet laser, operating in the 405–415-nm wavelength range. Figure 10.8 shows the voltage–current characteristics and the light output per uncoated facet of a cw InGaN diode laser at room temperature (Nakamura *et al.*, 1998), grown on a sapphire substrate. The devices were ridge waveguide structures with dimensions of $2 \times 600 \mu\text{m}$, and could be augmented by reflective facet coatings. The threshold current density was approximately 4 kA cm^{-2} , at a voltage of 5.5 V (under the uncertain assumption of a uniform current flow). A slope efficiency (differential) as high as 1.0 W A^{-1} has been obtained from such devices by the Nichia group. Output powers exceeding 50 mW were observed under high injection ($\sim 70 \text{ mA}$),

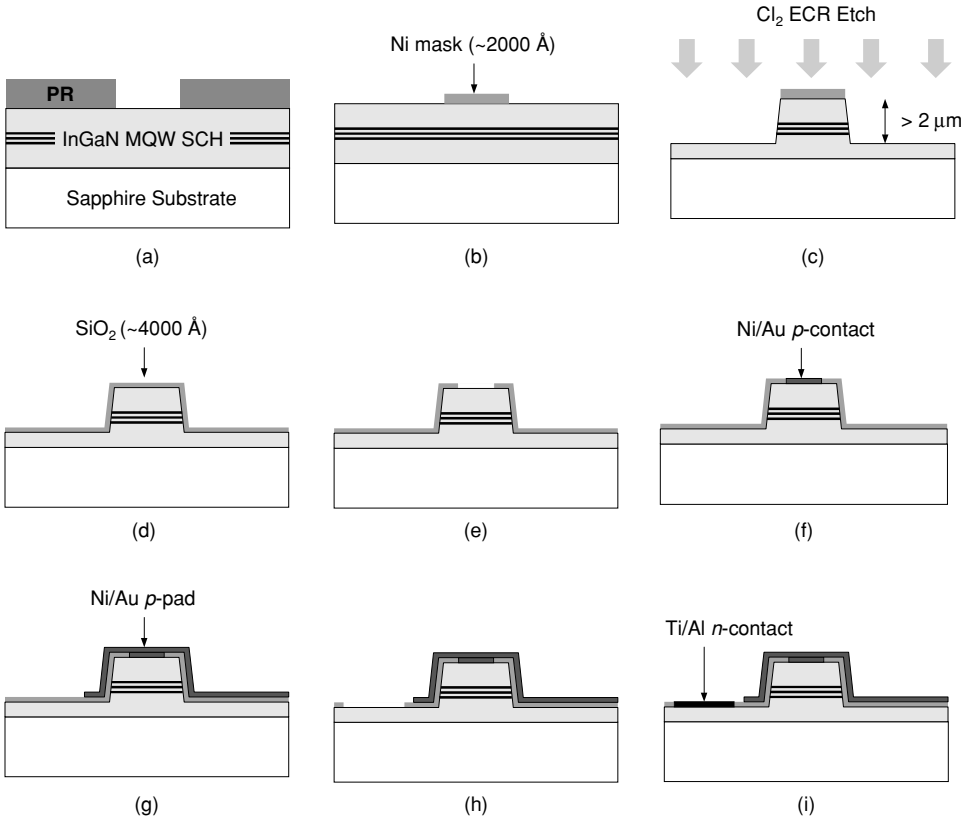


Figure 10.7: Schematic of a typical process sequence for an InGaN MQW diode laser: (a) mesa pattern; (b) Ni evaporation: lift-off; (c) mesa etching: electron cyclotron resonance; (d) SiO_2 deposition: PECVD; (e) p -window: BHF; (f) p -contact formation: lift-off; (g) p -pad formation: lift-off; (h) n -window opening; (i) n -contact formation: lift-off.

reaching the level required for the writing process in most current optical disk media. Due to the small ridge width, a rather stable fundamental transverse mode operation was possible, as shown in the far-field patterns of Figure 10.9. Near-field imaging of the InGaN MQW diode lasers has been reported from a number of laboratories to show how issues of lateral optical confinement need to be reconciled, and eventually optimized, with the constraints imposed by the lateral electrical injection (electrons) requirements. In particular, finite optical leakage and penetration to the n -contacts occurs relatively readily. More recently, by employing the ELOG-based substrate/template approaches, some of the cited performance parameters have improved, though we hasten to reiterate that the principal advantage of this approach is to reduce the device degradation. Threshold current densities have been reduced to the range of $2\text{--}3 \text{ kA cm}^{-2}$ in leading laboratories (Nichia, Sony, Xerox) and the slope efficiency increased up to about 1.5 W A^{-1} in best cases. The threshold

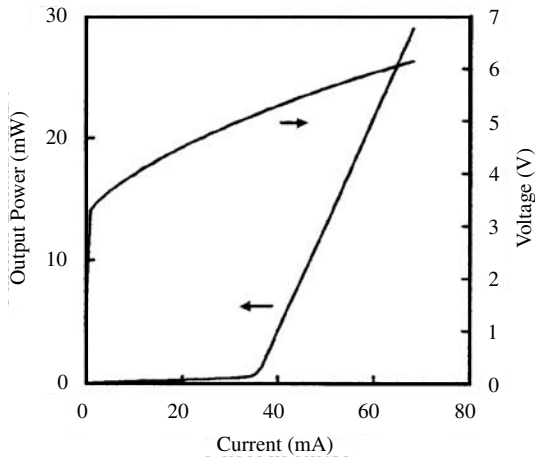


Figure 10.8: I - V characteristics and the optical power output per uncoated facet of a cw InGaN diode laser at room temperature. (From Nakamura *et al.* (1998).)

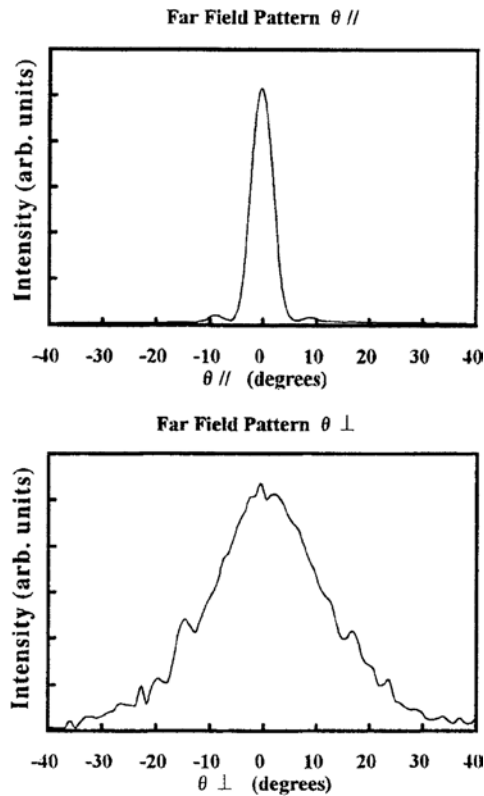


Figure 10.9: Far field emission pattern of InGaN MQW SCH/MQW diode laser in the planes parallel and perpendicular to the junction at 5 mW power. (From Nakamura (1999).)

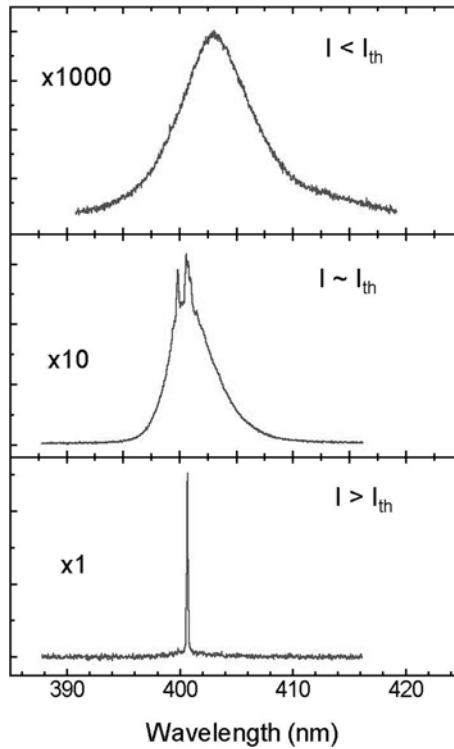


Figure 10.10: Emission spectrum below and above lasing threshold for an InGaN MQW diode laser grown on SiC substrate (Song *et al.*, 1998).

voltages likewise have dropped to the 4–5 V range, still dictated largely by the quality of the *p*-contacts. For operation at an output power level of about 30 mW (and with the cavity equipped with one high-reflectivity coating applied to the nonoutput facet), operating currents for $2 \times 600 \mu\text{m}$ sized ridge (index) waveguide devices are typically about 50 mA.

An example of the emission spectrum below and above lasing threshold is shown in Figure 10.10 for an InGaN MQW diode laser grown on SiC substrate (Song *et al.*, 1998), displaying a single longitudinal mode. In general, closer spectral analysis of the InGaN diode laser output frequently shows a complex longitudinal mode pattern where the presence of subcavities is apparent (that is, whose length is less than the device physical length). It is likely that specific crystalline defects, such as cracks that are frequently induced within the growth of the AlGaIn waveguide layers, form accidental resonators that provide the dominant optical feedback, hence supplementing the external resonator. However, good progress is being made to create stable, single-longitudinal-mode blue and violet lasers, though such a single-mode aspect is not necessary for optical storage applications (in contrast to the single-transverse-mode requirement).

10.3 PHYSICS OF OPTICAL GAIN IN THE InGaN MQW DIODE LASER

One striking aspect of the operation of the present InGaN QW lasers concerns the estimated electron–hole pair densities which are deduced from the electrical injection conditions. For an estimated carrier lifetime of several nanoseconds, the bulk equivalent electron–hole pair density at threshold in excess of 10^{19} cm^{-3} is obtained (Suzuki and Uenoyama, 1996). In fact, Nakamura and Fasol (1997) quoted a density of $2 \times 10^{20} \text{ cm}^{-3}$ for the first Nichia cw laser, an astonishing figure by the standards of known semiconductor lasers. Such a high electron–hole pair density is only partly explained by the somewhat higher effective masses in terms of formation of population inversion, when compared with GaAs. On the other hand, since the effective masses in ZnSe and GaN are of comparable magnitude and the threshold current density as well as the electron–hole pair density in a ZnCdSe QW laser are nearly two orders of magnitude lower than for InGaN QW lasers (for review of these lasers, see Gunshor and Nurmikko (1997)), an important question arises concerning other factors that influence the operation of the InGaN device. For example, in strong contrast with the optical properties of the binary GaN, the optical characteristics of the InGaN QW system at the bandedge include a spectrally very broad luminescence emission, even at cryogenic temperatures (linewidth on the order of 100 meV for In concentration $x \sim 0.10$). Up to injection levels of at least 10^{18} cm^{-3} , luminescence spectroscopy (including time-resolved) shows evidence for localized electron–hole pair states that reflect the large energy range available for such localization (Chichibu *et al.*, 1997a, Narukawa *et al.*, 1997, Jeon *et al.*, 1996). The degree of localization increases more rapidly with In concentration than expected for a simple random alloy, suggesting lattice disorder for the thin QWs which is both structural and compositional. How this can influence the optical gain and the necessary level of electron–hole pair injection in an InGaN MQW diode laser is a central subject in this section.

