

Fundamentals of Attosecond Optics

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To Hou Xun, my advisor and friend.

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Preface

The generation of attosecond optical pulses was first demonstrated in 2001. Along with a related process, high-order harmonic generation, which was discovered around 1988, this laser-like light is redefining ultrafast physics and chemistry. This book has been written to fill the need for a focused introduction to the emerging field of attosecond optics and its applications. It targets senior undergraduate students, graduate students, and scientists who want to enter the field. It should also be useful for senior scientists and engineers in industry who are developing the next generation ultrafast lasers. While reporting the latest research advances (including examples from our own group's research, with which we are intimately familiar), we focus on fundamental concepts and techniques. At the end of each chapter, we provide problems for teaching purposes and reference important papers for readers who are interested in the original work.

Many universities offer courses on the interaction of matter with ultrafast, high-power lasers. This book can serve as a stand-alone textbook for courses on attosecond optics. In addition, Chapters 2, 3, 6, 7, and 8 can be used as supplementary material for more general laser courses, and Chapters 4, 5, and 9 have information relevant to atomic physics courses.

The organization of the book builds from basic underlying theory to more complex ideas related to attosecond optics. The generation of attosecond optical pulses requires knowledge of femtosecond laser technologies. At the same time, the mechanisms of attosecond pulse generation are quite different from those of femtosecond lasers. We explain these mechanisms using both semiclassical models and quantum mechanics theories. In addition, high-order harmonic generation as compared to the field of attosecond physics is much more mature. We introduce the technique for generating attosecond train first because of its connection with high harmonic generation. We then explain gating methods for extracting single isolated pulses. Finally, we provide illustrative examples of attosecond applications.

In brief, Chapter 1 describes the motivations of attosecond research, including a brief review of the history and explanation of the connection between attosecond pulses and high harmonic generation. Chapters 2 and 3 focus on driving lasers as key tools used in attosecond generation. As most high-power lasers are based on chirped pulse amplification, we discuss it first, followed by details on how to generate few-cycle pulses. Chapter 3 looks at carrier-envelope phase stabilization. Chapters 4 and 5 set the theoretical foundations for single atom response. We first introduce the intuitive semiclassical model and then discuss quantum theory that describes the dipole phase. Chapter 6 discusses propagation effects, introducing several approaches for improving phase matching. Chapters 7 and 8 turn to attosecond pulse generation and characterization, covering two types of light sources: attosecond pulse train and single isolated pulses. Chapter 9 gives several examples of experimental applications of attosecond pulses.

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Finally, I would like to thank my wife, Shanshan Liao, for her love and affection, and for supporting my research over the last 25 years. I would also like to thank my sons, Wenbo and Libo, for their understanding.

Zenghu Chang

Author



pulse amplifier and inventing attosecond pulses.

Zenghu Chang joined the University of Central Florida as distinguished professor of physics and optics in 2010. Prior to this, he was Ernest K. and Lillian E. Chapin Professor of Physics at Kansas State University. He now serves as chair of the Optical Attoscience Technical Group of the Optical Society of America; he is also a fellow of the American Physical Society. After receiving his doctorate at the Xi'an Institute of Optics and Precision Mechanics, Chinese Academy of Sciences, in 1988, Dr. Chang served as an associate professor before spending time at the Rutherford Appleton Laboratory in the United Kingdom. From 1996 to 2000, he worked in the Center for Ultrafast Science at the University of Michigan. In 2001, Dr. Chang's group demonstrated the high harmonic cutoff extension using a long wavelength driving laser. Other notable contributions include demonstrating the first active carrier-envelope phase stabilization of grating-based chirped the double optical gating for the generation of single isolated

Quest for Attosecond Optical Pulses

The longest timescale that we can possibly observe is the age of the universe, which is approximately 14 billion years, or 4×10^{17} s. With the emergence of attosecond optics in the twenty-first century (1 as = 10^{-18} s), it is now possible to “see” things that happen in extremely short timescales. To imagine how short, just think that comparing 1 as to 1 s is equivalent to comparing 1 s to the age of our universe. Attosecond optics is a subfield of ultrafast optics and strong field physics.

1.1 Ultrafast Optics

Ultrafast optics is the study of the generation, characterization, and application of ultrashort light pulses. The definition of “ultrashort” has evolved over time. The motivation for generating shorter light pulses comes from the demands of many scientific and industrial areas. These demands fall into two main categories: high optical power delivery and high-speed imaging. Before 2001, nanosecond (1 ns = 10^{-9} s) to femtosecond (1 fs = 10^{-15} s) lasers were developed for ultrafast optics studies and application. In this chapter, we briefly review basic concepts used in ultrafast science, and then extend them to attosecond optics.

1.1.1 High-Power Applications

For mathematical convenience, it is a common practice to assume that the temporal envelope of a coherent light pulse and the transverse spatial profile of a laser beam are Gaussian functions.

1.1.1.1 Power, Peak Power, and Pulse Duration

For Gaussian pulses, the instantaneous power at time t can be expressed as

$$\mathcal{P}(t) = P_0 e^{-4 \ln 2 \left(\frac{t}{\tau}\right)^2}. \quad (1.1)$$

Here, the pulse duration τ is the full width at half maximum (FWHM) of the pulse envelope, and $e = 2.7182$ is the Euler constant. A 5 as Gaussian pulse is shown in [Figure 1.1](#).

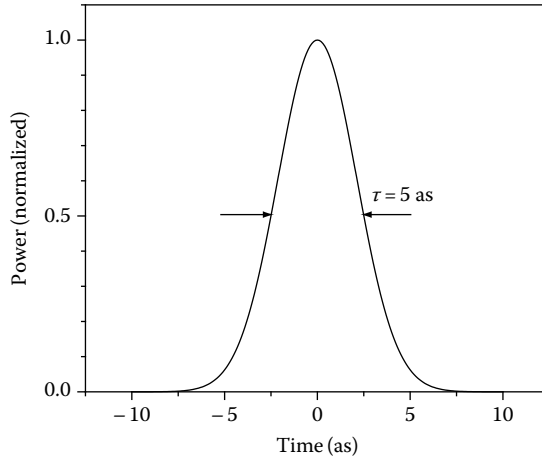


Figure 1.1 A Gaussian pulse with 5 as FWHM.

1.1.1.2 Pulse Energy

The peak power P_0 is related to the pulse energy ε by the expression

$$P_0 = \frac{\varepsilon}{\int_{-\infty}^{+\infty} e^{-4 \ln 2 \left(\frac{t}{\tau}\right)^2} dt} = \sqrt{\frac{4 \ln 2}{\pi}} \frac{\varepsilon}{\tau} \cong 0.94 \frac{\varepsilon}{\tau}. \quad (1.2)$$

The factor 0.94 originates from the Gaussian pulse shape. For a given optical pulse energy, the peak power is inversely proportional to the pulse width. Thus, high optical power can be reached by reducing the pulse duration, which is one of the motivations for generating ultrashort light pulses. It is often cheaper to reduce the pulse width than to increase the pulse energy.

Experimentally, pulse energy can be measured easily by converting light into heat or electric current. The temporal profile of pulses longer than 100 picoseconds ($1 \text{ ps} = 10^{-12} \text{ s}$) can be measured conveniently with fast photodiodes and oscilloscopes. Significant effort has been devoted to the development of methods for measuring the duration of femtosecond pulses, some of which are described in Chapter 2. The characterization of attosecond pulses is discussed in Chapters 7 and 8.

1.1.1.3 Fluence

Fluence is defined as the energy deposit on a unit area by a laser pulse. Its unit is Joule per centimeter square.

1.1.1.4 High-Power Lasers

Since the laser was invented in 1960, it has become the dominant high-power light source. Because of their temporal coherence, lasers can generate reproducible optical pulses much shorter than those generated by other light sources. In other words, high power can be generated with low pulse energy. High-power lasers can be classified into three categories: single shot (approximately one shot per hour), low repetition



Figure 1.2 A femtosecond high-power laser system. The whole laser fits on one 8' $\times$ 12' optical table. The cylindrical container above the laser beam is for cooling the laser gain medium with liquid nitrogen.

(~ 10 Hz), and high repetition (1 kHz or higher). The pulse duration of all these lasers can reach < 100 fs, but their energy decreases with repetition rate. The pulse energy of low repetition rate femtosecond lasers can be on the order of 1 J, whereas the pulse energy is on the order of mJ for most kHz lasers. For example, the Kansas Light Source laser in the author's lab generates 3 mJ, 30 fs pulses at 1 kHz, yielding a peak power of 0.1 terawatts ($1 \text{ TW} = 10^{12} \text{ W}$). A picture of the laser is shown in Figure 1.2. Laser powers as high as petawatt ($1 \text{ PW} = 10^{15} \text{ W}$) have been generated using femtosecond lasers. High-power femtosecond lasers will be discussed in detail in Chapters 2 and 3.

1.1.1.5 Average Power and Repetition Rate

It is important not to confuse the average power of a pulse train with the peak power of each individual pulse. For a pulse train with a repetition rate f_{rep} , the average power is given by

$$P_{\text{ave}} = f_{\text{rep}} \mathcal{E}. \quad (1.3)$$

Powermeters used in laser labs measure the average power, not the peak power. The average power of the Kansas Light Source is 3 W, which is lower than that of a light bulb. However, its peak power is 100 GW, which is much higher than the capacity of the Hoover Dam, 2 GW.

The repetition rate of a laser can be easily determined by using a fast photodiode and an oscilloscope, or a photodiode and pulse counter. Thus the pulse energy can be deduced from the average power measurement. The peak power can be determined by plugging in the measured pulse energy and duration values into Equation 1.2.

1.1.1.6 Intensity and Field Amplitude of CW Light

According to Maxwell's theory, light can be considered an electromagnetic wave. At a given spatial point, the electric field of a lineally

polarized monochromatic laser (also known as continuous wave laser or CW) with angular frequency ω can be expressed as

$$\mathcal{E}(t) = E_0 \cos(\omega t), \quad (1.4)$$

where E_0 is the field amplitude. Similarly, the magnetic field is $\mathcal{B}(t) = B_0 \cos(\omega t)$. The amplitude of the magnetic field is $B_0 = E_0/c$, where c is the speed of light in vacuum.

Light-matter interactions start with the response of the electrons to the light fields. The magnitude of forces acting on the electron by the electric and magnetic fields are eE_0 and $eB_0v(=eE_0[v/c])$, respectively, where $e = 1.6 \times 10^{-19}$ C is the charge of the electron, and v is the speed of the electron. When the speed of the electron is much lower than the speed of light, the contribution from the electric field is much larger than that from the magnetic field. In this nonrelativistic regime, the response of the electron to the light is directly related to the strength of the electric field, whereas the effects of the magnetic field can be ignored. This is the regime considered in this book.

The electric field amplitude of light is difficult to measure directly. This is because the field changes very rapidly (the light frequency is on the order of 10^{14} Hz). In Chapter 8, we show how the electric field oscillation in time and space is directly measured using attosecond electron probes. It is known in optics that the field amplitude is related to the intensity, I (also known as irradiance), by

$$I = \frac{1}{2} \epsilon_0 c E_0^2. \quad (1.5)$$

Here, $\epsilon_0 = 8.8542 \times 10^{-12}$ C²/N-m² is the electric permittivity of free space. The intensity is defined as the cycle-averaged power transmitted through a unit area. Unlike field amplitude, intensity can be determined by measuring the power and the laser beam size. In the laser community, it is a common practice to use Watt per centimeter square as unit of intensity.

1.1.1.7 Peak Intensity and Beam Size

A cylindrically symmetric laser beam with a Gaussian transverse profile can be expressed as

$$I(r) = I_0 e^{-2\left(\frac{r}{w}\right)^2}, \quad (1.6)$$

where

$1/e^2$ radius w is a measure of the beam size
 I_0 is the peak intensity

The intensity distribution of a 5 μ m Gaussian beam is shown in [Figure 1.3](#). For a CW beam with power P , the peak intensity is

$$I_0 = \frac{P}{\int_0^\infty e^{-2\left(\frac{r}{w}\right)^2} r^2 dr} = \frac{2P}{\pi w^2}. \quad (1.7)$$

One should pay attention to the number 2 in front of the power, which originated from the definition of the beam size. The intensity distribution $I(r)/I_0$ can be measured by sending the laser beam to a CCD camera,

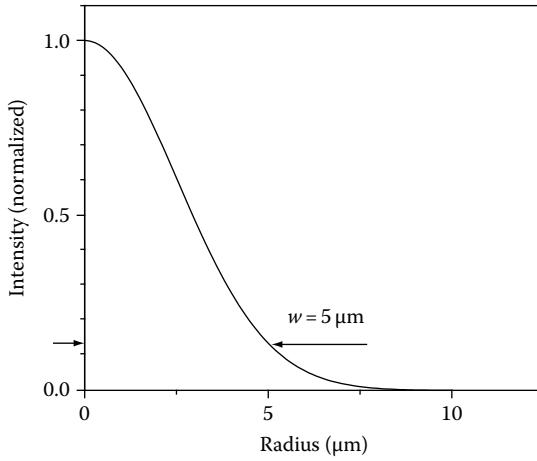


Figure 1.3 A Gaussian beam with 5 μm spot size.

which is very similar to a webcam. The value of w can then be obtained from a lineout of the CCD image.