The InGaN QW shows generally strong departures from a usual random alloy. A propensity for In clustering has been at least qualitatively established via many microprobe studies, the origin of which is the finite degree of thermodynamic immiscibility of the InN and GaN constituents in a normal solid solution. The clustering, or “compositional anomalies”, in the InGaN system in turn profoundly affect the bandedge electronic states which form the “electronic power supply” for optical emission, for both LEDs and diode lasers. In earlier literature, Nakamura and colleagues speculated that InGaN QWs might even be subject to spontaneous “quantum dot” formation (Narukawa *et al.*, 1997), the implication being that diode laser action could derive a benefit in such a blue nitride quantum dot laser in a manner already clearly demonstrated in “conventional” III–VI self-assembled systems, most notably the InAs/GaAs strained layer case.

We now consider how the compositional anomalies and possible structural disorder in InGaN QWs lead to a veritable competition of electronic excitations between localized and extended electronic states. A key issue is the nature of those bandedge electronic states that supply the requisite optical gain, given the demonstrably large departure of InGaN from a random alloy in terms of the In concentration fluctuations (x_{In}) on a microscopic (atomic) scale. Typically, the mean values of x_{In} in the laser devices are in the range of ~ 0.1 – 0.2 ; *ad hoc* arguments can be made for the probable partial cation segregation due to the differences in the bond energies and lattice constants for the InN and GaN binary endpoints. High-resolution electron microscopy studies have shown that “clustering” takes place in the InGaN QW and the term “quantum dot” has been applied to such clusters (Narukawa *et al.*, 1997). When this type of nanoscale heterogeneous semiconductor forms the active laser material, a question that arises concerns the competition between localization and many-electron correlations within the available electron–hole pair states, given the very high pair densities required even in the best present devices to reach lasing threshold ($>10^{19} \text{ cm}^{-3}$).

Early on, a number of reports appeared on spectroscopic features of (undoped) InGaN QW at the bandedge, including the optical gain regime. Gain experiments were performed by optical pumping, especially by Hangleiter and coworkers, usually by the so-called stripe excitation method (Frankowsky *et al.*, 1996), which yields a limited spectroscopic view and may not accurately correspond to the conditions in an electrically-injected device. Nakamura and coworkers have used the Hakki–Paoli method to acquire gain spectra on actual diode laser devices (Nakamura and Fasol, 1997) and an electrical injection-optical probe method has been applied to study the formation of optical gain in InGaN QW p – n junction heterostructures (Kuball *et al.*, 1997).

Significant insight into the optical gain spectra of the InGaN laser active medium has been acquired subsequently, based on the analysis of the spontaneous emission spectra of the diode laser, in conjunction with its threshold characteristics. This approach makes use of the fundamental relationships between spontaneous emission, stimulated emission, and absorption. This method, initially introduced by Henry *et al.* (1980) to semiconductor lasers has been applied to the detailed study of different III–V QW semiconductor lasers (Kesler and Hardy, 1990, Blood *et al.*, 1991, Gershoni *et al.*, 1993), as well as to the II–VI green-blue diode lasers illustrated later in this chapter (Ding *et al.*, 1994). The technique has important advantages over the Hakki–Paoli technique, including the problem in the latter method of separating the nonradiative component of the injection current from the radiative one and the inability to probe the absorbing regions of the spectrum at injection levels well below the lasing threshold. The application of the Hakki–Paoli method is further complicated for the InGaN diode lasers due to the frequent presence of extraneous longitudinal subcavity modes. For a gain medium possessing such idiosyncrasies as

the InGaN QW, the “Henry” approach is advantageous, given the central question of the fundamental gain characteristics and their relationship with radiative processes.

First gain spectroscopic measurements on actual InGaN QW diode lasers ($x = 0.15$) were made by Song *et al.* (1998), to show the pronounced extension of the gain spectra associated with the $n = 1$ QW transition into the low-energy region. At threshold, finite gain was found to be as much as 200 meV below its peak position, indicating a degree of broadening which is most uncharacteristic of common semiconductor lasers. A peak gain coefficient of approximately 3200 cm^{-1} was measured. Subsequently, Kneissl *et al.* (2000) measured the gain spectra over a range of indium compositions for the practical operating range of the violet InGaN diode laser, shown in Figure 10.11, where the reader’s attention is directed to the

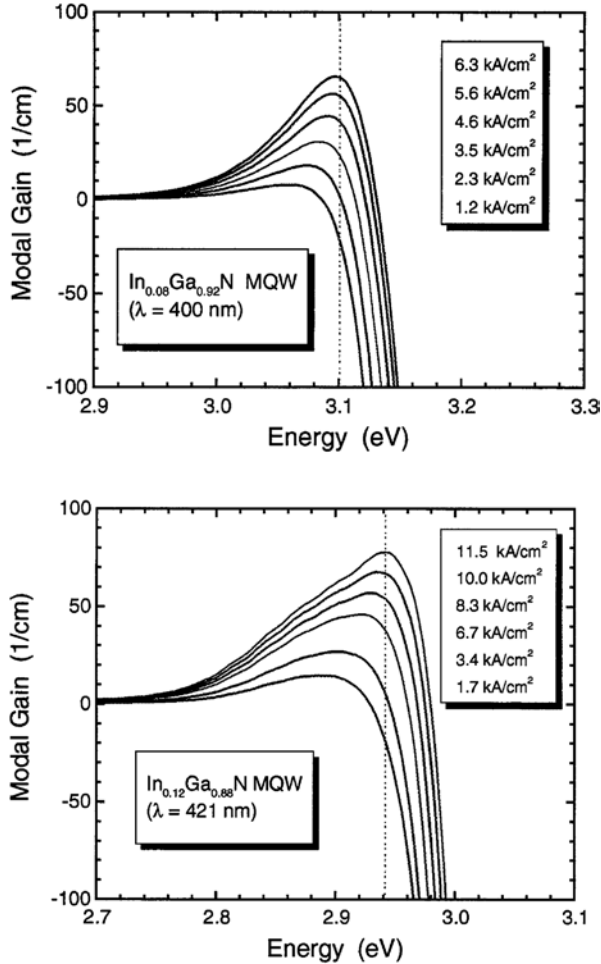


Figure 10.11: Gain/absorption spectrum of a InGaN MQW diode laser of two different In compositions at varying injection levels at room temperature. (From Kneissl *et al.* (2000).)

dependence of the gain bandwidth on the composition. As shown by Song *et al.* (1998), with increasing injection one reaches the transparency condition relatively easily at levels of injection that were not very different from that of the conventional LED regime. When the current is increased further, gain builds up over the large spectral range, indicative of the participation of a corresponding range of electronic states. The position of the peak gain blue shifts somewhat at higher injection levels, but considerably less than anticipated from a one-electron-state-filling picture, possibly due to many-body bandgap renormalization effects. Qualitatively, we may now understand one reason for the high electron–hole pair density required for laser operation, apart from extrinsic reasons such as unwanted optical losses. That is, while the spectrally integrated gain is, in fact, quite large, its peak value (determining the lasing threshold) is much diluted at the expense of the remarkably large broadening.

These observations provide an extension to arguments (Chichibu *et al.*, 1996, 1997, Narukawa *et al.*, 1997, Kuball *et al.*, 1997) that radiative recombination processes at the lowest interband transition in the InGaN QW are profoundly influenced by localized electron–hole pair states at room temperature, within an energy range that is nearly an order-of-magnitude larger than estimated for a simple random alloy. That is, the description of the system in terms of weak disorder, as usually applied to ternary and quaternary compounds in the III–V and II–VI semiconductors, is probably inapplicable. Whether this aspect of InGaN is intrinsic or subject to specific MOCVD growth conditions has yet to be established; suffice it to say that all available optical data on InGaN QWs and thin films to date display the striking extension of the bandedge states so that, for example, excitonic features in absorption at the $n = 1$ QW states have not been unambiguously identified. By contrast, gain spectroscopy performed on wide-gap ZnCdSe QW diode lasers by the method described below shows very clearly the characteristic influence of the strong excitonic enhancement of the peak gain and an overall optical response at the $n = 1$ heavy-hole exciton with the pronounced Coulomb correlations in evidence. Such effects are clearly masked by the disorder contributions in the InGaN QW, making it difficult to isolate predicted many-body interactions (Chow and Koch 1995, Chow *et al.*, 1997) in the dense electron–hole system within the active region of the blue diode laser.

To summarize, experiments show how filling of the localized states is a necessary prerequisite to the build up of a sufficient population inversion for threshold gain in the present devices. On the other hand, since transparency is reached at a rather low injection level ($n = p \sim 10^{18} \text{ cm}^{-3}$) it may be possible to reduce the threshold current by designing a laser resonator with very low optical losses. The near “clamping” of the Fermi level E_F at higher injection may be due to a significant increase in the effective density of states. However, the question that we will defer

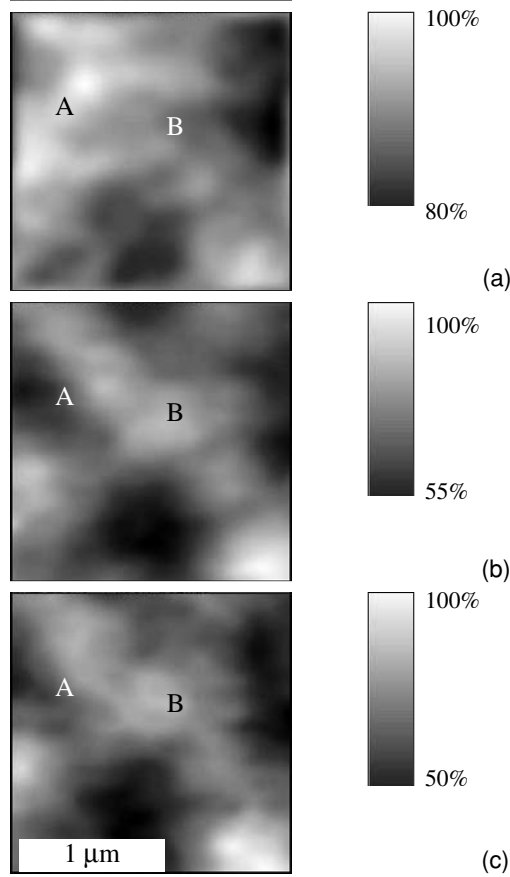


Figure 10.12: Collection-mode NSOM photoluminescence from an undoped InGaN/GaN MQW for three different wavelengths. Traces (a), (b), and (c) correspond to NSOM photoluminescence images at $\lambda = 450$ nm (2.75 eV), $\lambda = 460$ nm (2.70 eV), and at $\lambda = 470$ nm (2.64 eV), respectively. Markers “A” and “B” highlight regions of complementary optical contrast at different emission wavelengths. (From Vertikov *et al.* (1998)).

to the next section is whether these states are still localized or extended. *We wish to emphasize that the issue of the In compositional anomalies and QW structural disorder increase in severity very rapidly as the In concentration reaches about $x_{In} = 0.1$ and beyond; in fact for $x_{In} \ll 0.1$, InGaN behaves very nearly as a random alloy.* The work at Xerox PARC laboratories (Kneissl *et al.*, 2000), in the author’s group at Brown University, and elsewhere has shown how the gain spectra does indeed significantly narrow as the In concentration is reduced, for lasers operating in the violet ($\sim 395\text{--}405$ nm). On the other had, to maintain adequate electronic/optical confinement, one then needs to increase the Al concentration in the cladding layers, adding a different type of materials science challenge.

10.3.1 On the electronic microstructure of InGaN QWs

In the case of LEDs, there is general agreement that the pronounced compositional anomalies encountered in InGaN (in the sense of non-Poissonian cation distribution) as well as other QW disorder are actually beneficial in enhancing the overall light emission efficiency. This is so because of the strong localization that accompanies the compositional fluctuations on the spatial scale of the electron and hole Bohr radii. Evidence for this is the presence of soft absorption edges, large spectral redshifts (“Stokes shifts”) between absorption and luminescence, and the long ($>$ nanosecond) room-temperature lifetimes that show strong dispersion. However, the presence of the large average strain and local fluctuations in the strain, coupled with the exceptionally large piezoelectric coefficients in GaN/AlGaN heterostructures (Im *et al.*, 1998), make it difficult to determine the specific contributions of these effects to the nature of near-bandedge states existing in InGaN laser devices. Furthermore, the defect landscape arising from extended defects is subject to significant variations depending on the growth details, complicating an already camouflaged picture further. The case of the piezoelectric fields is particularly topical at this writing, in that the estimated piezofields (up to megavolts per centimeter) not only influence the energies of the conduction and valence band edges sizably (~ 100 meV) but can have a direct impact on the dilution of the electron–hole overlap and interaction. The former is of relevance to the one-electron matrix element, while the latter influence many-body electronic phenomena, specifically the excitonic effects described at the end of this chapter. Hangleiter and coworkers have studied the optical transitions in undoped InGaN QWs by time-resolved photoluminescence to argue that the observed spectral shifts are consistent with the built-in piezoelectric fields (Kollmer *et al.*, 1999). In an InGaN MQW diode laser, the high electron–hole pair density will, of course, significantly screen the piezoelectric contributions, but so far there are no detailed experimental studies to address this question in the actual devices.

There have been a number of experimental reports, employing microscope and microprobe techniques, that have provided real images of the In aggregation in InGaN epitaxial thin films and QWs. At one end of the spatial spectrum, Kisielowski *et al.* (1997) have employed the atomic resolution transmission electron microscopy scale to show evidence of clustering on the scale of a lattice constant in connection with studies of strain in the InGaN/GaN QWs. At the other end of the apparent size distribution, both cathodoluminescence (Chichibu *et al.*, 1997a) and near-field optical microscopy (NSOM) (Crowell *et al.*, 1998, Vertikov *et al.*, 1998) have been used to acquire luminescence-based images on a spatial scale on the order of 100 nm. Such a strikingly wide distribution of the cation aggregates contrasts very strongly with the random alloy behavior, where spatial compositional (and crystal potential energy) fluctuations are confined to the atomic scale. We note in

passing that the cathodoluminescence and the NSOM spatially-resolved techniques have been used to also show how the presence of threading dislocations and larger extended state defects such as nanopipes has little evident impact on the radiative efficiency of InGaN QWs (Liu *et al.*, 1996, Vertikov *et al.*, 1998a).

Among the reports on high-resolution microscopy of InGaN QWs and epilayers, Chichibu *et al.* (1997a) studied spectral variations of luminescence from InGaN/GaN QWs by cathodoluminescence at low temperatures. In that work, the mean In-cluster size was estimated to be less than 60 nm, though the cathodoluminescence images revealed In-rich (deficient) areas up to half a micron in diameter. NSOM is an attractive alternative to cathodoluminescence, allowing subwavelength resolution in photoluminescence measurements with energetically direct and accurately measurable carrier injection into the QW, together with simultaneous topographic imaging. Comparison of the spatially-resolved photoluminescence with other simultaneous optical microprobes, for example, reflectivity or transmission, can provide additional useful information. We show next an example of the use of *collection-mode* NSOM, where issues of carrier diffusion can be minimized when studying the local spectral variations in the photoluminescence emission on a sub-100 nm scale under high electron-hole pair injection such as encountered in a diode laser (Vertikov *et al.*, 1998).

Typical near-field photoluminescence images taken on four undoped 30 Å thick $\text{In}_x\text{Ga}_{1-x}\text{N}$ QWs with 90 Å thick GaN barriers are shown in Figure 10.12 for three different wavelengths of emission ($x \approx 0.2$). The injection of electron-hole pairs ($\sim 10^{19} \text{ cm}^{-3}$) into the QWs was made to occur resonantly by the choice of the excitation wavelength. Figure 10.12(a) shows the NSOM image recorded on the higher-energy side of the far-field photoluminescence spectrum at $\lambda = 450 \text{ nm}$ ($\hbar\omega = 2.75 \text{ eV}$), Figure 10.12(b) was recorded at the center of the spectrum at $\lambda = 460 \text{ nm}$ ($\hbar\omega = 2.70 \text{ eV}$), while Figure 10.12(c) is the NSOM image taken at $\lambda = 470 \text{ nm}$ ($\hbar\omega = 2.64 \text{ eV}$) on the lower-energy side of the photoluminescence spectrum (Vertikov *et al.*, 1998). The images reveal darker and brighter regions several hundred nanometers in extent. They strongly suggest that the light emission in the room-temperature InGaN MQW lasers occurs from the extended states, once localized states are filled. The gain spectroscopy on InGaN QW diode lasers discussed above shows that it is necessary to fill a large number of band tail states, over an energy range exceeding 100 meV, before the electron-hole pair quasi-Fermi level has reached a value at which the optical gain coefficient is sufficient for the onset of laser action (typically $g_{th} > 3000 \text{ cm}^{-1}$). The excitation and position dependences of the NSOM spectra in the upper and lower diagrams, respectively, Figure 10.13 give another view of this state-filling process. A significant part of the photoluminescence spectra in each particular point of the sample is broadened *homogeneously* (on the scale of the instrument resolution), and one can argue that

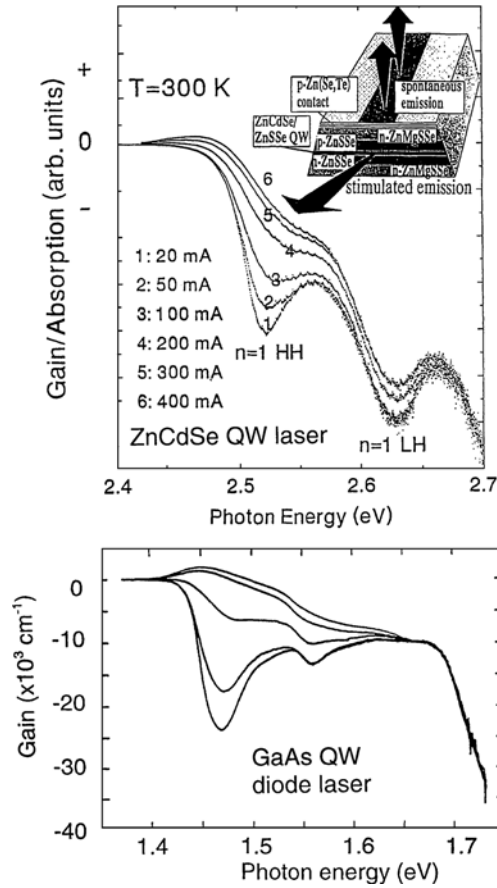


Figure 10.13: Comparison of the gain/absorption spectra for SQW ZnCdSe and GaAs SCH diode lasers at room temperature, as a function of injection current (upper and lower traces, respectively), in the vicinity of the $n = 1$ HH (heavy-hole) and LH (light-hole) transition. (From Ding *et al.* (1994) and Kesler and Hardy (1990).)

all localized states and a significant number of extended states, perhaps “mixed” with higher-energy localized states, contribute to the local radiative recombination under high injection levels. Also, sites with larger local bandgap (for example, Indeficient regions) can easily become interconnected through the carrier diffusion.

A different application of the NSOM imaging technique has been used to measure the electron–hole (ambipolar) diffusion in InGaN MQWs under optical injection (Vertikov *et al.*, 1999). The technique involves the setting up of an excitation interference grating with two blue laser beams, on a spatial scale of about 200 nm, and the direct imaging of the photoluminescence intensity variations with the NSOM fiber tip as the high-spatial-resolution light collector. In these types of experiments, one studies the grating contrast in the photoluminescence profiles; this

is diminished due to carrier diffusion. Numerical fitting of such photoluminescence profiles with the solutions of the diffusion equation for electron–hole densities

$$\Delta n(x,t) = \Delta p(x,t) \propto \left[e^{-t/\tau} + \gamma_0 e^{-(t/\tau)(1+(KL_D))^2} \cos(2\pi x/\Lambda) \right]$$

then gives diffusion length L_D . Here, τ is the (carrier-density-independent) recombination lifetime, γ_0 is the contrast parameter of the excitation grating, and Λ is the grating period. Vertikov *et al.* (1999) studied a number of InGaN MQW samples grown at different laboratories to find that the diffusion lengths at room temperature could vary very widely under low electron–hole injection, presumably reflecting the individual microstructure of a particular MQW. On the other hand, in the high-injection regime typical of a blue diode laser ($>10^{19} \text{ cm}^{-3}$) the values of diffusivity converge to those roughly expected for a “free” electron–hole gas.

10.3.2 Excitonic contributions in green-blue ZnSe-based QW diode lasers

In terms of basic optical properties, the wide-bandgap semiconductors are exceptional in the strength of the excitonic effects that dominate the linear optical response. Exciton binding energies exceeding $E_x > 40 \text{ meV}$ have been measured for ZnCdSe QWs and the nitride compounds are expected to possess considerably larger values. Noting that the exciton oscillator strength scales roughly as the square of E_x , the robustness of two-dimensional excitons (against screening and thermal dissociation) implies a significant contribution to the optical gain even at room temperature. While the compositional anomalies in InGaN QWs have made it very difficult to isolate excitonic features in their optical spectra to date, the heteroepitaxial quality of II–VI structures has allowed a good deal of such spectroscopy to be performed. Thus we have chosen to include selected examples of this important subject in this chapter, using work done with the II–VI materials as illustrative examples. In particular, both excitonic and biexcitonic spectrally distinct features have been clearly identified in ZnSe-based QWs up into a high-density regime at cryogenic temperatures where stimulated emission and laser action commences. We concentrate, in particular, on the room-temperature ZnCdSe QW diode laser device which is demonstrably subject to pronounced electron–hole Coulomb effects.

In a high-density electron–hole system (two-dimensional), the direct electron–hole Coulomb interaction is accompanied by exchange and correlation effects which profoundly increase in importance with the particle density. Physically, the two extreme limits are easily identified, for example, in a GaAs bulk crystal or quantum well: the low-density bound state (exciton) regime and the high-density electron–hole plasma. For the latter, the exchange and correlation effects are seen mainly through the bandgap renormalization effect which shifts the emission wavelength of such a semiconductor laser to longer wavelengths (by up to several tens

of meV). In terms of the actual gain spectrum (lineshape or magnitude), however, the many-body effects induce only rather small corrections to calculations made in the one-electron picture that usually include some phenomenological damping rates. Sophisticated theoretical approaches, converging to the semiconductor Bloch equations, have been applied to describe the gain spectra of various III–V lasers with impressive success (Haug and Koch, 1993) and these methods have been tested with the wide-gap semiconductors (Chow and Koch, 1995, Ell and Haug, 1989). While referring the reader elsewhere for expert discussion of the power of the semiconductor Bloch equations, we note here that the approach might be qualitatively described as having its starting point in the electron–hole plasma limit to which the electron–hole Coulomb interactions are added. In contrast, we adopt a viewpoint that takes the opposite limit as a starting point, that is, a gas of noninteracting (or weakly interacting) excitons subject to many-body interactions. This pathway is natural for an experimentalist when considering, for example, the ZnSe QW or future GaN QW laser in which the temperature is gradually increased from liquid helium to room temperature and where the role of bound electron–hole pair states at low temperatures is beyond dispute. Approaching the problem from this limit presents, however, significant challenges to the semiconductor Bloch equation approach, for example, due to the difficulty of including the screening by the bound states into the formalism. Yet evidence in the ZnCdSe QWs strongly suggests that the Coulomb correlations remain very potent, when compared, for example, with an GaAs laser.