1.1.1.8 Gaussian Beams and Gaussian Pulses

The intensity of a pulsed laser beam varies in both space and time. For a Gaussian pulse in a Gaussian beam, the intensity can be expressed as

$$I(r, t) = \frac{2P_0}{\pi w^2} e^{-2\left(\frac{r}{w}\right)^2} e^{-4 \ln 2 \left(\frac{t}{\tau}\right)^2} = \frac{1.88\epsilon}{\pi w^2 \tau} e^{-2\left(\frac{r}{w}\right)^2} e^{-4 \ln 2 \left(\frac{t}{\tau}\right)^2}. \quad (1.8)$$

The peak intensity at the center of the beam is

$$I_{0p} = \frac{1.88}{\pi} \frac{\epsilon}{w^2 \tau}. \quad (1.9)$$

Apparently, there are three approaches to obtain high intensity. The smallest spot size a laser beam can be focused to is the laser wavelength due to the diffraction from the limited aperture of the focusing optics, which is $\sim 1 \mu\text{m}$, as shown in Figure 1.4. The shortest laser pulse that can be generated is one optical cycle, which is 3 fs. For example, at the University of Michigan, a laser called λ^3 can focus 1 mJ, 8 fs pulses down to a $1.2 \mu\text{m}$ spot size for generating laser intensities above 10^{18} W/cm^2 , the so-called relativistic intensity. To reduce the pulse duration to less than 1 fs and to focus the light to less than $1 \mu\text{m}$, ultraviolet (UV) light or x-rays are needed.

1.1.1.9 Atomic Units

For bound electrons in atoms, the motion of an electron in a light field is determined by the relative strength of the external field to the atomic Coulomb field. In the Bohr model of the hydrogen atom, the electric field experienced by the electron in the ground state is

$$E_H = \frac{e^2}{4\pi\epsilon_0 a_0^2}, \quad (1.10)$$

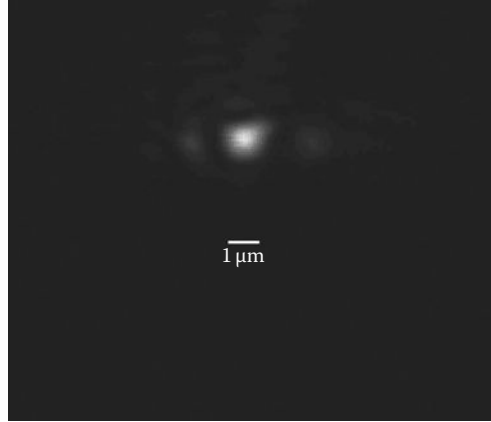


Figure 1.4 The CCD image of the focused λ^3 laser beam at the University of Michigan. The focal spot has a diameter $w = 1.2$. (Reprinted from O. Albert, H. Wang, D. Liu, Z. Chang, and G. Mourou, Generation of relativistic intensity pulses at a kilo hertz repetition rate, *Opt. Lett.*, 25, 1125–1127. With permission of Optical Society of America.)

where $a_0 = 0.0529 \text{ nm}$ is the Bohr radius, which is one atomic unit of length. The value of E_H then becomes $5.142 \times 10^9 \text{ V/cm}$, which is defined as one atomic unit of electric field. When the laser field amplitude E_0 is equal to E_H , the corresponding laser intensity is called one atomic unit of intensity, which is $3.55 \times 10^{16} \text{ W/cm}^2$. Atomic units are frequently used in attosecond physics and atomic physics literatures. Many equations can be simplified by taking advantage of $e = m_e = \hbar = a_0 = 1$ in atomic units, where $\hbar$ is Planck's constant h divided by 2π , $h = 6.626 \times 10^{-34} \text{ Js}$, and $m_e = 9.109 \times 10^{-31} \text{ kg}$ is the rest mass of the electron.

For visible and infrared lasers, the photon energy $\hbar\omega$ ($\sim 2 \text{ eV}$) is much smaller than the binding energy (or ionization potential) of many atoms. For the hydrogen atom, the ionization potential is

$$I_{p,H} = \frac{m_e e^4}{(4\pi\epsilon_0)^2 2\hbar^2} \frac{1}{n^2}, \quad (1.11)$$

where n is the principal quantum number. $I_{p,H} = 13.6 \text{ eV}$ when the electron is in the ground state ($n = 1$). When the laser intensity is much lower than one atomic unit, the motion of the electron in an atom can be described by a simple harmonic oscillator. The resonant frequency of the oscillator is related to the excitation energies. The amplitude of the displacement of the electron's motion is proportional to the amplitude of the light field. This is the linear optics regime.

1.1.1.10 Nonlinear Optics and Strong Field Physics

The intensity of light from a laser can be much higher, and as a result, the amplitude of the displacement of the bound electron no longer linearly follows the laser field. The first nonlinear optics phenomenon, second harmonic generation, was demonstrated in 1962. Nonlinear optics, which has become a subfield in optics, has been studied extensively with infrared (IR), visible, and UV lasers; however, not much has been done with extreme ultraviolet

(XUV) or x-rays because of the lack of high-power sources at shorter wavelengths. One frontier of attosecond optics research is the generation of high-power pulses for studying nonlinear processes in the soft x-ray range.

When the laser intensity approaches one atomic unit, an electron can be freed from the atom either by absorbing many photons or by tunneling through the Coulomb barrier formed by the superposition of the atomic potential and the laser field. The ionization of the electron by a laser field and the subsequent recombination are directly related to the generation of attosecond pulses using lasers, which is discussed in Chapters 4 and 5.

At even higher laser intensities ($>10^{18}$ W/cm²), the freed electron from atoms can be accelerated by the laser field at a speed close to the speed of light. The change in electron mass leads to nonlinear optics in the relativistic regime. Generating attosecond pulses at such high intensities has been proposed. In this book, we consider only the laser–matter interaction in the nonrelativistic regime.

1.1.2 High-Speed Imaging

The human eye is a marvelous optical instrument. At a simplistic level, its operation is similar to that of a digital video camera. Our brains function as imaging processors and storage devices. However, things we can see are limited by the spectral response, spatial resolution, and temporal resolution of our eyes. Visible light extends from a wavelength range of 400–720 nm. The response time of human eyes is approximately 50 ms and the spatial resolution is approximately 50 μ m. Just as microscopes were invented to enhance our capability to see small objects, so were high-speed cameras developed to observe fast events.

Microscopic and macroscopic objects move with various speeds. There are two characteristic speeds in nature: the speed of light (3×10^8 m/s) and the speed of sound (~ 300 m/s). Because of the difference in mass, it is possible to accelerate electron and ions to nearly the speed of light. As an example, the proton and antiprotons in the Large Hadron Collider are moving at 99.999999% of the speed of light. On the other hand, macroscopic objects move much slower. The speed of a bullet is approximately 1000 m/s, on the order of the speed of sound. When we take a sequence of pictures of a moving body with a video or movie camera, the exposure time, Δt , of each frame should be much shorter than the time that the object takes to move across its size, D ; otherwise the image will be blurred. As a rule of thumb, we can estimate the exposure time by the equation

$$\Delta t \approx 0.1 \frac{D}{v}, \quad (1.12)$$

where v is the speed of the object. The prefactor determines the blurriness caused by the motion. For a bullet, that exposure time should be approximately 1 μ s.

There are two approaches to control the exposure time. One is a mechanical shutter, as in most of the old-style movie cameras. Another way is to control the duration for which the light illuminates the object.

1.1.2.1 Framing Camera

The fastest mechanical shutter speed is approximately 1 ms, but much faster shutters (down to <100 ps) have been constructed electronically. The electronic camera works by first converting the light images into electron replicas. The electron images are then gated, by turning the voltage applied to a microchannel plate (MCP) on and off. MCPs are commonly used in detectors for electrons, ions, and high-energy photons (UV to x-ray). An MCP looks like a round thin glass plate, with a diameter of 20–100 mm and thickness of 0.5 mm. It is made by fusing several millions of thin glass tubes together. The diameter of each tube or channel is approximately $10\text{ }\mu\text{m}$. The wall of each tube is approximately $1\text{ }\mu\text{m}$. When an incident particle or photon hits the input surface, it knocks out electrons. The electrons are accelerated inside the channels by the electric field created by applying a 1000 V voltage difference between the front and the back surfaces of the plate. The electrons may hit the wall of the channel and knock out more electrons. This process is repeated approximately 12 times. As a result, for one incident particle, 1000 electrons can be released from the rear surface, which yields a gain of 1000 per plate. Two plates can be stacked together to provide a gain of 10^6 , which is sufficient for many applications. When a pulse voltage is applied on the MCP, the gain can be turned on and off very rapidly, which serves as a shutter.

Figure 1.5 shows plasma evolution in a cylindrical target bombarded by 50 high-power laser beams. In the experiment, the beams from various directions were focused at the target center to concentrate their intensities, which pinched the cylinder. The MCP was coated with four pairs of

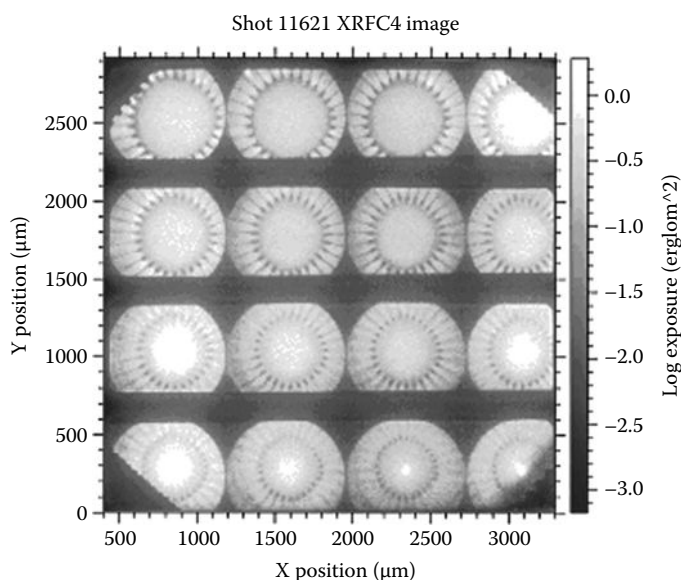


Figure 1.5 A movie of laser produced plasma. (Reprinted from Voss, S.A. et al., *IEEE Trans. Plasma Sci.*, 27, 132, 1999. With permission.)

horizontal strip electrodes, which could be gated one after another. The electric gating pulses propagated from left to right. The images started at the top left corner of the photo and moved to the right, and then from the left of the second row to the right, etc. Each image started as the previous one ended. The whole sequence lasted for approximately 1 ns. The images were taken for studying laser fusion, a possible clean energy source when the fossil fuels run out. The National Ignition Facility in California uses similar cameras to take images of the fusion target irradiated by 192 laser beams, with a total energy of 1.8 MJ. The initial confinement fusion happens on the nanosecond timescale.

MCPs are also used in time-of-flight (TOF) electron spectrometers for measuring attosecond pulses, as well in XUV grating spectrometers to measure attosecond spectra, as discussed in Chapters 4, 7, and 8. In these applications, DC voltages are applied to the MCPs.

The stroboscope is another tool for imaging fast processes. It uses a series of flashes to illuminate a moving object. In this case, the exposure time is the duration of each flash. It is obvious that the time resolution of a stroboscope can be set at a new level using attosecond light pulses.

1.1.2.2 Streak Camera

The shutter time can be reduced even further, down to a subpicosecond, if one spatial dimension is imaged. Such a device is called a streak camera. The principle of an optical streak camera, which works by converting temporal information into spatial information, is depicted in the left graph in Figure 1.6. The time resolution Δt is determined primarily by the scanning speed v_s . The key component of an electronic streak camera is a streak tube. A schematic of a conventional streak tube is shown in the right graph in Figure 1.6. It consists of a photocathode, a focusing lens, a deflection plate, and a phosphor screen. When the optical pulses are incident on the photocathode through a slit, a narrow beam of electron pulses is released into vacuum. The photoelectric conversion is considered to be instantaneous. In other words, the electron pulse can be considered a

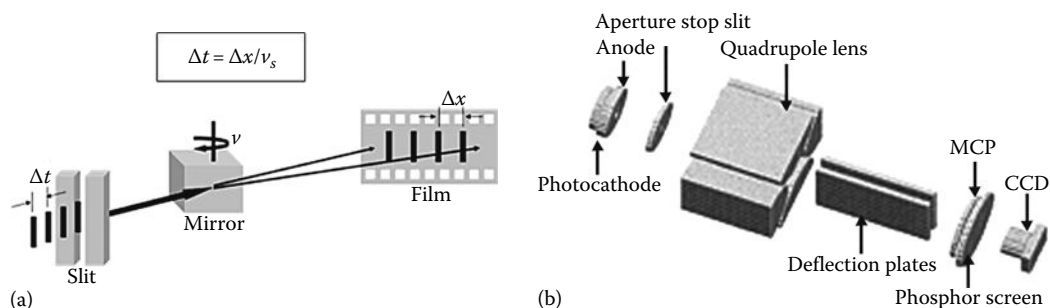


Figure 1.6 (a) The principle of measurement of four light pulses shape by an optical streak camera. The rotating mirror sweeps the light pulses on a film. v_s is the linear scanning velocity on the film. The time interval, Δt , can be determined from the spacing of the spots, Δx , on the film by a simple equation. (b) An electronic streak camera with 280 fs resolution. (Reprinted from Shakya, M.M. and Chang, Z., *Appl. Phys. Lett.*, 87, 041103, 2005. With permission.)

replica of the optical pulses. The electron optic lens images the slit to the phosphor screen, which can then be recorded by a CCD camera. The kinetic energy of the photoelectrons is concentrated to a small range close to zero, which leads to a small chromatic aberration. When a linearly ramping voltage is applied to the deflection plates, it sweeps the electron across the phosphor screen in the direction perpendicular to the slit. Different temporal portions of the electron pulse are displaced at different positions on the screen. For a known sweep speed, one can determine the electron pulse profile and thus the optical pulse profile. The scanning speed in such a device can exceed the speed of light in vacuum, c , and thus very high temporal resolution can be achieved.