Chronologically, localized excitons were first invoked by Ding *et al.* (1992) to define optical gain in the ZnCdSe QW, with supporting and complementary evidence reported by other groups (Kawakami *et al.*, 1994, Alferov *et al.*, 1994), especially by Cingolani *et al.* (1996). Here we consider the experimental evidence in the case of a room-temperature electrically injected green-blue diode laser in an attempt to gauge the physical role of the electron–hole pair correlations in a practical wide-gap semiconductor device.

Figure 10.13 compares gain/absorption spectra for a ZnCdSe (Ding *et al.*, 1994) and GaAs SQW diode laser in the vicinity of the $n = 1$ QW heavy-hole exciton transition (Kesler and Hardy, 1990) obtained in each case by the same experimental technique of correlating edge-stimulated emission with top spontaneous emission. Note how, as a function of increasing current injection, the heavy- and light-hole exciton features are rapidly bleached in the GaAs QW so that at laser threshold any semblance to excitonic resonances is absent. This is in striking contrast with the case for the II–VI laser, where a partially bleached heavy-hole exciton resonance and a nearly intact light-hole resonance are clearly present at laser threshold, with gain appearing some 60 meV below the heavy-hole exciton resonance. While in broad agreement with predictions of the semiconductor Bloch equations (Ell and

Haug, 1996), important differences remain concerning the details of the gain spectrum. In the authors' opinion the microscopic details of the system, in terms of the appropriate many-body description, remain theoretically incomplete for now. For some phenomenological ideas and interpretations, as well as experimental details leading to Figure 10.13, the reader is referred to Ding *et al.* (1994). The role of the Coulomb interactions remains practically relevant, for example, in the context of present efforts to fabricate wide-bandgap semiconductor quantum wire lasers where the bound exciton states might be expected to be increasingly stable against dissociation and screening. In any event, the typical range of enhancement of the peak gain for room-temperature ZnCdSe QW lasers is in the range of a factor of 3–4, clearly an important positive feature in designing low-threshold devices. In considering the laser threshold condition, we remark that while the excitonic enhancements also imply an accelerated spontaneous (radiative) decay rate, the concentration of oscillator strength in the gain spectrum near the $n = 1$ heavy-hole exciton resonance specifically enhances the peak gain coefficient, whereas the spontaneous emission rate is proportional to the integral of the corresponding spontaneous emission spectrum.

As yet another experimental illustration of the impact of the electron–hole Coulomb correlation at room temperature, we mention results where ZnCdSe QW diode lasers have been studied in a high magnetic field, applied perpendicular to the QW layer plane (Song *et al.* 1997). In this study $\text{Zn}_{1-x}\text{Cd}_x\text{Se}/\text{Zn}_{1-x}\text{S}_x\text{Se}/\text{Zn}_{1-x}\text{Mg}_x\text{S}_y\text{Se}_{1-y}$ SCH SQW diode laser structures were used with a rather large Cd fraction to ensure strong electron–hole confinement in the QW. This choice of QW design parameters was made in order to approach the quasi-two-dimensional limit, that is, to enhance the exciton binding energy, which for this concentration range of Cd is estimated to be at least 40 meV, based on earlier studies on the ZnCdSe/ZnSSe QW (Pelekanos *et al.*, 1992).

The spectrum of an index-guided diode laser device operating in a single longitudinal mode is shown in Figure 10.14 in magnetic fields up to 29 T (Song *et al.*, 1997), where we see the mode hopping occurring as discrete jumps between the photon energies of $\hbar\omega = 2.2899$ eV (for $B = 0$ –18 T), $\hbar\omega = 2.2901$ eV ($B = 18$ –28 T), and $\hbar\omega = 2.2921$ eV ($B = 28$ –29 T). The diode laser was operated well above threshold at an injection level of $I = 18$ mA ($\sim 1.4I_{th}$, where $I_{th} = 13$ mA). From the measured room-temperature electron–hole lifetime under comparable injection (Ding *et al.*, 1994), we calculate an electron–hole pair density of $1.9 \times 10^{12} \text{ cm}^{-2}$ in the ZnCdSe single QW. In a free-carrier picture, a Landau shift, linear in the magnetic field and of the order of $\Delta E_L \sim 0.5 \text{ meV T}^{-2}$ would be expected (due to either the shift of the lowest conduction band Landau level or the jumps of the Fermi level between such occupied levels). Clearly, the experimental result is about one order of magnitude smaller and incompatible with free electron–hole behavior. On the other hand, the

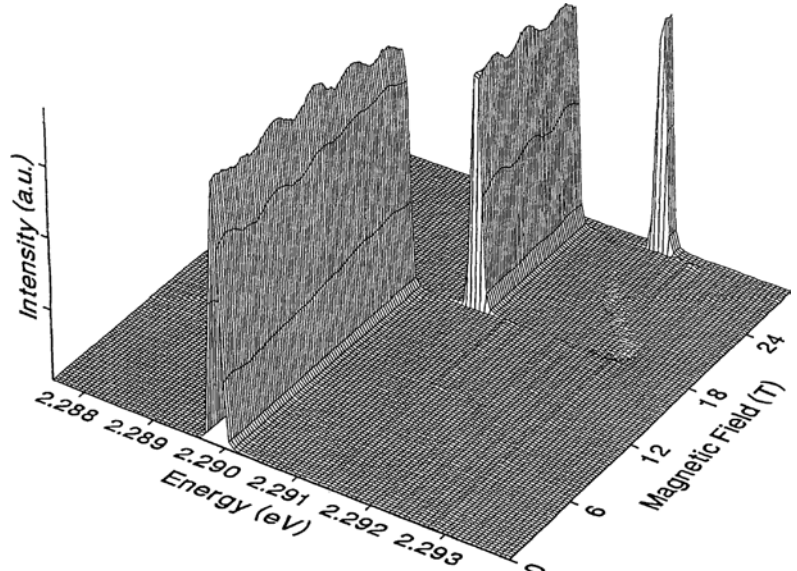


Figure 10.14: Emission spectra of a single-mode ZnCdSe QW diode laser in magnetic fields up to 29 T at room temperature. (From Song *et al.* (1997).)

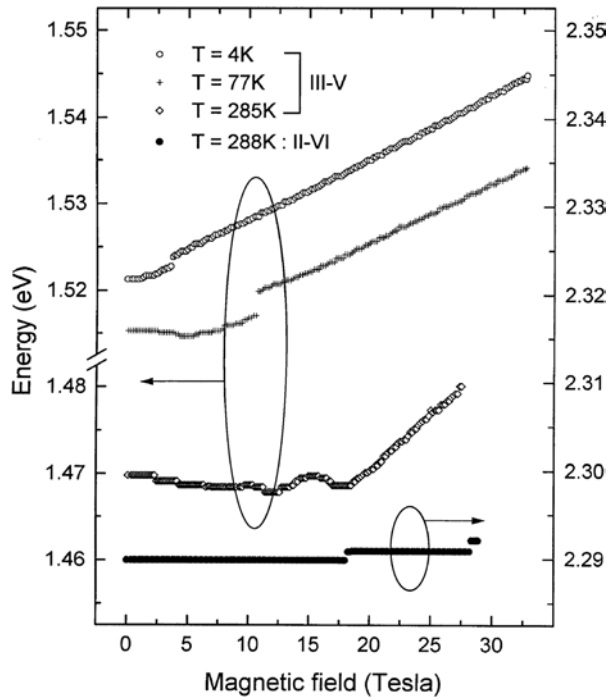


Figure 10.15: The emission photon energy for a GaAs QW diode laser (upper traces) and ZnCdSe QW diode laser (lowest trace) as a function of magnetic field, oriented perpendicular to the layer plane. (From Song *et al.* (1997).)

agreement is much better when compared with the diamagnetic shifts measured for the $n = 1$ heavy-hole exciton transition in ZnCdSe QWs, typically, $\Delta E_{dia} \sim 2 \mu\text{eV T}^{-2}$, with the reduced exciton mass of $\mu^* \sim 0.1m_0$. Given the exciton binding energy of about 40 meV, this implies that at $B = 30$ T, we are still in the regime where $E_x > \hbar\omega_c$, the electron cyclotron energy. In Figure 10.15 we compare the behavior of the ZnCdSe QW diode laser with GaAs-based QW diode lasers. In strong contrast, while exhibiting temperature-dependent “Landau-level jumping” (oscillations) of the Fermi level at low B fields (in terms of the Fermi-level), the spectral shifts in the emission of the GaAs QW diode laser in Figure 10.15 evolve into a linear cyclotron-like shift, with some influence of the many-body interactions on the effective masses evident, however.

10.4 SUMMARY

In this chapter we have examined the device science and engineering features of the violet edge-emitting InGaN MQW diode lasers, which are making their entry to the technological and commercial arenas. We have included the key current research issues of the high-threshold current density and the continued efforts to create artificial substrate templates for reducing the misfit (threading) dislocation density for improved device performance and lifetime. We have noted how, at a rather fundamental level, the InGaN alloy active QW medium has characteristic compositional (and structural) disorder that impacts the gain spectrum of the laser. This feature, which increases in seriousness with the indium concentration, may restrict the operation of the devices at practical threshold current densities to the violet, leaving the longer blue and green regions to await future developments, perhaps involving complementary material approaches. In the next chapter we consider the device prospects for a different device geometry for surface emitting lasers, based on vertical microcavities.

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11

Prospects and properties for vertical-cavity blue light emitters

11.1 BACKGROUND

VCSELs have gained importance in recent years for applications where beam quality, prospects for high-density arrays, and inherent compatibility with planar processing are particularly important. In the case of resonant-cavity LEDs (RCLEDs), the quasi-beam-like directionality in the spontaneous emission and possible enhancements to the radiative recombination rates likewise have spurred active research. VCSEL technologies that rely on III–V semiconductor heterostructures have now risen to a dominant position within the semiconductor laser industry, supplying high-performance components that play an increasingly vital role in optical communications technology. Both GaAs- and phosphide-based QW VCSELs are making significant headway in penetrating into the 1.3–1.5 μm wavelength region, following spectacular device successes in the roughly 650–900 nm range in the 1990s.

To date, the shortest wavelength VCSELs that have been implemented have reached the short end of the red (~ 630 nm). There are a number of reasons, both fundamental and practical, that make the development of blue and green VCSELs and RCLEDs in the wide-gap semiconductors challenging. In terms of the technological approaches and prospects for short-wavelength VCSELs and RCLEDs, this chapter is speculative in tone, given the early stages of research. At this writing, it is unclear what combination of epitaxial growth and device design/processing schemes might result in a technologically viable VCSEL, for instance. On the other hand, there are ample fundamental physical reasons that suggest that microcavity emitters based on wide-gap semiconductors, and the nitrides in particular, have special properties that offer unique opportunities both in terms of the basic physics and device performance. One intriguing open question relates to the microscopic physics of the stimulated emission, given earlier indications of the importance of the strongly coupled exciton–photon modes in ZnSe-based VCSEL structures (Kelkar *et al.*, 1997).

As the examples in preceding chapters illustrate, the nitride optoelectronic materials frequently stand apart from other III–V semiconductors in terms of their device-related properties. This fact extends to the device science and engineering of the vertical-cavity blue and near-ultraviolet emitters. True enough, the generic concepts of planar microcavity devices apply here; however, their detailed implementation departs appreciably from the longer wavelength III–V VCSELs and RCLEDs. We have already discussed the gain characteristics of InGaN MQWs in Chapter 10 which, at least in the present approaches to epitaxial growth by MOCVD methods, limit the wavelength range of edge-emitting lasers roughly to 400–430 nm.

From a device engineering point of view, the primary challenge presented by the nitride vertical-cavity emitters is two-fold. First, the fabrication of a high-Q optical resonator is nontrivial. Secondly, dictated mainly by the presently low conductivity of *p*-GaN and its alloys, the electrical injection schemes must incorporate lateral current spreading schemes. In the following, we consider these issues separately, while showing examples of solutions from the authors' laboratories. We note that important parallel work is being pursued, for example, at UC Santa Barbara, and wish to acknowledge important ideas put forth by K. Iga and his collaborators (Honda *et al.*, 1995). More recently, Mackowiak *et al.* (2001) have put forth innovative theoretical ideas concerning current injection into a nitride-based VCSEL.

11.2 OPTICAL RESONATOR DESIGN AND FABRICATION: DEMONSTRATION OF OPTICALLY-PUMPED VCSEL OPERATION IN THE 380–410-nm RANGE

One major challenge for a blue/violet nitride VCSEL concerns the choice of high-reflectivity mirror materials and the fabrication of the microcavity resonator structure. *In-situ* as-grown AlGaIn/GaN DBRs are feasible in principle, but, due to the small index-of-refraction contrast, require a large number of layer pairs. Nonetheless, several groups have demonstrated the epitaxial growth of such DBRs (Someya and Arakawa, 1998, Langer *et al.*, 1999, Krestnikov *et al.*, 1999, Ng *et al.*, 2000). Furthermore, there are reports that argue for the observation of vertical cavity or “surface” lasing under optical intense pumping from GaN or InGaIn MQW or thin-film heterostructures that are encased by *in-situ* grown AlGaIn/GaN DBRs (Redwing *et al.*, 1996, Krestnikov *et al.*, 1999). Arakawa and coworkers have shown stimulated emission employing a “hybrid” structure composed of one *in-situ* grown AlGaIn/GaN DBR and one dielectric DBR (Someya *et al.*, 1999).

11.2.1 All-dielectric DBR resonator

Here we illustrate optically-pumped VCSEL action near $\lambda = 403$ nm from an InGaN MQW heterostructure within an optical cavity formed by a pair of dielectric DBRs. One specific fabrication technique has been shown to produce microcavity resonators with quality factor Q approaching 1000 (Song *et al.*, 1999). Briefly, a multi-layer $\lambda/4$ stack of $\text{SiO}_2/\text{HfO}_2$ was first deposited on the surface of the InGaN/GaN heterostructure wafer by reactive ion-beam sputtering. The structure was then flip-chip mounted and bonded onto a permanent host substrate and the substrate separated by pulsed excimer laser ablation (Kelly *et al.*, 1997, Wong *et al.*, 1998). After appropriate chemical cleaning (ECR etch), a second $\text{HfO}_2/\text{SiO}_2$ multi-layer dielectric stack (DBR) was deposited directly onto the exposed AlGaIn layer surface for completion of the optical resonator. Figure 11.1 shows a cross-sectional scanning electron microscope (SEM) image of a completed structure where the nitride heterostructures included a MQW active region (10–20 QWs) $\text{In}_{0.1}\text{Ga}_{0.9}\text{N}$ quantum wells ($L_w = 30$ Å) with GaN barriers ($L_B = 50$ Å), surrounded by $\text{Al}_{0.07}\text{Ga}_{0.93}\text{N}$ upper and lower outer cladding layers. Growth was optimized to virtually eliminate crack formation and to provide a high degree of optical flatness despite the relatively thick AlGaIn layer. Good morphology is crucial to the realization of true VCSEL operation; AFM studies of both the surface of the as-grown heterostructure as well as the ECR-etched, excimer-laser-separated GaN layer

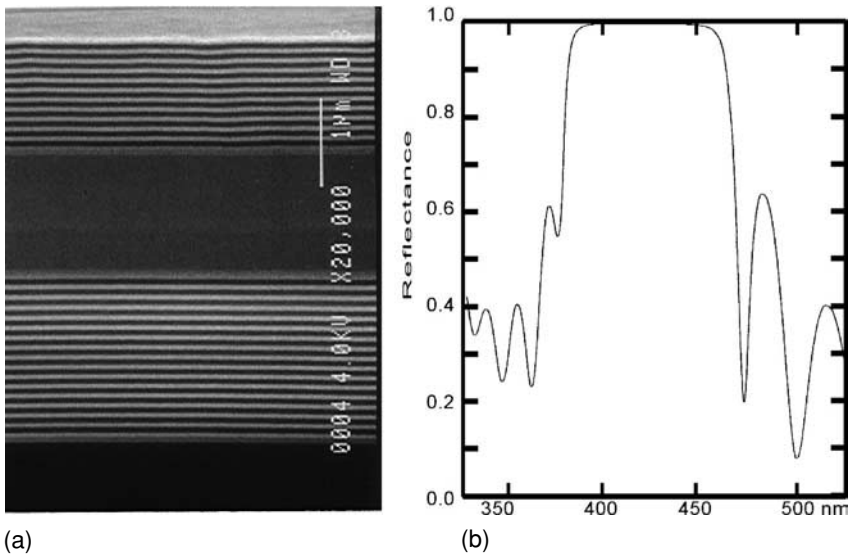


Figure 11.1: (a) Cross-sectional SEM image of a completed vertical cavity InGaIn QW structure. (b) Reflectance spectrum of a ten-layer pair reference $\text{HfO}_2/\text{SiO}_2$ DBR stack. (From Song *et al.* (1999).)

indicate a mean roughness of 2–3 nm over areas on the order of several hundred square microns. In the absence of good morphology, lasing can be obtained but is readily dominated by in-plane stimulated emission. Optically-pumped quasi-cw VCSEL operation was achieved by photoexcitation at 355 nm, outside the reflectance band of the DBRs and slightly below the bandgap of the AlGaIn cladding layer, ensuring a predominant creation of electron-hole pairs directly into the InGaIn QWs. Figure 11.2(a) shows the spontaneous emission spectrum at temperature $T = 258$ K at an average incident power of approximately 17 mW (Song *et al.*, 2000). Several well-defined cavity modes are seen, with a typical modal linewidth of approximately 0.6 nm, limited by scattering from residual morphological roughness. Figure 11.2(b) shows the onset of stimulated emission (at threshold input power of $P_{th} = 32$ mW). A well-defined far-field pattern was acquired, shown in Figure 11.3, with a full-width-at-half-maximum radiation angle of approximately 5° , for the *linearly polarized* nearly Gaussian beam emerging from a 20- μm diameter aperture.

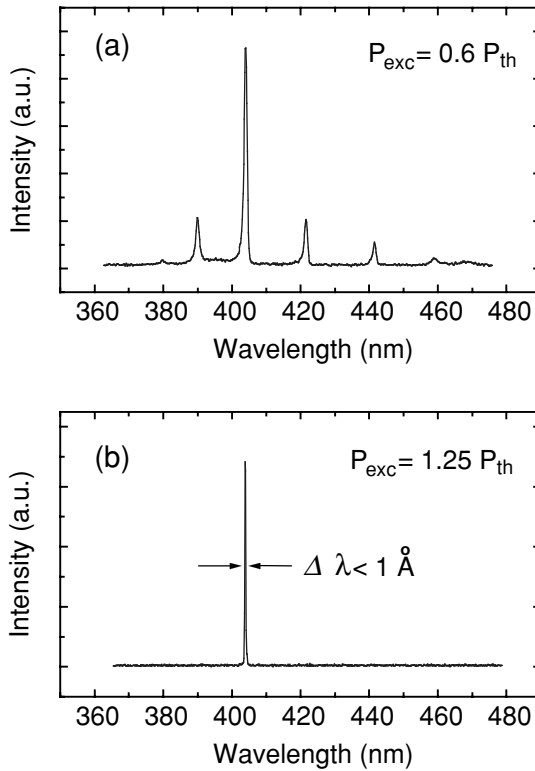


Figure 11.2: (a) Spontaneous emission spectra of the optically pumped InGaIn MQW VCSEL below threshold. (b) Stimulated emission spectra above threshold under quasi-cw pumping conditions at $T = 258$ K. (From Song *et al.* (2000a).)

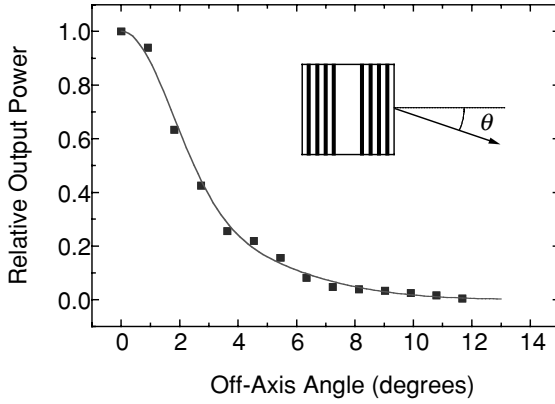


Figure 11.3: Beam profile measurement for the linearly polarized VCSEL output at $\lambda = 403$ nm. The radiation angle is measured relative to the beam center axis.

From the conditions of excitation we estimate that the threshold conditions of this type of optically pumped VCSEL correspond to approximately those in the cw edge-emitting diode lasers in terms of equivalent electrical injection (assuming an approximately 1 ns carrier lifetime). The threshold gain of the VCSEL is estimated to be several thousand cm^{-1} , which is also comparable to that of InGaN quantum well edge-emitting lasers (Nakamura, 1999).

11.2.2 Stress engineering of AlGaIn/GaN DBRs

The low contrast in index of refraction between AlN and GaN necessitates the use of a large number of layer pairs to achieve high reflectivities. The associated spectral bandwidths are typically relatively small. Someya and Arakawa (1998) reported the crack-free growth of a 35-pair $\text{Al}_{0.34}\text{Ga}_{0.66}\text{N}/\text{GaN}$ DBR with reflectivity up to 96% at 390 nm. Langer *et al.* (1999) reported a maximum reflectivity of 93% at 473 nm with 30 pairs of $\text{Al}_{0.41}\text{Ga}_{0.59}\text{N}/\text{GaN}$ DBRs. Ng *et al.* (2000) explored DBR mirrors consisting of binary AlN and GaN for increased contrast in the index of refraction. A network of cracks was observed which was attributed to the large tensile stress between the two binary compounds. It has been discovered (Han *et al.*, 2001) that the use of AlGaIn interlayers is effective in controlling mismatch-induced stress and suppressing the formation of cracks which otherwise occurred during growth of AlGaIn directly upon GaN epilayers. This idea has been applied in conjunction with *in-situ* monitoring to control stress evolution during growth of AlGaIn/GaN DBR mirrors by metal-organic vapor phase epitaxy. The employment of an AlN interlayer at the beginning of a thick (~ 5 μm) DBR growth leads to a substantial modification of the initial stress evolution. Tensile growth stress can be brought

under control and nearly eliminated through multiple insertions of AlN interlayers. Using this technique, crack-free growth of 60 pairs of $\text{Al}_{0.20}\text{Ga}_{0.80}\text{N}/\text{GaN}$ DBR mirrors has been achieved over the entire 2-in wafer with a maximum reflectivity of at least 99%.