Streak cameras sensitive to x-rays can measure optical pulses down to subpicoseconds and do not rely on nonlinear effects, which have also been used to study laser fusions. When streak cameras are used in synchrotron facilities, subpicosecond dynamics in crystals are studied by time-resolved diffraction, although the x-ray pulses from the synchrotron are 100 ps long. A factor that limits the resolution is the time spread due to initial velocity difference of the electrons when they travel from the photocathode to the deflection field. The streaking technique has been extended to measure attosecond light pulses, which is discussed in Chapter 8.

1.1.2.3 Pump–Probe Technique

For imaging fast processes, synchronization between the events and the camera is crucial. In the stroboscope, the flashing light can be considered as a “probe” light, whereas the signal that triggers the process is the “pump.” The pump–probe technique has been widely applied to study many aspects of fast processes, such as emission, absorption, and reflection, because one can use slow detectors to record a fast process. The time resolution is determined by the length of the probe pulse. It is particularly useful when the process is reproducible. A typical pump–probe setup is shown in Figure 1.7. It is a common practice to use a Mach–Zehnder interferometer to control the time delay between the pump and probe pulses. The advantage of the stroboscope and the pump–probe is that the response of the detector can be slow. In experiments, the fast events are measured as a function of time delay. Examples of attosecond pump–femtosecond laser probe experiments are given in Chapter 8.

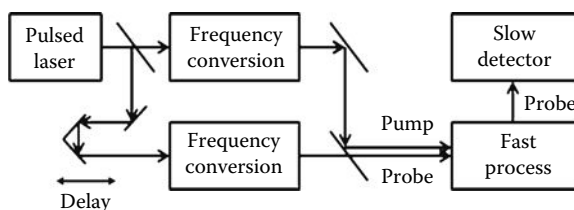


Figure 1.7 A pump–probe setup.

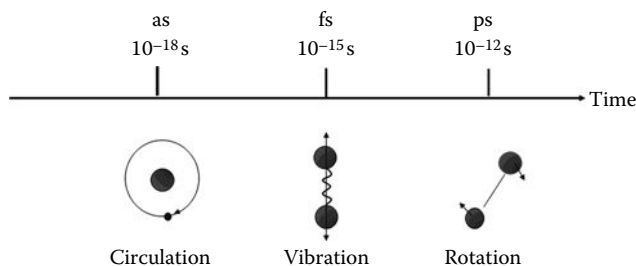


Figure 1.8 Timescale of dynamics in atoms and molecules.

1.1.3 Timescale of Electron Dynamics: The New Frontier

Ultrafast and high-power lasers operate in the UV, visible, and IR spectrum, and have been used extensively in the study of ultrafast processes on the nanosecond to femtosecond timescales. In molecules, the rotational and vibrational periods are on the order of picoseconds and femtoseconds, respectively, as illustrated in Figure 1.8. Consequently, femtosecond lasers have been an indispensable tool for studying molecular dynamics. In 1999, Ahmed H. Zewail won the Nobel Prize in Chemistry for his studies of the transition states of chemical reactions using femtosecond spectroscopy.

1.1.3.1 Atomic Unit of Time

In Bohr's hydrogen atom, the electron in the ground state circulates the nucleus in a time period,

$$T_H = 2\pi \frac{\hbar}{2I_{p,H}} = 2\pi T_a = 152 \text{ as}, \quad (1.13)$$

where

$I_{p,H} = 13.6 \text{ eV}$ is the ionization potential of the hydrogen atom

$T_a = 24.2 \text{ as}$ is the atomic unit of time, which is the time for the electron to travel across a distance that equals the Bohr radius

To shoot a movie of the electrons around the orbit in this old quantum model, the exposure time of each frame should be on the order of 10 as, as depicted in Figure 1.9. In fact, the atomic unit of time is the characteristic

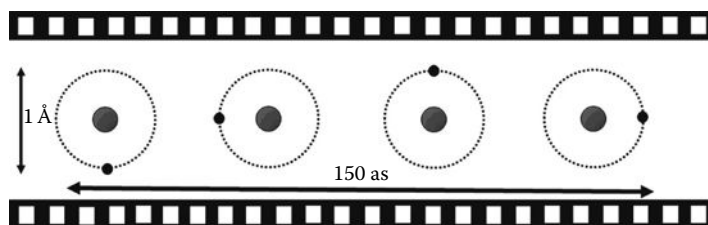


Figure 1.9 A movie of an electron circulating a proton in the hydrogen atom. The quantum effects are ignored.

timescale of electron motion in many atoms and molecules. The study of electron dynamics in atoms and molecules calls for optical pulses of attosecond duration. Such extremely short pulses were first measured in 2001. Chapter 9 introduces some proof-of-principle experiments using attosecond optical pulses.

In the frequency domain, a transform-limited Gaussian pulse with 24 as FWHM corresponds to a 73 eV FWHM power spectrum, which is much broader than the entire visible light range. In other words, attosecond pulses are inherently XUV light or x-rays.

1.2 Attosecond Light Pulses

Ultrafast lasers in the near infrared (NIR) and visible wavelength range have been developed for the aforementioned applications. Since the invention of the laser in 1960, the duration of coherent NIR-visible pulses has decreased from hundreds of microseconds to 6 fs, as shown in Figure 1.10. Various pulse shortening techniques have been invented, such as mode locking, which is discussed in Chapter 2. However, by the year 1987, the optical pulse length was approaching the limit, i.e., one optical cycle of visible light, which is only a few femtoseconds. Progress has been made on ultrafast x-ray lasers based on stimulated emissions in laser produced plasma, but the pulse duration of such lasers is still longer than 1 ps. X-ray free electron lasers that are under construction may reach pulses around 100 fs. The costs of building one of these lasers are on the one-billion-dollar level. A breakthrough happened in 2001, when XUV pulses shorter than 1 fs were generated by interacting atoms with intense femtosecond NIR lasers.

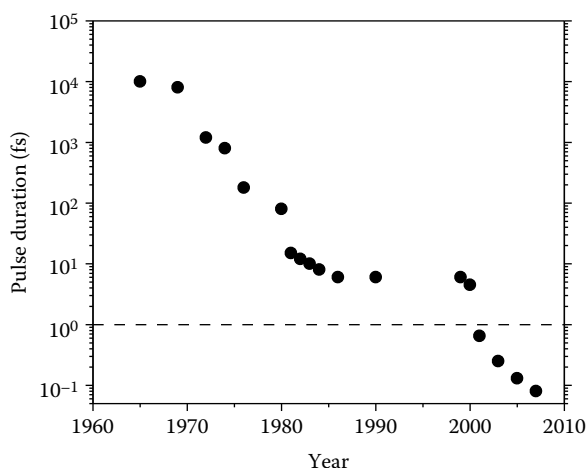


Figure 1.10 The progress of ultrafast lasers. (Adapted from P.B. Corkum and Z. Chang, The attosecond revolution, *Opt. Photon. News*, 19, 24, 2008. With permission of Optical Society of America.)

1.2.1 Mathematical Description of Attosecond Optical Pulses

Coherent attosecond optical pulses are laser-like pulses. Thus, the theory of ultrafast laser optics can be applied to attosecond pulses.

1.2.1.1 Time Domain

Equation 1.14 is the electric field for a linearly polarized CW wave at a given point in space. Similarly, a coherent optical pulse can be expressed by its time-dependent electric field

$$\varepsilon(t) = E(t) \cos[\omega_0 t + \phi(t)]. \quad (1.14)$$

It is evident from Equation 1.14 that a pulse is determined by three quantities: $E(t)$, the time-dependent amplitude (envelope); ω_0 , the carrier (center) angular frequency; and $\phi(t)$, the temporal phase.

Although the electric field is a real physical quantity, mathematically it is more convenient to use the complex representation. Therefore, we can express the field as

$$\varepsilon(t) = E(t)e^{+i[\omega_0 t + \phi(t)]}, \quad (1.15)$$

where $i = \sqrt{-1}$. In this book, we adopt the sign convention for the exponent used in the electric engineering community. The same sign convention is used in Fourier transforms, discussed in greater detail in later chapters. In atomic physics literatures, a different sign convention has been used, where the sign before i in the exponent is negative.

For Gaussian pulses,

$$\varepsilon(t) = E_0 e^{-2 \ln 2 \left(\frac{t}{\tau}\right)^2} e^{i[\omega_0 t + \phi(t)]}, \quad (1.16)$$

where E_0 is the peak amplitude. The intensity profile is given by

$$I(t) = I_0 e^{-4 \ln 2 \left(\frac{t}{\tau}\right)^2}. \quad (1.17)$$

The peak intensity I_0 and the peak field amplitude are related by

$$I_0 = \frac{1}{2} \epsilon_0 c E_0^2. \quad (1.18)$$

A pulse is Fourier transform limited when the temporal phase equals a constant or is a linear function of time. The linear dependence of the temporal phase is equivalent to a shift of the center frequency, as seen in Equation 1.16. [Figure 1.11](#) shows a transform-limited 25 as pulse with an 8 nm carrier wavelength.

The carrier wavelength, λ_0 , the optical period, T_0 , and the central angular frequency, ω_0 , are related to one another by

$$\lambda_0 = c T_0 = c \frac{2\pi}{\omega_0}. \quad (1.19)$$

In this example, $T_0 = 26.7$ as, $\omega_0 = 0.24$ rad/as. The corresponding photon energy is

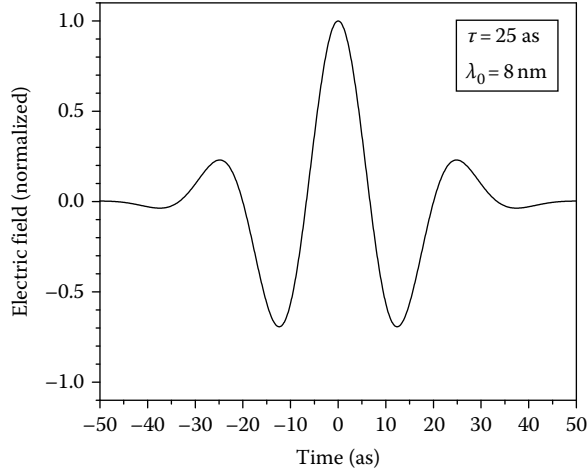


Figure 1.11 The electric field of a transform-limited 25 as Gaussian pulse.

$$\hbar\omega_0[\text{eV}] = \frac{1240}{\lambda_0[\text{nm}]} = 155 \text{ eV}. \quad (1.20)$$

The pulse can be considered as a single cycle when $\tau \approx T_0$. The pulse in Figure 1.11 is a single cycle pulse.

1.2.1.2 Temporal Phase and Chirp

A pulse is said to be chirped when the instantaneous frequency of the pulse changes with time. If the temporal phase is a quadratic function of time, i.e., $\phi(t) = bt^2$ (where b is the chirp parameter), then the chirp is linear. The instantaneous frequency can be expressed mathematically as

$$\omega(t) \equiv \frac{d}{dt}[\omega_0 t + \phi(t)] = \omega_0 + 2bt. \quad (1.21)$$

The pulse is positively chirped when $b > 0$. In this case, the instantaneous frequency under the pulse envelope increases with time. Figure 1.12

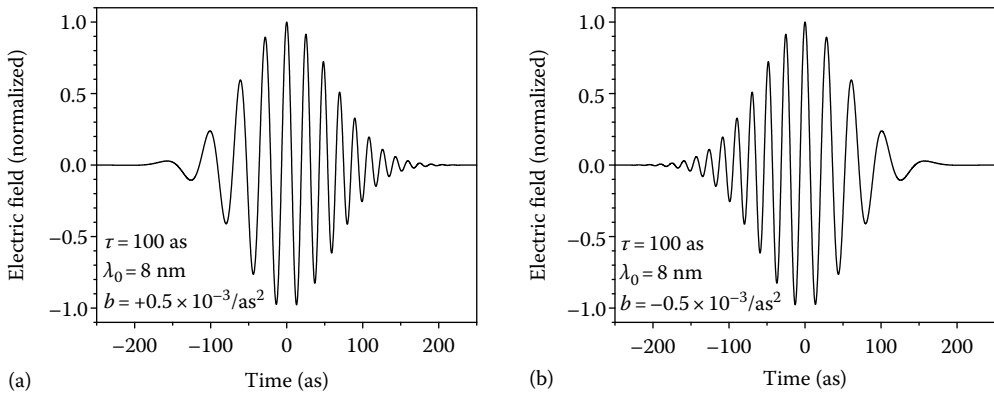


Figure 1.12 (a) A positively chirped pulse and (b) a negatively chirped pulse.

shows two 100 as Gaussian pulses centered at 8 nm; one is positively chirped ($b = +0.5 \times 10^{-3} \text{ as}^{-2}$) and the other is negatively chirped ($b = -0.5 \times 10^{-3} \text{ as}^{-2}$). It is evident that the field oscillation becomes faster from cycle to cycle under the envelope for positively chirped pulses.

Like laser pulses, a linearly chirped Gaussian pulse can be specified with a complex parameter

$$\Gamma = a - ib, \quad (1.22)$$

where the pulse width parameter is defined as

$$a = 2 \ln 2 \frac{1}{\tau^2}. \quad (1.23)$$

The pulses expressed using the Γ parameter are given by

$$\varepsilon(t) = E_0 e^{-\Gamma t^2} e^{i\omega_0 t}. \quad (1.24)$$

1.2.1.3 Frequency Domain

The pulse can also be described in the frequency domain, where the electric field is obtained by performing a Fourier transform of the field in the time domain

$$\tilde{E}(\omega) = \int_{-\infty}^{+\infty} \varepsilon(t) e^{-i\omega t} dt. \quad (1.25)$$

Two different sign conventions in the transform have been used in the physics and engineering communities. In this book, we adopt the latter, which is why the sign in the exponent is negative. In general, the transform gives

$$\tilde{E}(\omega) = U(\omega) e^{i\varphi(\omega)}, \quad (1.26)$$

where

$U(\omega)$ is the spectrum amplitude

$\varphi(\omega)$ is the spectral phase

Of the four quantities that describe light pulses, $U(\omega)$ is most easily measured. The power spectrum, $U^2(\omega)$, of attosecond pulses can be measured with a grating spectrometer. It can also be converted into electrons and measured by TOF spectrometers. Chapters 7 and 8 discuss techniques for measuring the spectral phase of attosecond pulses. Once $U(\omega)$ and $\varphi(\omega)$ are determined, we can describe the pulse in both the spectral and the temporal domain.

For a Gaussian pulse with a linear chirp, the spectral amplitude can be expressed as

$$U(\omega) = U_0 e^{-2 \ln 2 \frac{(\omega - \omega_0)^2}{\Delta\omega^2}}, \quad (1.27)$$

which is also a Gaussian function. The spectral phase is then given by

$$\varphi(\omega) = -\frac{1}{4} \left(\frac{b}{a^2 + b^2} \right) (\omega - \omega_0)^2, \quad (1.28)$$

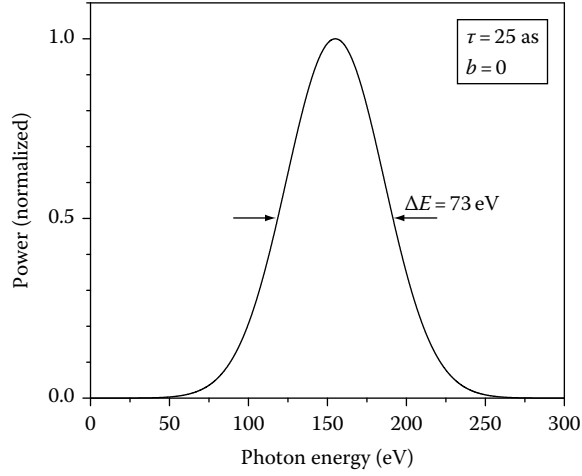


Figure 1.13 Power spectrum of a transform-limited 25 as pulse.

which is a parabolic function. For a positively chirped pulse, $b > 0$, the prefactor of the spectral phase, $-1/4(b/a^2 + b^2)$, is negative.

The FWHM of the power spectrum is

$$\Delta\omega = \frac{4 \ln 2}{\tau} \sqrt{1 + \left(\frac{b}{a}\right)^2}. \quad (1.29)$$

The bandwidth in terms of the photon energy is $\Delta\mathcal{E} = \hbar\Delta\omega$, which is 73 eV for a transform-limited 25 as pulse. The power spectrum of the 25 as pulse that corresponds to Figure 1.11 is shown in Figure 1.13. Notice that the wings of the spectrum extend to a range from 50 to 270 eV, approximately 220 eV, which is approximately three times that of the FWHM. An electron TOF spectrometer for measuring such a broadband spectrum with high resolution is discussed in Chapter 7.

1.2.1.4 Time-Bandwidth Product

We know from Equation 1.29 that the product of the pulse width and the spectral bandwidth of a linearly chirped Gaussian pulse satisfies the relation

$$\tau \cdot \Delta\mathcal{E} = 4 \ln 2 \sqrt{1 + \left(\frac{b}{a}\right)^2} \hbar. \quad (1.30)$$

For transform-limited Gaussian pulses, $\tau[\text{as}] \cdot \Delta\mathcal{E}[\text{eV}] = 1825$. Thus, for pulses shorter than 1 fs, the spectral bandwidth should be larger than 1.8 eV, and the wings of the spectrum extend to a 5.5 eV range, which is much larger than the range of visible light.

Equation 1.30 is a manifestation of the uncertainty principle in quantum mechanics,

$$\Delta t \cdot \Delta E \geq \hbar. \quad (1.31)$$

Here, Δt and ΔE are the uncertainties in simultaneous time and energy measurements.

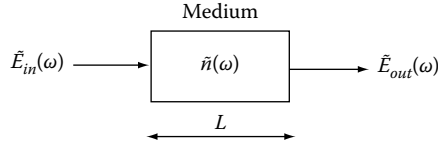


Figure 1.14 Pulse propagation through a linear medium.

1.2.2 Propagation of Attosecond Pulse in Linear Dispersive Media

The bandwidth of Gaussian pulses of 100 to 1 as duration is in the photon energy range of 18 eV–1.8 keV, which correspond to radiation ranges of XUV to soft x-rays. Unlike visible and NIR light, such pulses cannot propagate through air. The requirement of a vacuum environment leads to an inconvenience in the generation, measurement, and application of attosecond pulses.

Consider the case that the intensity of the XUV light is weak, so that the nonlinear response of the medium can be neglected. A medium can be described by a frequency-dependent index of refraction; therefore, it is much easier to handle as a propagation problem in the frequency domain. When a plane wave propagates in an absorptive and dispersive material, as depicted in Figure 1.14, the electric field exiting the medium can be described in the frequency domain

$$\tilde{E}_{out}(\omega) = \tilde{E}_{in}(\omega)e^{-i\omega\tilde{n}(\omega)L}, \quad (1.32)$$

where

L is the length of the medium

$\tilde{n}(\omega) = n_R(\omega) - in_I(\omega)$ is the complex index of refraction of the material

Note the negative sign in front of $in_I(\omega)$, which is the convention used in optics.

1.2.2.1 Index of Refraction and Scattering Factor

The frequency of a wave does not change when entering a linear medium from vacuum, but the wavelength changes from the value in vacuum, λ , to the value in the medium, $\lambda_m = \lambda/n_R(\lambda)$. For XUV and x-ray, the complex index of refraction at wavelength λ is expressed in terms of the atomic scattering factors, f_1 and f_2 , by the expression

$$\tilde{n}(\lambda) = 1 - \frac{1}{2\pi} N r_e \lambda^2 (f_1 + if_2), \quad (1.33)$$

where N is the number of atoms per unit volume. The classical electron radius, r_e , is found by

$$r_e = \frac{1}{4\pi\epsilon_0} \frac{e^2}{m_e c^2} = 2.82 \times 10^{-15} \text{ m}. \quad (1.34)$$

The values of the scattering factors of atoms in the 10–30,000 eV photon energy range can be found at the Advanced Light Source online database.

For example, the scattering factors for argon and Al are shown in Figures 1.15 and 1.16.

Some fine structures, such as the Fano resonance discussed in Chapter 9, are not included. It is also worth pointing out that the values are based on photoabsorption measurements of elements in their elemental state. For molecules or condensed matter, it is assumed that they may be modeled as a collection of noninteracting atoms. This assumption may not be valid for energies close to the absorption thresholds.

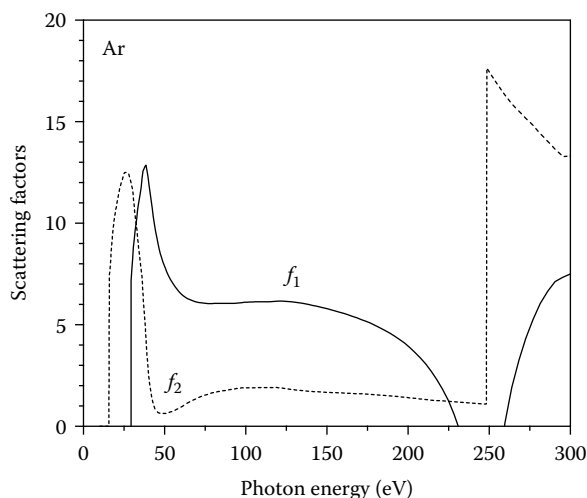


Figure 1.15 Scattering factor of an Ar atom. (Calculated using Advanced Light Source, Lawrence Berkeley National Lab, http://henke.lbl.gov/optical_constants/, accessed April 12, 2010.)

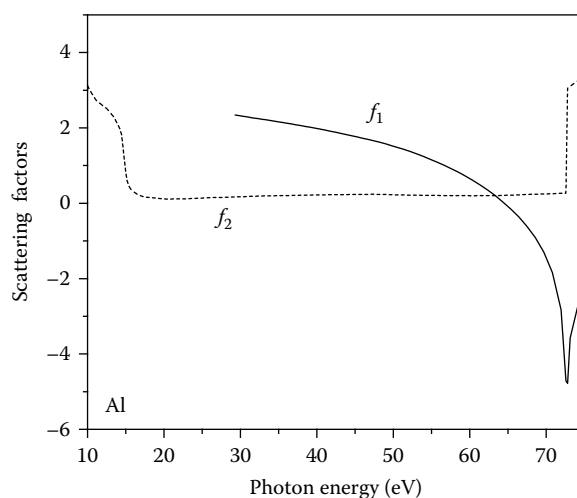


Figure 1.16 Scattering factor of an Al atom. (Calculated using Advanced Light Source, Lawrence Berkeley National Lab, http://henke.lbl.gov/optical_constants/, accessed April 12, 2010.)

1.2.2.2 Photoabsorption Cross Section and Transmission

The photoabsorption cross section is related to the scattering factor by

$$\sigma(\lambda) = 2r_e\lambda f_2. \quad (1.35)$$

For a monochromatic light expressed by Equation 1.4, the change of the electric field amplitude is

$$E_{0,out} = E_{0,in} e^{-\frac{1}{2}N\sigma(\lambda)L}. \quad (1.36)$$

The transmittance, T , is defined as the ratio of the output intensity to the input intensity

$$T(\lambda) = e^{-N\sigma(\lambda)L}. \quad (1.37)$$

The transmittance can also be expressed in term of absorption coefficient

$$\alpha(\lambda) = N\sigma(\lambda), \quad (1.38)$$

in essence,

$$T(\lambda) = e^{-\alpha(\lambda)L}, \quad (1.39)$$

which is called the Beer–Lambert law.

1.2.2.3 Gas Medium

As discussed later, attosecond pulses are normally generated in gases. Therefore, Equation 1.37 can be rewritten as

$$T(\lambda) = e^{-N_0\sigma(\lambda)P_{gas}L}, \quad (1.40)$$

where N_0 is the number density of the gas, equaling 2.5×10^{19} atoms/cm³ for a standard gas at room temperature and atmospheric pressure. P_{gas} is therefore the gas pressure in the unit of atmosphere. It is clear that the transmission is determined by the pressure–length product, $P_{gas}L$, instead of the length alone, as in the case of absorption by solid materials. As an example, the transmission of 5 mm of argon gas with a pressure of 50 torr in the 10–100 eV range is shown in Figure 1.17. The transmission is ~50% in the 20–250 eV range. When argon gas is used to generate attosecond pulses, the absorption by the generation medium poses an ultimate limitation on the conversion efficiency from the driving laser energy to the attosecond pulse energy, as discussed in Chapter 6.

The transmission for 0.5 m and a pressure of 500 mtorr is the same as in Figure 1.17. Thus, a good vacuum is required for a long propagation distance.

The absorption for an XUV photon leads to the ionization of the gas atoms. The photoabsorption cross section in Equation 1.35 is also called the photoionization cross section. In an attosecond streak camera, the absorption by the gas atoms is used to convert attosecond photon pulse into its electron replica, as discussed later in this chapter.

1.2.2.4 Thin Film

Thin metal filters are frequently used in attosecond pulse generation to block NIR laser light and transmit attosecond XUV light. The transmission

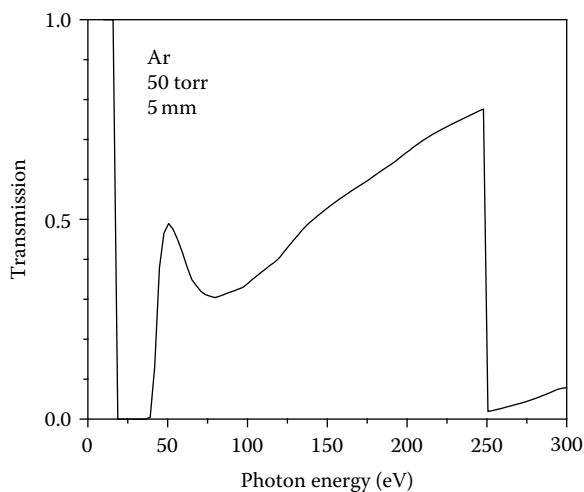


Figure 1.17 Transmission of argon gas. (Calculated using the tools provided at the Advanced Light Source, Lawrence Berkeley National Lab, http://henke.lbl.gov/optical_constants/gastrn2.html, accessed April 12, 2010.)

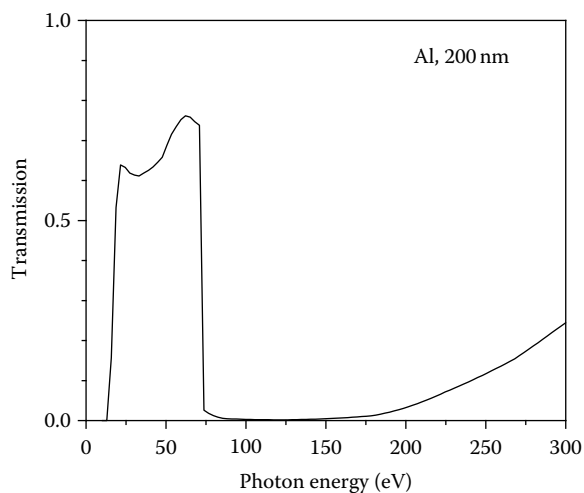


Figure 1.18 Transmission of a 200 nm Al filter. (Calculated using Advanced Light Source, Lawrence Berkeley National Lab, http://henke.lbl.gov/optical_constants/filter2.html, accessed April 12, 2010.)

of a 200 nm Al filter is shown in Figure 1.18. It is a good band-pass filter in the 20–70 eV range. Such filters are extremely fragile.