A very useful real-time *in-situ* stress monitoring is based on wafer curvature measurements with a multi-beam optical stress sensor (MOSS) (Taylor *et al.*, 1998). Slopes of the stress–thickness product traces versus time during deposition cycles can be converted to instantaneous stress. Figure 11.4 shows the *in-situ* stress–thickness curves recorded by MOSS of two DBR structures consisting of 30 pairs of $\text{Al}_{0.20}\text{Ga}_{0.80}\text{N}/\text{GaN}$ layers, approximately 415 and 385 Å thick, respectively (Waldrip *et al.*, 2001). The upper curve (a) is from a DBR structure grown directly atop the 1- μm thick GaN layer with no interlayers. The quarter-wavelength AlGaIn/GaN DBR as a whole acts as a pseudoalloy in terms of inducing an accumulation of tensile stress energy. By contrast, curve (b) shows the *in-situ* MOSS for growth where an AlN interlayer (nominally 150 Å thick) was inserted between the HT GaN layer and the DBR structure. The use of an AlN interlayer reduces the in-plane lattice constant and consequently exerts a compressive stress during the initial growth of AlGaIn/GaN DBR structures. The observed compressive stress gradually decreases and passes through a stress-free region at around 0.5 μm ; a

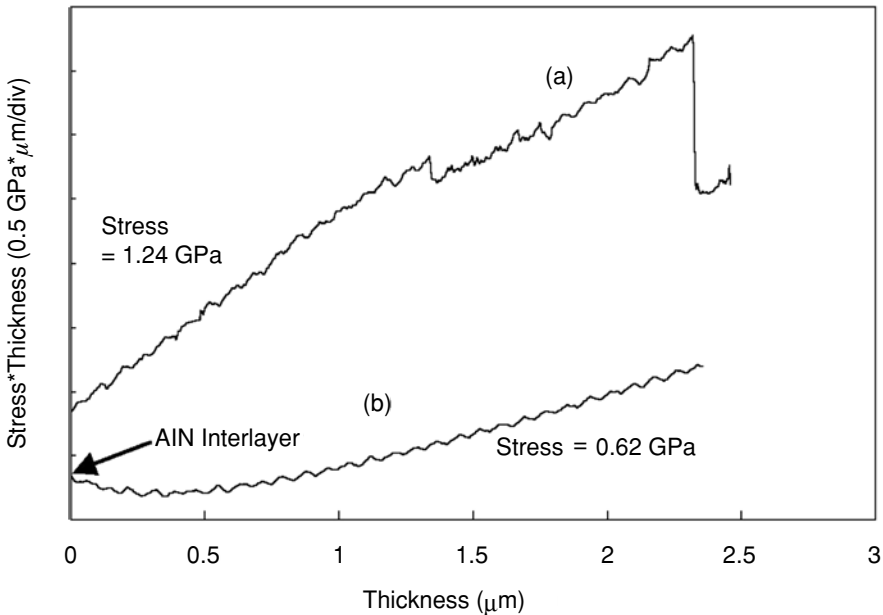


Figure 11.4: Stress–thickness versus thickness plots recorded by the *in-situ* stress sensor (MOSS) for two 30-pair AlGaIn/GaN DBRs: (a) grown directly on a GaN template, and (b) grown on an AlN interlayer. (From Waldrip *et al.* (2001).)

constant tensile stress (~ 0.62 GPa) is developed and sustained throughout the rest of the DBR growth. In this case, the surface morphology was found to be crack-free over the entire 2-in wafer.

Such DBRs, with high-quality morphology and a peak reflectivity $R \sim 0.991$, have been applied to demonstrate quasi-cw operation, at room temperature, of an optically-pumped $\text{In}_x\text{Ga}_{1-x}\text{N}$ ($x \sim 0.03$) MQW VCSEL at near $\lambda = 383$ nm (Zhou *et al.*, (2000)). The vertical cavity scheme combined a high reflectivity *in-situ* grown multi-layer $\text{GaN}/\text{Al}_{0.25}\text{Ga}_{0.75}\text{N}$ and postgrowth dielectric $\text{SiO}_2/\text{HfO}_2$ DBR. A photograph of the side profile of the low-divergence beam of circular cross-section is shown in the inset of Figure 11.5, which also displays the input/output power characteristics of a device with lasing threshold at pump power of 30 mW (average VCSEL output powers up to 3 mW were measured). However, finite thickness variation across the wafer led to spectral shifts of the cavity modes (relative to InGaN MQW gain spectrum) so that significant increases in threshold were encountered for devices fabricated from near the edge of the wafer. When accounting for the optical excitation volume, the fractional absorption of the pump, and using an electron-hole recombination time of approximately 0.5 ns, we estimate that the device is about

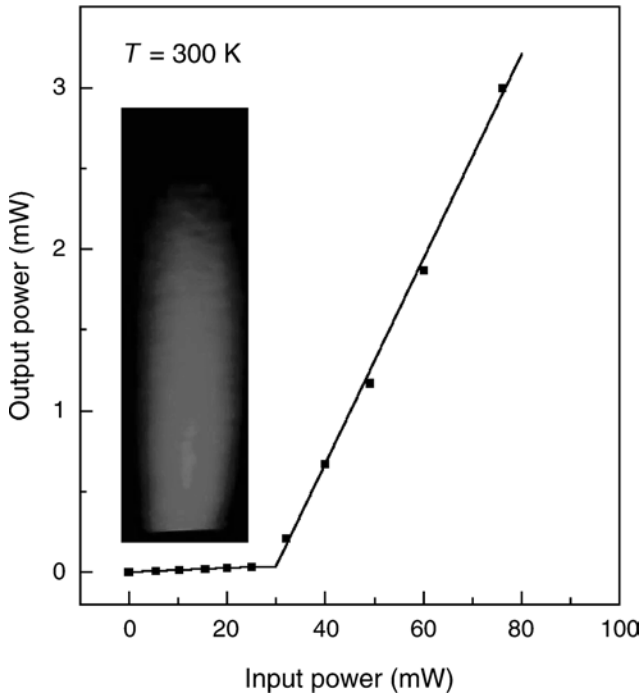


Figure 11.5: Average input versus output power of a violet optically pumped VCSEL device. The inset shows the beam far-field (side) profile captured on a screen. (From Zhou *et al.* (2000).)

25% efficient and that the threshold corresponds roughly to a carrier density of approximately 10^{19} cm^{-3} .

11.3 ELECTRICAL INJECTION: DEMONSTRATION RESONANT-CAVITY LEDs

A major challenge for a nitride vertical-cavity diode emitter involves the need for lateral injection of the holes into the *p*-side of the junction. The low conductivity of typical MOCVD-grown *p*-GaN (on the order of $1 (\Omega \text{ cm})^{-1}$) dictates that a high-conductivity *intracavity* layer be inserted between the top *p*-GaN layer and the adjacent DBR. This layer must not contribute to optical losses so that both its intrinsic absorption as well as its morphology (in terms of optical scattering) must be considered carefully. In principle, two approaches can be taken in this direction. Firstly, as shown below, appropriately processed indium-tin oxide (ITO) is a useful thin-film material both from the standpoint of low optical losses and in terms of forming an electrical contact to *p*-GaN. Secondly, we show that it is now possible to use a nitride-based (Esaki) tunnel junction which represents a class of versatile layered structures that have been applied to conventional III–V heterostructures to increase their design flexibility.

As was the case with the high-Q microcavity resonators described above, initial efforts at creating electrically-injected devices have employed either all-dielectric DBRs or a hybrid configuration with one AlGaIn *in-situ* grown DBR. As an example of the first approach, Figure 11.6(a) shows the schematic of a recent resonant-cavity LED (Song *et al.*, 2000a) where the design included the deposition of a 1000 Å thick intracavity hole current spreading layer of ITO onto the *p*-GaN cap layer of the as-grown nitride heterostructure. ITO has been used for transparent contacts on *p*-GaN. The resistivity of the magnetron-sputtered and thermally-annealed ITO was approximately $4 \times 10^{-4} \Omega \text{ cm}$ and its single-pass optical absorption was as low as 1% in the 400–500 nm wavelength range. The microcavity devices could be operated under continuous current injection condition to about 1 kA cm^{-2} without degradation, indicating their relative robustness (Song *et al.*, 2000a). The current–voltage characteristics of the 30–40 μm diameter RCLED devices, when compared with those of standard LED devices equipped with a conventional *p*-contact (Ni/Au), show that about 1.5 V of extra bias is required to reach the high injection level of approximately 1 kA cm^{-2} . This overhead is due to the ITO and we note that it is unclear from fundamental considerations what kind of a heterointerface resistance (junction) can be expected for the ITO/*p*-GaN system. Figure 11.6(b) shows an optical microscope image of one device emitting at a current density of 0.2 kA cm^{-2} , demonstrating a very uniform luminescence over the entire active area defined by a 35-μm ITO *p*-contact. Under pulsed injection, we have been

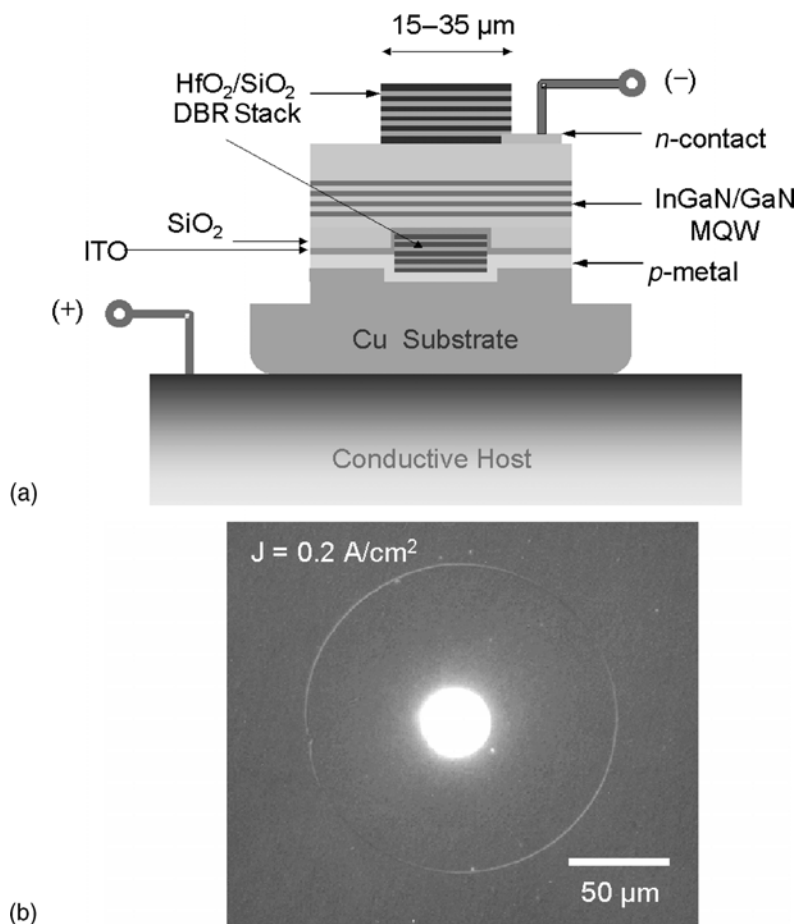


Figure 11.6: (a) Schematic drawing of an InGaN MQW RCLED incorporating two dielectric DBRs and an intracavity ITO hole current spreading layer. (b) Photograph of the light emission pattern from 30- μm aperture.

able to reach current densities up to about 15 kA cm^{-2} in these types of device designs, with $0.5 \mu\text{s}$ pulses at a duty cycle of $\sim 1\%$. The emission spectrum of such a RCLED is shown in Figure 11.7, with the emission collected within an approximately 5° forward solid angle. The cavity for the case shown is relatively thick, approximately 16λ but considerably thinner structures have also been fabricated (a few λ) by the chlorine ECR etching technique without loss of device quality. The spectrum, centered at around $\lambda = 430 \text{ nm}$, shows the evidence for a high-quality resonator through the definition of the cavity modes. The modal linewidth is approximately 0.6 nm , implying a cavity Q-factor of approximately 750.