1.2.2.5 Spectral Phase

A frequency-dependent index of refraction means that the phase velocities of each frequency component of an attosecond pulse are different. Consequently, group velocity dispersion (GVD) is introduced, and pulse broadening or compression may occur in a dispersive media. The GVD can be determined from the spectral phase.

When a transform-limited pulse propagates through the medium shown in Figure 1.14, the spectral phase introduced by the dispersion is given by

$$\varphi(\omega) = -\beta(\omega)L. \quad (1.41)$$

The propagation constant, which is the phase shift per unit length, can be expressed as

$$\beta(\omega) = \frac{\omega}{c} n_I(\omega) = \frac{2\pi}{\lambda} n_I(\lambda) = \frac{2\pi}{\lambda_m}. \quad (1.42)$$

Here, we use the symbol β instead of k in discussing pulse propagation.

It can be expanded into a Taylor series in the following manner:

$$\begin{aligned} \beta(\omega) = & \beta(\omega_0) + \left. \frac{d\beta}{d\omega} \right|_{\omega_0} (\omega - \omega_0) + \frac{1}{2} \left. \frac{d^2\beta}{d\omega^2} \right|_{\omega_0} (\omega - \omega_0)^2 \\ & + \sum_{m=3}^{\infty} \frac{1}{m!} \left. \frac{d^m\beta}{d\omega^m} \right|_{\omega_0} (\omega - \omega_0)^m. \end{aligned} \quad (1.43)$$

1.2.2.6 Carrier-Envelope Phase

The first term in this expansion is a constant phase shift, which determines the phase delay of the carrier wave. It is related to the phase velocity by

$$v_p = \frac{n_I(\omega)}{c} = \frac{\beta(\omega_0)}{\omega_0}. \quad (1.44)$$

The second term is related to the group velocity of the pulses by

$$v_g = \frac{1}{\left. \frac{d\beta}{d\omega} \right|_{\omega_0}}, \quad (1.45)$$

which causes a delay of the pulse envelope.

These first two terms become important when the carrier-envelope (CE) phase, φ_{CE} , is considered. The CE phase is defined as the offset between the peak of the pulse envelope and the peak of the carrier oscillation, as shown in Figure 1.19 for $\varphi_{CE}=0$, and Figure 1.20 for $\varphi_{CE} = -\pi/2$ rad.

The shift of the CE phase in a dispersive material is

$$\Delta\varphi_{CE} = \left(\frac{1}{v_p} - \frac{1}{v_g} \right) L, \quad (1.46)$$

where the quantity

$$\frac{L}{v_g} = L \left. \frac{d\beta}{d\omega} \right|_{\omega_0} = t_g, \quad (1.47)$$

is the group delay.

Although the effects of the CE phase of attosecond pulses have not yet been studied experimentally, it has been demonstrated that the CE phase of high-power NIR lasers plays a critical role in the generation of attosecond pulses. The techniques of stabilizing and controlling the CE phase of femtosecond lasers are discussed more thoroughly in Chapter 3.

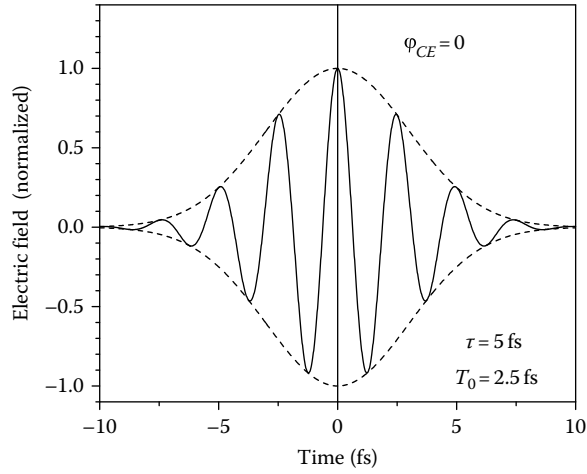


Figure 1.19 Electric field of a 5 fs pulse. The carrier-envelope phase is 0 rad.

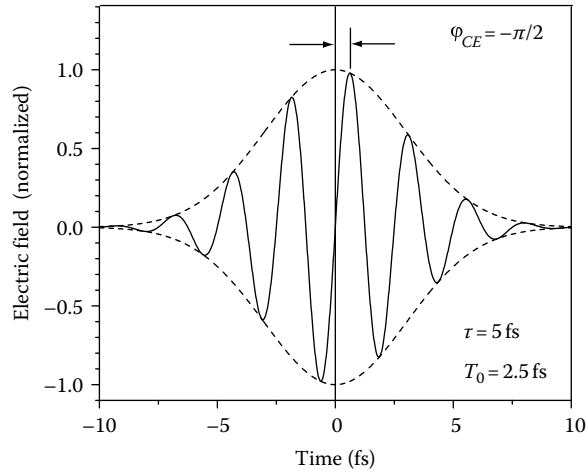


Figure 1.20 Electric field of a 5 fs pulse. The carrier-envelope phase is $-\pi/2$ rad.

1.2.2.7 Group Velocity Dispersion and Group Delay Dispersion

The third term in the Taylor expansion causes the variation of the pulse width. The group velocity dispersion is defined as

$$GVD = \frac{d^2\beta}{d\omega^2} \Big|_{\omega_0} = \frac{d}{d\omega} \left(\frac{1}{v_g} \right) = -\frac{1}{v_g^2(\omega_0)} \frac{dv_g}{d\omega}. \quad (1.48)$$

By examining Equation 1.48, it is clear that the unit of GVD is $[\text{time}^2]/[\text{length}]$, for example, as^2/mm .

Likewise, the group delay dispersion (GDD) is defined as

$$\text{GDD} = -\frac{d^2\varphi}{d\omega^2} \Big|_{\omega_0}, \quad (1.49)$$

where the unit of GDD is [time²]. For attosecond pulses, it is convenient to use [as²]. The dispersion of mirrors can be specified by GDD. For the dispersive materials considered here,

$$\text{GDD} = \text{GVD} \cdot L. \quad (1.50)$$

The later terms in the Taylor series are called high-order phases, and can be ignored when the spectrum is narrow. For the generation of few-cycle NIR lasers, the distortion of high-order phases (the third to the fifth order) should be considered.

GDD can also be defined by the group delay in the following way:

$$-\frac{d^2\varphi}{d\omega^2} = -\frac{d}{d\omega} \left(\frac{d\varphi}{d\omega} \right) = \frac{dt_g}{d\omega}. \quad (1.51)$$

Using this definition, the unit of GDD is attosecond per electronvolt. The two units are related by

$$\text{GDD}[\text{as}^2] = \frac{\text{GDD}[\text{as}/\text{eV}]}{1.516} \times 10^3. \quad (1.52)$$

1.2.2.8 Pulse Broadening and Compression

For a transform-limited input pulse with width τ_{in} , if we neglect the higher order phases, the output pulse duration can be calculated by the expression

$$\tau_{out} = \tau_{in} \sqrt{1 + \left(\frac{L}{z_D} \right)^2}, \quad (1.53)$$

where the dispersion length

$$z_D = \frac{\tau_{in}^2}{(4 \ln 2) \text{GVD}}. \quad (1.54)$$

Equation 1.53 can be rewritten as

$$\tau_{out} = \tau_{in} \sqrt{1 + \left(4 \ln 2 \frac{\text{GDD}}{\tau_{in}^2} \right)^2}, \quad (1.55)$$

which can also be applied to mirrors.

If the input pulse is positively chirped, then the pulse duration will be shortened by a material with negative GVD. The pulse is stretched when the GVD is positive. Consequently, for initially negatively chirped pulses, the opposite is true.

1.2.2.9 GVD of Filters

To calculate the index of refraction, we first need to know the atomic density. For Al, the density is 2.7 g/cm³ and the mass of each Al atom is $27m_p$, where $m_p = 1.67 \times 10^{-24}$ [g] is the mass of the proton. Thus the atomic number density of solid Al is $N = 1 \times 10^{23}$ cm³. From Equation 1.33, we know

$$\tilde{n}(\lambda) - 1 = -\frac{1}{2\pi} \times 1 \times 10^{23} \times 2.82 \times 10^{-13} \times \left(\frac{1.24}{\hbar\omega} \times 10^{-4} \right)^2 (f_1 + if_2), \quad (1.56)$$

where the photon energy is in electronvolts. The real part, $n_R(\lambda) = \text{Re}[\tilde{n}(\lambda)]$ is related to the GVD. The $n_R - 1$ for argon gas and solid Al are shown in Figures 1.21 and 1.22. We can see that the index of refraction can be less than one for XUV light, which implies that the phase velocity is larger than the speed of light in vacuum.

The propagation constant difference is

$$\beta(\omega) - \beta_0(\omega) = \frac{\omega}{c} [n_R(\omega) - 1], \quad (1.57)$$

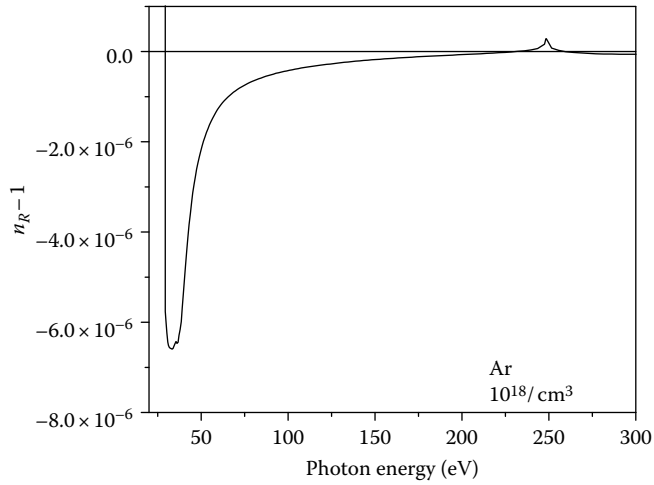


Figure 1.21 The real part of the index of refraction of argon gas with 30 torr pressure.

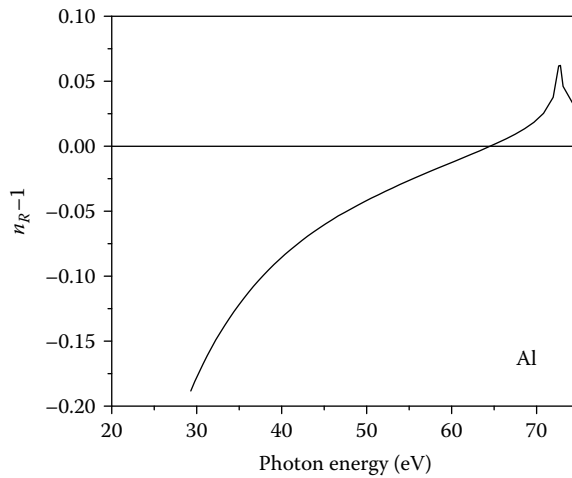


Figure 1.22 The real part of the index of refraction of Al.

where β_0 is the propagation constant in vacuum. The values are shown in Figures 1.23 and 1.24 for argon and Al, respectively.

The group delay in the medium per unit distance minus that in the vacuum, t_{g0} , is then given by

$$t_g(\omega) - t_{g0} = \frac{d(\beta - \beta_0)}{d\omega} = -\frac{1}{c} \left[n_R(\omega) - 1 + \hbar\omega \frac{dn_R(\omega)}{d(\hbar\omega)} \right]. \quad (1.58)$$

Figures 1.25 and 1.26 show the group delays of 1 mm of argon gas at 30.4 torr and a 1000 nm Al filter, respectively. The corresponding GVDs are shown in Figure 1.27 and 1.28. It can be seen in Figure 1.28 that the GVD changes signs at ~ 50 eV. The GVD is negative below 50 eV, which can be used to compensate the positive chirp of the attosecond pulses (Figure 1.28).

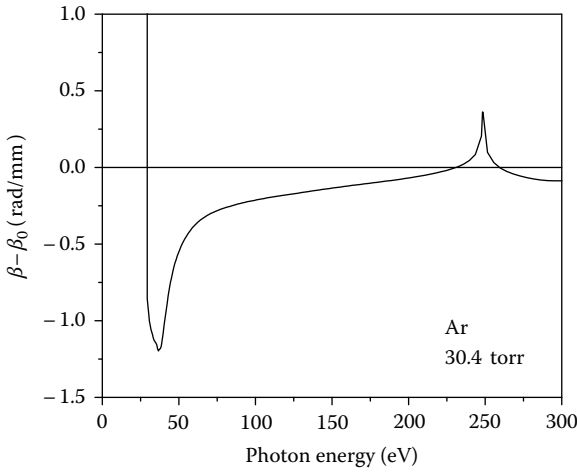


Figure 1.23 The propagation parameter of Ar.

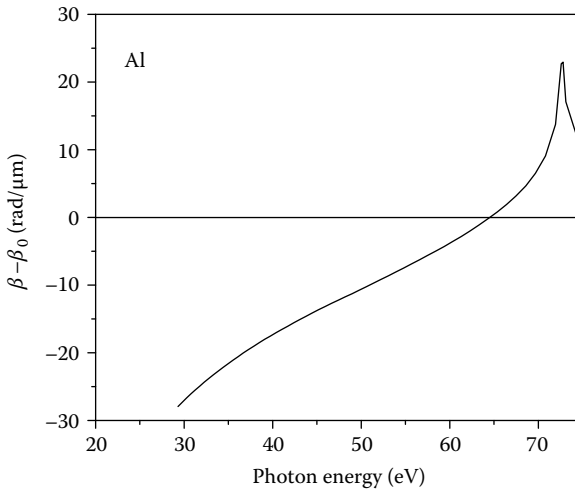


Figure 1.24 The propagation parameter of Al.

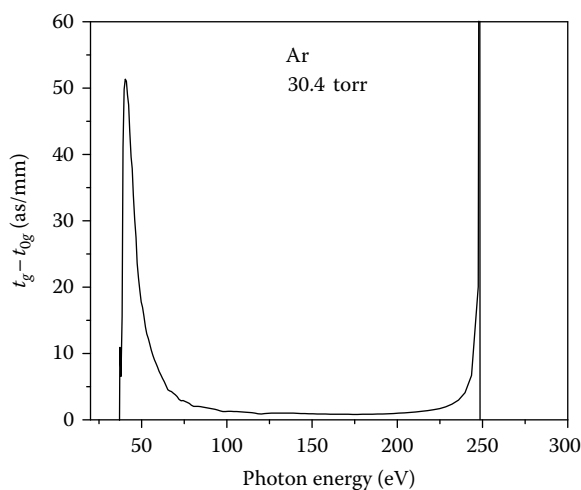


Figure 1.25 The group delay per mm in argon gas at 30.4 torr.

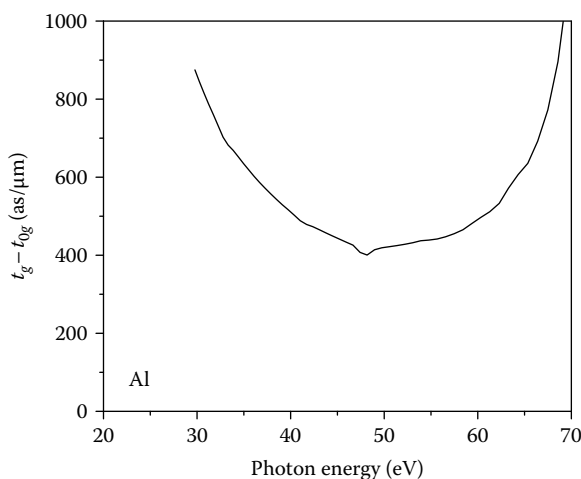


Figure 1.26 The group delay per micrometer in Al.

1.3 Overview of Attosecond Pulse Generation

As mentioned earlier, the FWHM bandwidth of 100 to 1 as duration is in the 18 eV to 1.8 keV range. No laser gain media has yet been found to support such a broad bandwidth, although the x-ray free electron laser may cover the bandwidth in the future. At the present time, the broadest bandwidth laser material is Ti:Sapphire, with its center wavelength at 790 nm. The FWHM of the gain cross section curve is 120 nm, corresponding to 2 fs.

Before the year 2000, post-laser pulse compression was done by broadening the spectrum through self-phase modulation, which is a third-order nonlinear process. It can extend the spectrum to near IR and UV, but the spectrum width is still much less than 18 eV.

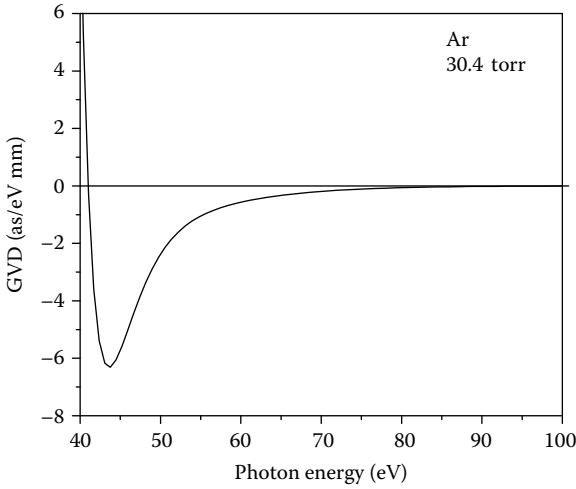


Figure 1.27 The GVD of argon gas at 30.4 torr.

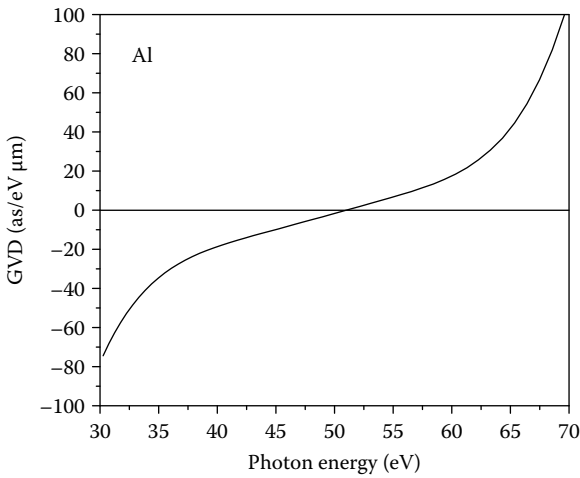


Figure 1.28 The GVD of Al.

1.3.1 Pulse Compression by Perturbative Harmonic Generation

Another approach to shorten pulses is by harmonic generation. For example, in second harmonic generation, the electric field amplitude of the frequency-doubled pulses is

$$E_{2\omega}(t) \propto E_{\omega}^2(t), \quad (1.59)$$

where $E_{\omega}(t)$ is the amplitude of the pulse at the fundamental frequency.

The intensity of the frequency-doubled pulses is

$$I_{2\omega}(t) \propto I_{\omega}^2(t), \quad (1.60)$$

where $I_{\omega}(t)$ is the intensity of the pulse at the fundamental frequency.

For Gaussian pulses, this leads to

$$e^{-4 \ln 2 \left(\frac{t}{\tau_{2\omega}}\right)^2} = \left[e^{-4 \ln 2 \left(\frac{t}{\tau_{\omega}}\right)^2} \right]^2 = e^{-8 \ln 2 \left(\frac{t}{\tau_{\omega}}\right)^2}, \quad (1.61)$$

where τ_{ω} and $\tau_{2\omega}$ are the FWHM of fundamental and second harmonic pulses, respectively. The two are related by

$$\tau_{2\omega} = \frac{\tau_{\omega}}{\sqrt{2}}. \quad (1.62)$$

Similarly, in the perturbative regime, for an even higher order nonlinear process, q ,

$$\tau_{q\omega} = \frac{\tau_{\omega}}{\sqrt{q}}. \quad (1.63)$$

Thus, we expect that a 5 fs pulse could be compressed to 0.5 fs by using a 100th order harmonic generation. The problem with this approach is that the conversion efficiency decreases exponentially with harmonic order. Consequently, the photon flux is extremely low for harmonic orders 100, which makes it useless for practical applications.

Perturbative harmonic generation occurs when the laser intensity is $< 10^{12} \text{ W/cm}^2$. Many solid materials will be damaged at an intensity $> 10^{12} \text{ W/cm}^2$. When atoms are used as the nonlinear medium, only the odd harmonics are generated due to the symmetry of the atom and the electric field. In this case, the intensity as a function of harmonic order is depicted in Figure 1.29.

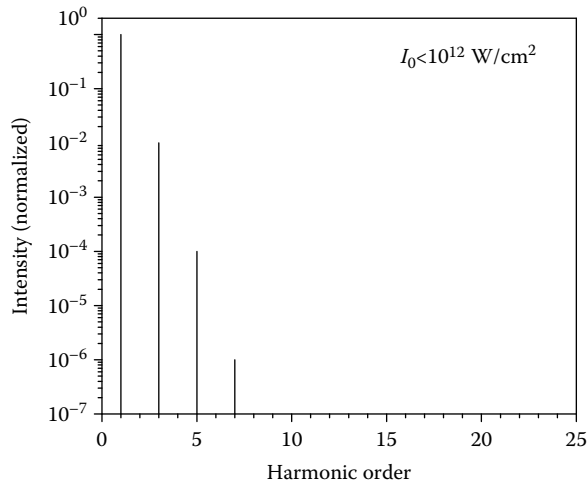


Figure 1.29 Power spectrum of harmonics generated in the perturbative regime.

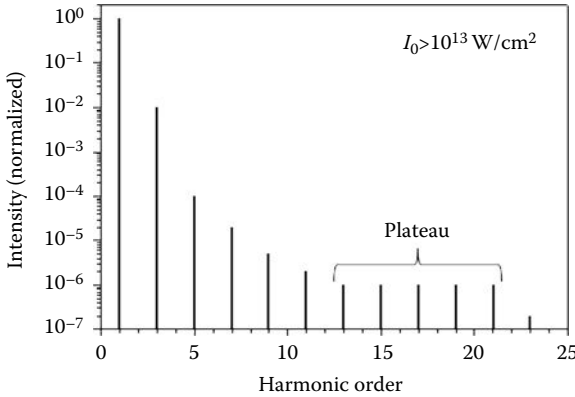


Figure 1.30 The power spectrum of high-order harmonics generated in the nonperturbative regime.

1.3.2 High-Order Harmonic Generation

A new type of harmonic generation phenomena was discovered around 1987–1988. When a linearly polarized, short pulse laser beam with intensity on the order of 10^{14} W/cm² interacts with noble gases, odd harmonics of the fundamental frequency—up to tens or even hundreds in order—emerge in the output beam. Noble gases were used because of their large binding energies. The harmonic spectrum is depicted in Figure 1.30. The intensity of the first few order harmonics decreases quickly as the order increases, after which the intensity remains almost unchanged over many harmonic orders, forming a plateau. Finally, the signal cuts off abruptly at the highest order. The appearance of the intensity plateau is the signature of this nonperturbative laser–atom interaction.

For NIR lasers, the photon energy is ~ 1.5 eV. In other words, the five plateau harmonics span 6 eV, which is broad enough to support attosecond pulses.

1.3.2.1 Attosecond Pulse Train

When several or more plateau harmonics are selected, we expect them to form an attosecond pulse train instead of a single isolated pulse, even if their phases are the same, because of the deep modulation of spectrum amplitude. A semi-classical model was proposed to explain the shape of the high harmonic spectrum, which also suggests that they correspond to an attosecond pulse train.

Suppose that the lowest order and the highest order in the plateau are q_L and q_H , respectively. Then the field in the time domain is

$$\varepsilon(t) = \sum_{q=q_L}^{q_H} E_q e^{i(q\omega_1 t + \varphi_q)}, \quad q = \text{odd numbers} \quad (1.64)$$

where

ω_1 is the carrier frequency of the laser

E_q and φ_q are the amplitude and phase of the q th harmonic

Assuming their amplitudes and phases are identical, then

$$\varepsilon(t) = E e^{i(q_L \omega_1 t + \varphi)} \sum_{q=0}^{q_H - q_L} e^{iq \omega_1 t}. \quad (1.65)$$

The sum results in

$$\frac{e^{i\left(\frac{q_H - q_L}{2} + 1\right)2\omega_1 t} - 1}{e^{i2\omega_1 t} - 1} = e^{i\frac{1}{2}(q_H - q_L)\omega_1 t} \frac{\sin\left[\left(\frac{q_H - q_L}{2} + 1\right)\omega_1 t\right]}{\sin \omega_1 t}. \quad (1.66)$$

Thus the field can be written as

$$\varepsilon(t) = E \frac{\sin\left[\left(\frac{q_H - q_L}{2} + 1\right)\omega_1 t\right]}{\sin \omega_1 t} e^{i\left[\frac{1}{2}(q_L + q_H)\omega_1 t + \varphi\right]}. \quad (1.67)$$

This field can be considered as a pulse with a carrier frequency equal to the frequency at the center of the plateau.

The intensity profile of the pulse is

$$I(t) = I_0 \frac{\sin^2\left[\left(\frac{q_H - q_L}{2} + 1\right)\omega_1 t\right]}{\sin^2 \omega_1 t}. \quad (1.68)$$

It has primary peaks at $t = 0, \pm T_1/2, \pm T_1, \dots$. The repetition rate of the primary pulses is half of an optical cycle, T_1 . For the five plateau harmonics in Figure 1.30, the intensity distribution in the time domain is shown in Figure 1.31. Here, we assume the period of the fundamental wave is 2.5 fs, corresponding to 750 nm center wavelength.

The width of each primary peak is

$$\Delta t = \frac{T_1}{\frac{q_H - q_L}{2} + 1}, \quad (1.69)$$

which can be shorter than 1 fs when there are sufficient numbers of plateau harmonics. For the five harmonics in the example, $\Delta t = 500$ as.

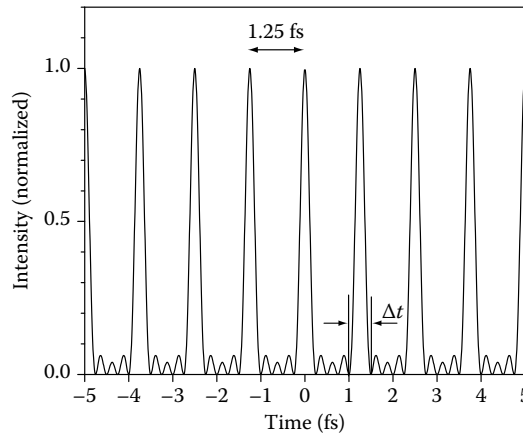


Figure 1.31 A train of attosecond pulses corresponding to five plateau harmonics.