As a second example of current progress, we show a vertical-cavity LED which combines the hybrid resonators structure with a hole injection scheme that is based

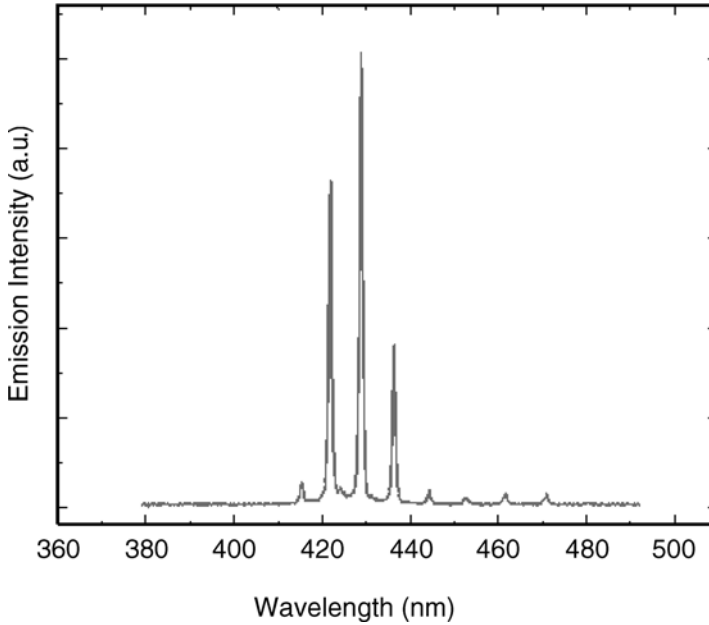


Figure 11.7: Emission spectrum of the RCLED device with two dielectric mirrors.

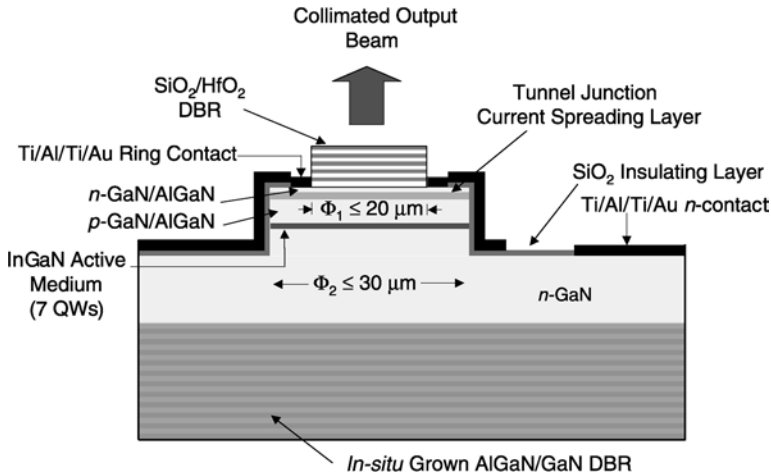


Figure 11.8: Schematic of an RCLED device with one as-grown GaN/AlGaIn and one dielectric DBR mirror, incorporating a nitride tunnel junction as the top current spreading layer.

on the use of a p^{++}/n^{++} InGaIn/GaN tunnel junction. (ITO has been also usefully employed for the hybrid resonator case.) A schematic diagram of the overall vertical-cavity emitter is shown in Figure 11.8 (Diagne *et al.*, 2001). Similar to the optically-pumped VCSELs, AlN strain-relief layers were used in the deposition of a

60-layer-pair quarter-wave GaN/Al_{0.25}Ga_{0.75}N bottom DBR stack. Those growths that yielded a root-mean-square average roughness no worse than 4 nm over an area of $1 \times 1 \text{ mm}^2$ were deemed suitable for continuation of the epitaxy. The active p - n junction region was grown directly atop the GaN/(Al,GaN) DBR, composed typically of 7 In_{0.08}Ga_{0.92}N QWs ($L_w = 40 \text{ \AA}$) with GaN barriers ($L_B = 60 \text{ \AA}$), and surrounded by approximately 1000- $\text{\AA}$ thick Al_{0.07}Ga_{0.93}N current blocking/carrier confinement layers. The tunnel junction in the GaN system (Takeuchi *et al.*, 2001, Jeon *et al.*, 2001) was included in the superstructure of the nitride segment and capped by a n -GaN layer, as shown in Figure 11.8. Note that in this case the positive bias to the device was applied through a contact to this n -layer. Lateral current spreading on the scale of $\sim 100 \text{ }\mu\text{m}$ was obtained, given the nearly 100 times higher n -type conductivity of GaN. The tunnel junction itself was grown atop the 1000- $\text{\AA}$ thick p -GaN layer as a p^{++}/n^{++} InGaN/GaN bilayer with the thicknesses of the layers approximately 150 $\text{\AA}$ and 300 $\text{\AA}$, respectively. The doping levels were approximately $1 \times 10^{20} \text{ cm}^{-3}$ Mg and $6 \times 10^{19} \text{ cm}^{-3}$ Si for the junction. The vertical cavity was completed by capping the structure with a multi-layer $\lambda/4$ stack of SiO₂/HfO₂ ($R > 0.995$), deposited by reactive ion beam sputtering. The top dielectric DBR was patterned so that the device had an effective optical aperture varying from 10 to 30 μm .

The current density versus voltage of a typical device is shown in Figure 11.9 up to a high continuous injection level ($\sim 1 \text{ kA cm}^{-2}$), which shows evidence of series resistance, assigned in part to the presence of the tunnel junction. Nonetheless,

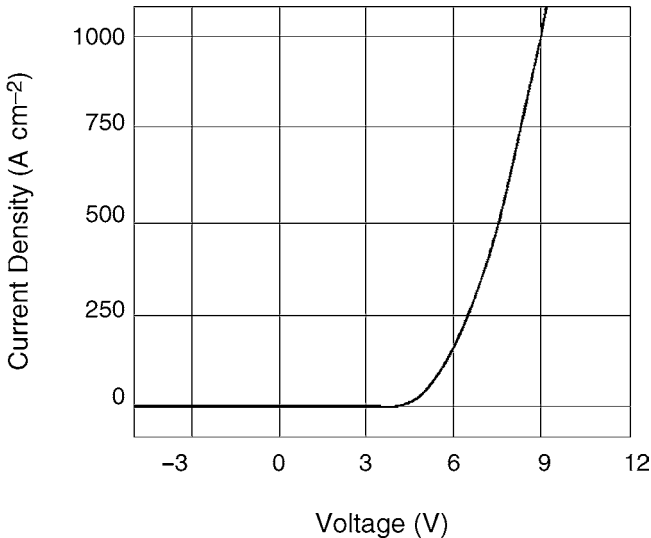


Figure 11.9: Current density versus voltage of a hybrid RCLED device with a 20 μm diameter mesa defining the vertical current path.

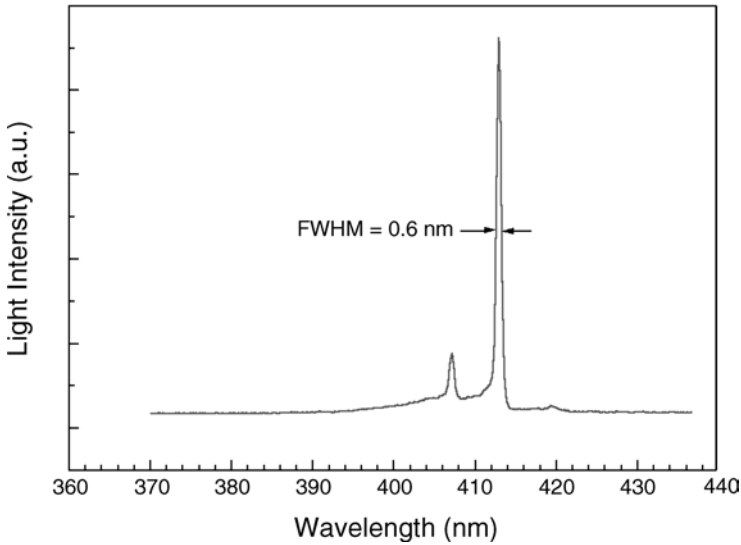


Figure 11.10: Emission spectrum of the RCLED hybrid device. (From Diagne *et al.* (2001).)

lateral current spreading was clearly accomplished so that the far-field light emission from the devices was uniform in its average intensity across the emitting aperture. However, when examining the emission under high spatial resolution ($\sim 1 \mu\text{m}$) evidence was found of some tendency towards “filamental” vertical conduction, reflective perhaps of the influence of local compositional or doping inhomogeneities within the tunnel junction. Figure 11.10 shows the output spectrum of a typical device at an operating current density of approximately 0.2 kA cm^{-2} . The emission was observed in the direction normal to the planar device, within an angular view of approximately 10° . While the optical resonator is rather thick ($> 10\lambda$), only two vertical cavity modes are seen, demonstrating the restrictive spectral bandwidth of the AlGaIn DBR. The dominant mode at $\lambda = 413 \text{ nm}$, which coincides with the high-reflectivity region of the DBR and the peak of the QW photoluminescence emission, has a spectral linewidth of approximately 0.6 nm . This value is comparable with the linewidths previously measured in the best structures fabricated in the author’s laboratory that were designed for optically pumped VCSEL operation.

The examples shown above suggest that the implementation of vertical-cavity LEDs has provided the important building blocks for demonstrating a diode VCSEL in the violet. We note that the incorporation of an InGaIn/GaN p^{++}/n^{++} tunnel junction has also been exploited in the demonstration of a monolithically integrated two-color blue/green LED (Ozden *et al.*, 2001a). The VCSEL challenge is two-fold: high-quality epitaxy and relatively complex device processing. For example, layer

thickness and composition (Al, In) control over a 2–3-in wafer area is still quite difficult, yet required so that spectral overlap, for example, between the high-reflectivity band of the AlGaIn DBR and the maximum of the InGaIn MQW gain remain in spectral synchrony.