The relationship between the plateau harmonics and the attosecond pulses train is very similar to that between a set of multiple slits and their interference pattern. In the above analysis, we assume that the phases of all the plateau harmonics are the same. The measurements of the phases were a major challenge. The first result was obtained in 2001, 13 years after the discovery of the high harmonic generation. The predicted attosecond pulse train structure was also confirmed at that time. The measurement technique is explained in detail in Chapter 7.

Equation 1.69 can also be written as

$$\Delta t \cdot \left(\frac{q_H - q_L}{2} + 1 \right) \hbar \omega_1 = 2\pi \hbar, \quad (1.70)$$

which is another manifestation of the uncertainty principle.

1.3.2.2 Three-Step Model

A semi-classical model was proposed in 1993, which revealed the mechanism of high-order harmonic generation, which is discussed in detail in Chapter 4. Consider the interaction of an atom, namely a hydrogen atom, with a linearly polarized laser field. The field-free Coulomb potential is shown in Figure 1.32. As the intensity reaches the level of 10^{14} W/cm^2 , the field near the peak of the each oscillation is comparable to the atomic Coulomb field. The superposition of the laser field and the Coulomb field transforms the potential well that binds the electron into a potential barrier, as depicted in Figure 1.33.

Consequently, the electron in the ground state tunnels through the barrier (the first step). The freed electron moves in the laser field like a classical particle, and its trajectory can be calculated using Newton's second law. In one laser cycle, the electron first moves away from the nucleus, after which it is driven back when the electric force of the laser field changes direction. During the return journey, the electron can acquire

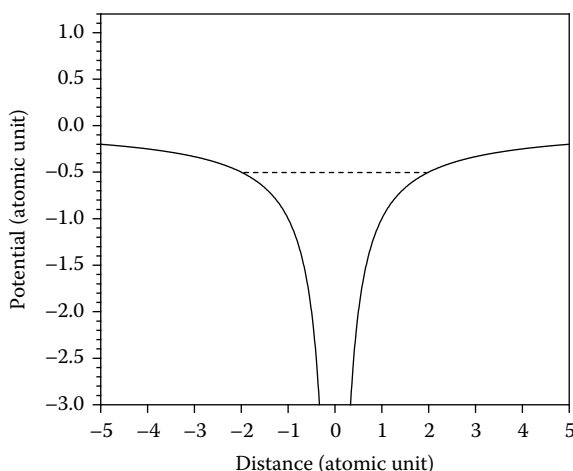


Figure 1.32 The potential energy curve of a hydrogen atom. The dashed line indicates the ground state.

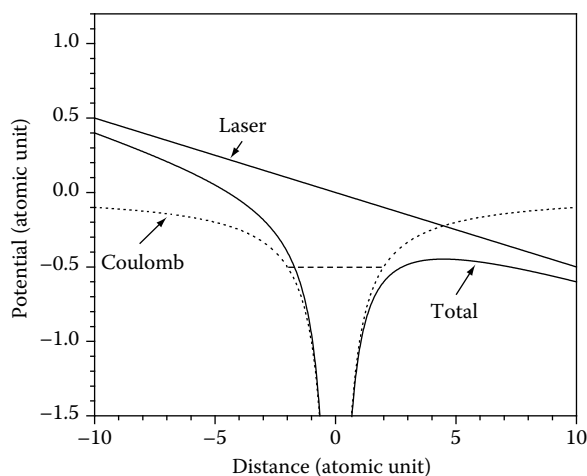


Figure 1.33 Formation of the Coulomb barrier. The laser field in the figure is 0.05 a.u.

kinetic energy up to hundreds of electron volts (the second step). Finally, the electron recombines with the parent ion with the emission of a photon (the third step).

When all electrons released near one peak of a laser cycle are considered, the emitted photons form an attosecond pulse. Since there are two field maxima in one laser cycle, two as pulses are generated, which explains the repetition rate shown in Figure 1.31. For a laser pulse that contains many cycles, an attosecond pulse train is produced. This is the origin of the primary peaks in Figure 1.31. The pulse train corresponds to discrete harmonic peaks in the frequency domain, as shown in Figure 1.30. In other words, high harmonic generation and the attosecond pulse train are two manifestations of the same nonperturbative interaction.

It is interesting to compare the pulse shortening by the perturbative method to the nonperturbative method. In the former case, we use the nonlinearity of the chosen order. The higher the nonlinearity, the more effective the shortening. For high harmonic generation, the intensity of the plateau harmonic order q does not depend on the intensity, as in the perturbative case, I^q . In the nonperturbative regime, the pulse is shorter than one laser cycle because the whole three-step process occurs in sub-laser-cycle timescales. Quantitative descriptions of the semi-classical model are given in Chapter 4. The full-quantum model is laid out in Chapter 5.

1.3.2.3 Single Isolated Attosecond Pulses

The attosecond pulse train corresponding to high-order harmonics is useful for some applications. In general, however, single isolated attosecond pulses are required for performing pump–probe experiments with well-defined time to start and to observe a process. Such pulses can be generated by suppressing all the pulses in the train except one, which can be accomplished by using single-cycle driving lasers or pulse extraction switches with a subcycle opening time. Various gating techniques are

discussed in Chapter 8. By 2008, the shortest single isolated pulses, which were generated from neon gas by using 3.3 fs driving lasers centered at 720 nm, were 80 as and contained 0.5 nJ of energy. Their spectrum was centered at 80 eV.

In many cases, a strong attosecond pulse is accompanied by weak pre- or post-pulses. This is because it is difficult to completely turn off attosecond emission using the gating methods developed so far. The satellite pulses are the leakages of the pulses in the train shown in [Figure 1.31](#). The definition of single isolated attosecond pulses is yet to be standardized. In other words, we need to define a maximum intensity ratio between the satellite pulse and the main pulse, η . When the ratio is less than η , the main pulse can be considered as a single isolated one. Here we suggest choosing $\eta = 0.1$, which corresponds to the case that the satellite pulse is one order of magnitude weaker than the main pulse.

In the spectrum domain, the power spectrum of a single Gaussian pulse without any satellite pulses is also a Gaussian function. Such a spectrum is called an XUV continuum. There is no modulation in the spectrum. The modulation depth of the power spectrum corresponding to a main pulse with satellite pulses is related to the number of satellite pulses and their intensity, as in the case of double-slit or multi-slit interference.

Consider the case that a Gaussian high-intensity pulse is accompanied by a weak pulse. Assume that they have same pulse duration. The electric field in the frequency domain is

$$\tilde{E}(\omega) = U_m e^{-2 \ln 2 \frac{(\omega - \omega_0)^2}{\Delta\omega^2}} + U_s e^{-2 \ln 2 \frac{(\omega - \omega_0)^2}{\Delta\omega^2} + i(\omega - \omega_0)T_d}, \quad (1.71)$$

where

U_m and U_s are the peak values of field of the main and satellite pulses, respectively

T_d is the delay between U_m and U_s

ω_0 and $\Delta\omega$ are the central frequency and the bandwidth, respectively

The power spectrum can be expressed as

$$\begin{aligned} I(\omega) &= |\tilde{E}(\omega)|^2 \\ &= \{I_m + I_s + 2\sqrt{I_m I_s} \cos[(\omega - \omega_0)T_d]\} e^{-4 \ln 2 \frac{(\omega - \omega_0)^2}{\Delta\omega^2}}, \end{aligned} \quad (1.72)$$

where $I_m = U_m^2$ and $I_s = U_s^2$.

The contrast of the pulses is

$$\eta = \frac{I_s}{I_m}. \quad (1.73)$$

The modulation depth in the power spectrum is

$$\frac{4\sqrt{I_m I_s}}{I_m + I_s + 2\sqrt{I_m I_s}} = \frac{4\sqrt{\eta}}{(1 + \sqrt{\eta})^2}. \quad (1.74)$$

For $\eta = 0.1$, the modulation depth is 73%, which is quite large. When the contrast $\eta = 0.001$, the depth is approximately 12%, which is still obvious in the spectrum.

1.3.3 Measurement of Attosecond Pulse Duration

The streak camera principle described in Section 1.1.2 has been used to measure the duration of single isolated attosecond pulses. When a beam of attosecond pulses is focused on an atomic gas target that serves as the photocathode, the bound electrons in the atoms can be ejected to vacuum by the XUV photons. The streak camera discussed in Section 1.1.2 converts temporal information to spatial information, because it is much easier to measure spatial information. An attosecond streak camera converts temporal information into momentum information. The momentum of electrons can be measured using the well-developed TOF spectrometer techniques. Laser fields are used to sweep the photoelectrons in the momentum space.

1.3.3.1 Response of the Gas Photocathode

Noble gases are commonly used as the detection gas because of the large ionization potential. The gas target is located in a high vacuum that is shared with the electron TOF spectrometer. Due to the low photon flux of the attosecond pulses, the challenge is to have a high length-pressure product to absorb enough photons. The length of the gas should be smaller than the focal spot of the streaking laser, typically $<50\text{ }\mu\text{m}$; otherwise the gas atoms outside of the laser beam contribute to the background pressure. Figure 1.34 illustrates the transmission of a $50\text{-}\mu\text{m}$ long neon gas target with 1 torr pressure. The absorption at 65 eV is 0.1%, and the detection efficiency is thus very low.

1.3.3.2 Momentum Streaking

As in conventional streak cameras, the photoelectron pulse is considered a replica of the attosecond XUV pulse. The momentum distribution of the electron is determined by Einstein's photoemission law,

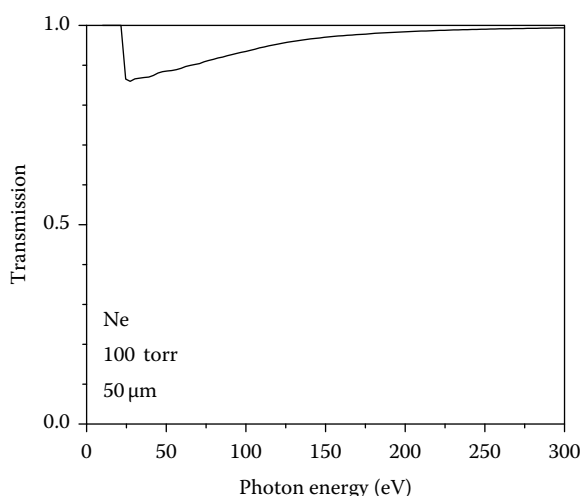


Figure 1.34 Transmission of a $50\text{ }\mu\text{m}$, 1 torr neon gas target. (Calculated using Advanced Light Source, Lawrence Berkeley National Lab, http://henke.lbl.gov/optical_constants/gastrn2.html, accessed April 12, 2010.)

$$p_0(\omega_X) = \sqrt{2m_e(\hbar\omega_X - I_p)}, \quad (1.75)$$

where

ω_X is the frequency of Fourier component of the attosecond pulse

I_p is the binding energy of the atom

If an electron is released to the laser field at time t_0 , then its equation of motion is

$$\frac{d\vec{p}}{dt} = -e\vec{\mathcal{E}}_L(t). \quad (1.76)$$

The expression can be simplified by using the magnetic vector potential, $\vec{\mathcal{A}}$, which is defined as

$$\vec{\mathcal{B}}_L = \nabla \times \vec{\mathcal{A}}_L, \quad (1.77)$$

where $\vec{\mathcal{B}}_L$ is the magnetic induction. From Faraday's induction law,

$$\nabla \times \vec{\mathcal{E}}_L = -\frac{\partial \vec{\mathcal{B}}_L}{\partial t} = -\frac{\partial(\nabla \times \vec{\mathcal{A}}_L)}{\partial t} = -\nabla \times \left(\frac{\partial \vec{\mathcal{A}}_L}{\partial t} \right), \quad (1.78)$$

we have

$$\vec{\mathcal{E}}_L = -\frac{\partial \vec{\mathcal{A}}_L}{\partial t}. \quad (1.79)$$

Replacing the electric field in Equation 1.79 by the one in Equation 1.76 leads to

$$\frac{d\vec{p}}{dt} = e \frac{d\vec{\mathcal{A}}_L}{dt}. \quad (1.80)$$

Thus,

$$\vec{p}(t) - \vec{p}_0 = e[\vec{\mathcal{A}}_L(t) - \vec{\mathcal{A}}_L(t_0)]. \quad (1.81)$$

In experiments, the momentum of the electron is measured after the laser pulses pass through the gas target, i.e., $\vec{\mathcal{A}}_L(t = \infty) = 0$. Thus, the measured momentum is

$$\vec{p}(t_m) - \vec{p}_0 = -e\vec{\mathcal{A}}_L(t_0). \quad (1.82)$$

1.3.3.3 Time to Momentum Conversion

We assume that the laser is monochromatic and propagates in the x direction. For simplicity, we further assume that the laser is linearly polarized along the z direction. The electric field of the laser can then be written as $\vec{\mathcal{E}}_L(t) = E_0 \cos(\omega_0 t) \hat{z} = E_0 \cos(2\pi[t/T_0]) \hat{z}$. In this case,

$$\vec{p}(t_m) - \vec{p}_0 = \frac{e}{\omega_0} E_0 \hat{z} \sin\left(2\pi \frac{t_0}{T_0}\right). \quad (1.83)$$

The range of t_0 variation is the attosecond pulse duration, τ_x .