Finally we mention that the application of the lateral epitaxial overgrowth techniques discussed in Chapters 9 and 10 may become quite useful also for future vertical cavity emitters. The inclusion of ELOG process/growth steps should aid in creating flexibility for designing and implementing blue/violet RCLEDs and VCSELs in at least two different ways. Firstly, the patterned growth can be adapted for creating a buried bottom dielectric DBR mirror. An illustration is shown in Figure 11.11, where a patterned $\text{HfO}_2/\text{SiO}_2$ multi-layer dielectric stacks was deposited on a GaN buffer layer prior to subsequent regrowth of GaN. The particular dielectric stack was terminated with an SiO_2 layer so that the ELOG process would occur normally. The second application of the lateral epitaxy pertains to building in a current aperturing scheme in order to alleviate the problem that arises from the competition between vertical and lateral transport in the nitride devices (especially on the *p*-side). Figure 11.12 shows the schematic of a possible arrangement where an SiO_2 -defined current aperture is implemented in conjunction with lateral epitaxial growth.

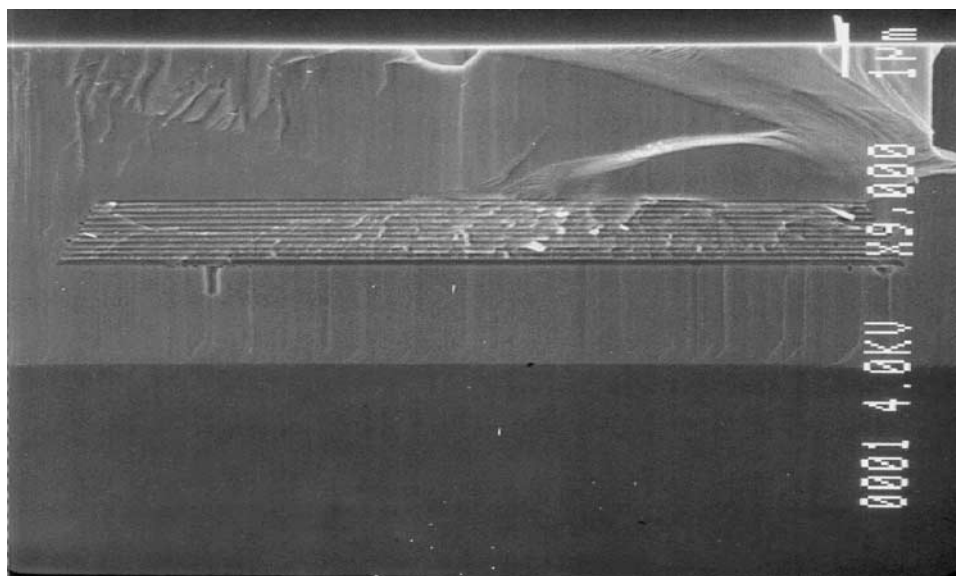


Figure 11.11: Illustration of the use of lateral epitaxial GaN overgrowth to “bury” a dielectric DBR. The imperfections are due to breaking of the sample so as to gain an edge view in this cross-sectional SEM image.

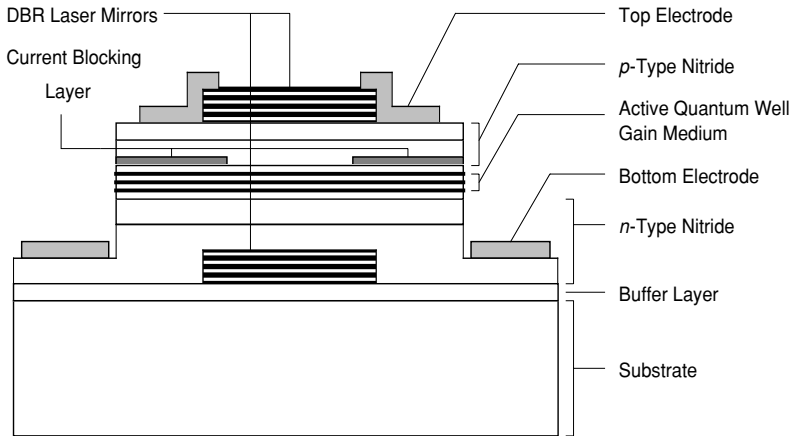


Figure 11.12: Schematic drawing illustrating a possible blue VCSEL device structure which features a buried dielectric DBR and current confining aperture.

11.4 SUMMARY

As illustrated through specific examples in this chapter, good progress is in evidence in the development of the key building blocks that are necessary for the realization of blue and near-ultraviolet vertical-cavity diode lasers and LEDs. We have reviewed the strategies for crafting high-Q-factor resonators, with examples of optically-pumped VCSEL operation, and shown initial examples of vertical-cavity LED emitters. While advances in the laboratory are encouraging, one should not underestimate the difficulties that are still faced in efforts to create practical short wavelength VCSELs. Yet the payoff for introducing such a new class of optoelectronic devices can be significant, with a wide range of applications. If we envision the eventual availability of short-wavelength VCSEL arrays (as matrix addressable LED arrays have now been introduced (Ozden *et al.*, 2001)), the application base expands even further, especially in the areas of future ultracompact chipscale chemical and biological sensing and diagnostic systems.

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12

Concluding remarks

We began in Chapter 1 by discussing applications that required compact blue-green lasers; hence, it seems only fitting to end by examining the extent to which these requirements have been fulfilled. In this final chapter, then, we attempt to gather up some of the diverse topics that this book has treated and establish the current state of the art in the *application* of compact blue-green lasers.

The preceding chapters have covered three principal approaches to creating compact blue-green lasers. In the first approach, blue-green light is generated through nonlinear frequency conversion of infrared semiconductor diode lasers or diode-pumped solid-state lasers. We saw that since these nonlinear processes tend to be rather weak, the desire for efficient generation of blue-green light has stimulated the development of high-power *infrared* lasers as well as the invention of a host of device configurations intended to boost the nonlinear conversion efficiency. These configurations include resonator-enhancement schemes, intracavity SHG, and waveguide implementations.

An alternative approach – the “upconversion laser” – directly excites a blue-green laser transition by combining the energy of two or more lower-energy pump photons through excited state absorption or cooperative energy transfer processes. Upconversion lasers using both bulk and fiber-optic media have been demonstrated.

Finally, we examined semiconductor diode lasers that are pumped by electrical injection and directly produce blue-green photons. Two main materials systems have been used to fabricate these devices: GaN and ZnSe. We saw that while the GaN system has so far produced more impressive laser devices, work done in both systems has been crucial for an understanding of devices based on these widegap semiconductors.

We might now ask “Where have the efforts described in this book led? What is the current status and outlook for compact blue-green lasers? Have they been used yet for the applications described in Chapter 1?” We close this treatise by briefly commenting upon these questions.

First, it is probably worth commenting that although some fifteen years have passed since breakthroughs in infrared diode laser technology prompted the first flurry of excitement over new prospects for compact blue-green sources, the need for such sources in a wide variety of applications has not ebbed. In fact, it might well be argued that the explosive growth of the internet and the growing convergence of the information and entertainment industries has enhanced interest in certain applications of compact blue-green lasers – for example, higher-density DVDs and large-format, high-resolution color displays. Furthermore, the advancement of biomedical technologies continues to sustain interest in compact, efficient, short-wavelength sources for these applications.

For some applications, the remarkable development of GaN semiconductor lasers has been a major boon. The impressive breakthroughs and tremendous progress made in these devices have finally led, at the time of this writing, to the availability of development devices operating in the violet (~ 405 nm) at powers up to ~ 30 mW in a single transverse mode (according to the website for Nichia Corp., www.nichia.com). These characteristics are sufficient to allow these devices to be used in applications where the relatively low power level and narrow range of available wavelengths do not constitute critical limitations.

For example, GaN laser diodes have now been used for demonstrations of high-density optical data storage, in which the expected advantages described in Chapter 1 have been realized. To cite one example, Akiyama *et al.* (2001) have reported demonstrating a rewritable optical disk with a capacity of 27 GB using a GaN laser. Numerous other descriptions of work to develop high-density optical data storage using GaN lasers could also be cited (for example, Ko *et al.* (2001), Ichimura *et al.* (2000)).

GaN laser diodes are also well suited to certain biomedical applications, where high power is not essential, and the violet wavelength is very effective for exciting fluorescence, spectral analysis of which reveals information about the sample under observation. For example, Gustafsson *et al.* (2000) have used an InGaN laser diode in a compact fluorescence sensor that can be used to monitor the condition of vegetation and to distinguish between healthy and pre-malignant skin tissue; Girkin *et al.* (2000) have reported on the use of a 406-nm InGaN laser diode for confocal microscopy of biological media stained with fluorescent dyes.

When GaN laser diodes are used with an extended cavity, it becomes possible to tune them sufficiently to make them useful for other spectroscopic applications (Leinen *et al.*, 2000). Gustafsson *et al.* (2000a) have reported using extended-cavity violet laser diodes for spectroscopy of potassium atoms. The same group has also mixed the ~ 400 nm output of a GaN laser diode with the 688-nm emission from a red diode laser to create a source tunable around 254 nm that can be used for mercury detection (Alnis *et al.*, 2000).

While GaN laser diodes are nearly ideal sources for applications like these, the relatively low power and limited wavelength range limits their use in other important applications. Furthermore, the prospects for obtaining significantly higher powers and a broader range of wavelengths from short-wavelength semiconductor diode lasers in the future are unclear at this time. Hence, for applications requiring substantially higher powers or wavelengths longer than ~ 410 nm, sources based on SHG are more likely to play a key role, at least in the near future. Commercially-available, high-power (~ 10 W) green lasers based on intracavity SHG are becoming increasingly popular as replacements for argon lasers. High-power diode-pumped blue and green lasers based on SHG and SFG continue to be extensively developed for other applications where higher powers or longer blue-green wavelengths are required, such as laser projection displays (Hollemann *et al.* 2000).

Upconversion lasers continue to be pursued in the laboratory, but are still more accurately described as “promising” rather than “practical”. Fiber upconversion lasers, in particular, remain intriguing because of the attractive features of the fiber geometry, but so far have not progressed to a stage suitable for use in applications. However, the pursuit of these lasers has also reinvigorated interest in the spectroscopy and laser characteristics of rare-earth-doped solid-state lasers and has broadened and deepened our understanding of these systems in ways that have had implications for more “conventional” lasers based on these systems.

Hence, some of the potential benefits of compact blue-green lasers that were anticipated fifteen years ago have begun to be realized, as some of the very diverse technical directions that have been pursued have begun to yield practical devices. It is probably true of nearly any technology that as it matures, there is a progressive convergence between what is sought in principle and what can be achieved practically. Applications for which there emerges no practical solution may become dormant, while the availability of a new solution may spark the invention of new applications. In the case of compact blue-green lasers, fifteen years of intense effort directed at developing sources for specific applications have clarified which approaches are more practical than others, what the limitations and advantages of each implementation are, which approaches “match up” best with preconceived applications, and the ease or difficulty of developing a novel laboratory technology into a product and bringing it to market. The current result of this process is that specific types of compact blue-green lasers are beginning to be used in specific applications to which they are especially well suited, as described above. On the other hand, the availability of versatile devices like the GaN laser diode are stimulating the conception of new applications in which such components can be used to great advantage. For the remainder, there is still more work to be done in finding lasers suitable for existing applications and new applications for existing blue-green lasers.

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