For τ_x much shorter than the laser period T_0 ,

$$\vec{p}(t_m) - \vec{p}_0 \approx eE_0 t_0 \hat{z}. \quad (1.84)$$

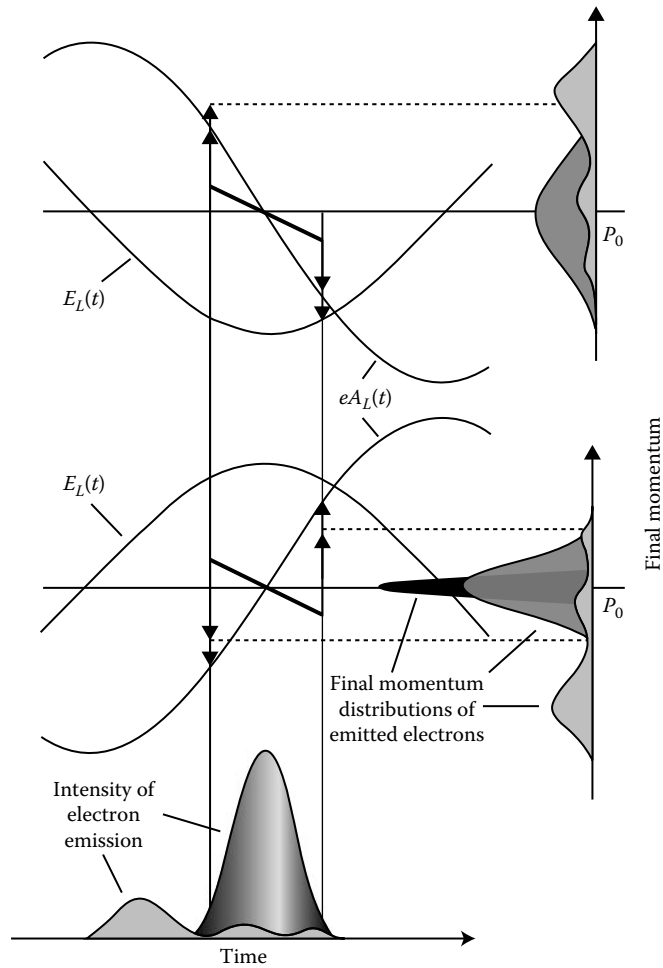


Figure 1.35 The principle of attosecond streaking. (From Kienberger, R. et al., *Nature*, 427, 817, 2004. With permission.)

For the same initial momentum, this equation establishes the mapping of electron release time, t_0 , to the measured momentum, $\vec{p}(t_m)$, as shown in Figure 1.35. Therefore, the electrons in different temporal portions of the attosecond pulse will have different momentum shifts. The peak of the attosecond pulse should be located at the zero-crossing point of the vector potential for maximum shifting. The attosecond pulse must be shorter than half of an optical cycle to avoid the momentum degeneracy between two different releasing times.

For simplicity, we only consider the electrons that are emitted in the laser polarization direction, i.e., in the positive z direction. For a transform-limited XUV pulse with duration τ_x , the width of the momentum distribution after the streaking is

$$\Delta p = \sqrt{\Delta p_0^2 + \left(\frac{dp(t_m)}{dt_0} \tau_x \right)^2}, \quad (1.85)$$

where Δp_0 is the width of the momentum distribution without streaking. The streaking speed is

$$\frac{dp(t_m)}{dt_0} = eE_0. \quad (1.86)$$

Once Δp and Δp_0 are measured at a given streaking speed, Equation 1.86 can be used to determine the attosecond pulse duration.

1.3.3.4 Time Resolution

Suppose the time resolution of a detector is Δt . For two identical pulses of an infinite small duration separated by Δt , the detector will display a trace as shown by the solid line in Figure 1.36, which is a superposition of two pulses broadened by the detector. The time resolution of the detector is defined as the minimum separation between the two, such that the modulation depth of the solid line is $8/\pi^2 \approx 81\%$. For such a modulation depth, we say that the two pulses can still be resolved.

For a transform-limited pulse with a Gaussian shape, $\tau_x \Delta \varepsilon_x / h = 0.441$. Here, τ_x and $\Delta \varepsilon_x$ are the FWHM of the pulse envelope and the power spectrum, respectively. In atomic units, $\tau_x \Delta \varepsilon_x = 0.441 \times 2\pi$ and $\Delta \varepsilon_x = p_0 \Delta p_0$, while the momentum bandwidth is $\Delta p_0 = 0.882\pi / \tau_x p_0$. The Rayleigh criterion requires that

$$\Delta p_2(\Delta t) = \sqrt{\Delta p_0^2 + \Delta p(\tau_x)^2}, \quad (1.87)$$

where

$\Delta p_2(\Delta t)$ is the separation of the two peaks in momentum space when the pulses are separated in time by Δt

$\Delta p(\tau_x)$ is the broadening of each peak due to the streaking

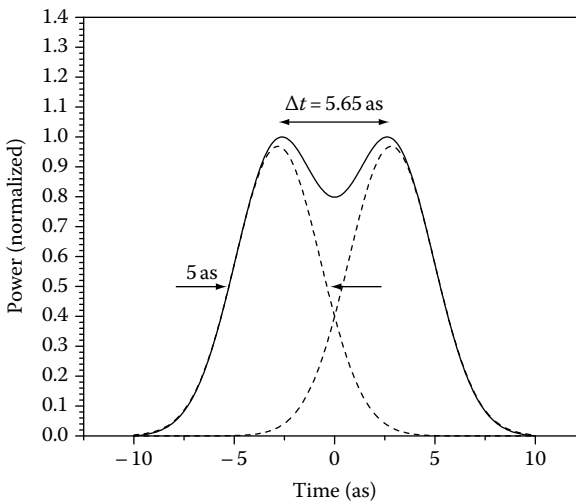


Figure 1.36 Time resolution defined by the Rayleigh criterion. We assume that a δ function pulse is broadened by the detector to a Gaussian shape with 5 as FWHM.

In terms of streaking speed,

$$eE_0\Delta t = \sqrt{\Delta p_0^2 + (eE_0\tau_x)^2}. \quad (1.88)$$

We can set $\Delta t = 2\tau_x$, which is the case where the contribution of Δp_0 equals $\Delta p(\tau_x)$. This leads to

$$\Delta t = \frac{\Delta p_0}{eE_0}. \quad (1.89)$$

As an example, the duration of the XUV pulse is $\tau_x = 5$ a.u. = 121 as, and it is centered around the 23rd order harmonic of Ti:Sapphire laser. When neon is used as the detection gas, $p_0 = 1$ a.u., $\Delta p_0 = 0.55$ a.u. To achieve $\Delta t = 10$ a.u., E_0 should be 0.055 a.u., which corresponds to a peak intensity of 1×10^{14} W/cm².

One can also define the time resolution in a less stringent way. The shift of the momentum should be at least 20% of the initial momentum bandwidth, to distinguish the laser streaking effect from other minor momentum shifting mechanisms, such as the intensity fluctuation in the XUV pulse generation process. In this case, it is 0.11 a.u. In atomic units, the maximum momentum shift by the laser field is $-A_0(0) = (T_0/2\pi)E_0(0)$. The required laser field amplitude for 0.11 a.u. of momentum shift is 6.23×10^{-3} a.u., which corresponds to a peak intensity of 1.38×10^{12} W/cm² and a vector potential of 13.7 fs MV/cm. The intensity can be achieved by focusing 0.5 μ J, 20 fs pulses to a 30 μ m radius spot. Under this intensity, the above-threshold ionization of the second gas target by the IR laser is almost zero.

A more precise and powerful measurement method based on streaking is discussed in Chapters 7 and 8. It can determine the pulse shape and phase of either an attosecond pulse train or single isolated pulses.

1.4 Properties of Attosecond XUV Pulses

Pulse duration is the most important specification of attosecond pulses. Other properties are also important for various applications.

1.4.1 Pulse Energy

At the present time, the conversion efficiency from laser to attosecond pulse is still very low, on the order of 10^{-6} or less. It is limited by the phase matching in the gas target, which will be discussed in Chapter 6. The attosecond pulse energy is also limited by the NIR laser that drives the nonperturbative process, which, in many cases, is only a few millijoules. The energy of attosecond pulses is on the order of nJ or less. It is highly desirable to generate high-power attosecond pulses for studying nonlinear optics in the XUV and x-ray wavelength range.

The energy of an attosecond pulse can be measured with a Si photodiode and an oscilloscope. The band gap of Si is 1.1 eV, and one XUV photon can therefore produce more than one photoelectron. The number of electrons generated from one XUV photon is defined as the quantum

efficiency of the detector, which can be calibrated at Synchrotron facilities. The electron will charge up an internal capacitor (~ 10 nF). The discharge of the capacitor through the resistor produces a voltage that can be measured by an oscilloscope. From the peak voltage value, the discharge time, and the quantum efficiency, one can determine the number of photons in the attosecond pulse.

1.4.2 Divergence Angle

Unlike in visible light topics, lenses are rarely used in the XUV and soft x-ray range. It is difficult to find materials that have indices of refraction significantly different from 1 and with small absorption. It is also difficult to use mirrors near normal incidence because of the low reflectivity. Figure 1.37 shows the reflection curve of an iridium mirror at normal incidence. The reflectance is approximately 1% in the 40–80 eV range. It is almost zero for photon energy higher than 150 eV. On the other hand, for attosecond second pulse below 30 eV, the reflectance is reasonably high.

Mirrors with glancing (or grazing) incidence are commonly used for XUV beams above 30 eV. The reflection of a gold mirror at 5° glazing incidence angle is shown in Figure 1.38. To avoid using large mirrors and to keep aberrations small, the divergence angle of the attosecond beam should also be small. It was found that the divergence angle of the XUV beam is smaller than that of the driving laser, and can be calculated using the theory described in Chapter 6.

1.4.2.1 XUV Mirrors at Glancing Incidences

The enhancement of reflection at glancing incidence is related to the “total internal reflection” subject in optics. Line as shown in Figure 1.22, the index of refraction of Al and other mirror material is less than one in a

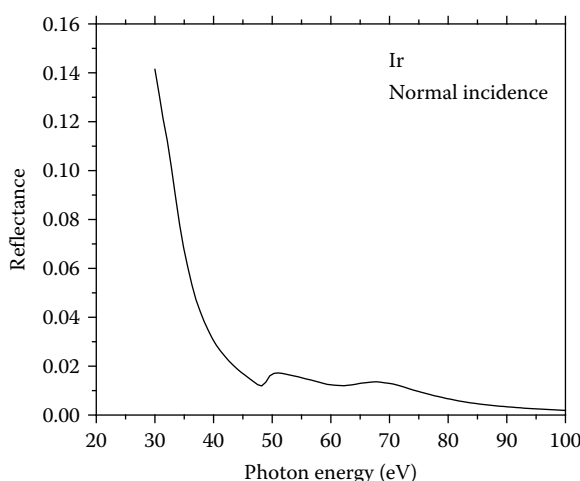


Figure 1.37 The reflectance of an Ir mirror at normal incidence. (Calculated using Mirror Reflectivity, Lawrence Berkeley National Lab, http://henke.lbl.gov/optical_constants/mirror2.html, accessed April 12, 2010.)

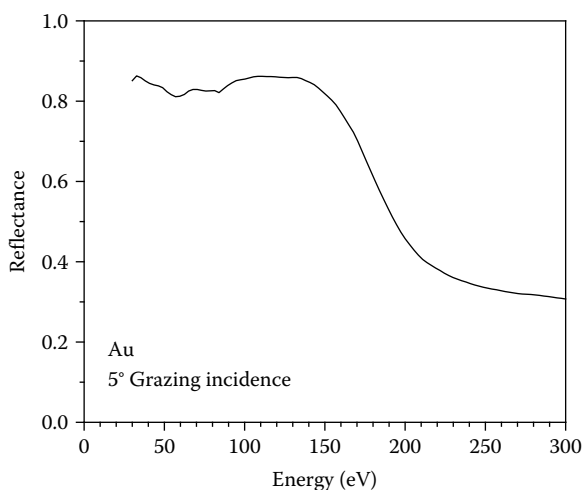


Figure 1.38 The reflectance of an Au mirror at a 5° glancing incidence. (Calculated using Mirror Reflectivity, Lawrence Berkeley National Lab, http://henke.lbl.gov/optical_constants/mirror2.html, accessed April 12, 2010.)

broad XUV wavelength range, which means that air is optically denser than metal! Consequently, total internal reflection occurs when the XUV beam is incident on the mirror surface. As an example, for $n_R = 0.999$, the total reflection angle is

$$\theta_c = \sin^{-1}(n_R) = 87.4^\circ, \quad (1.90)$$

which corresponds to a glazing incident angle of 2.6° .

1.4.2.2 Multilayer XUV Mirrors

A multilayer XUV mirror is an option for experiments where normal incidence is required. Like the dielectric mirrors for visible light, multilayer mirrors enhance the reflection by constructive interference between the reflections of each layer. It was found that Mo/Si stacks provide high reflection at 13.5 nm, as shown in Figure 1.39. There are 40 periods in the coating, and the peak reflectance reaches 70%, which is considered to be excellent for XUV light. In comparison, it is quite common to achieve >99% reflection with multilayer mirrors in the visible light range.

The bandwidth of the standard Mo/Si mirror is only ~ 4.2 eV, which supports a 400 as pulse. To reflect even shorter attosecond pulses, one can reduce the periods of layers. As an example, when 2 periods are used, the bandwidth is 10 times broader (as shown in Figure 1.40), which could be used with pulses shorter than 50 as. However, the reflectance is less than 3% with such few layers.

1.4.3 Challenges and Opportunities in Attosecond Optics

A typical attosecond generation setup is shown in Figure 1.41. It consists of a high repetition rate femtosecond Ti:Sapphire laser system, a vacuum

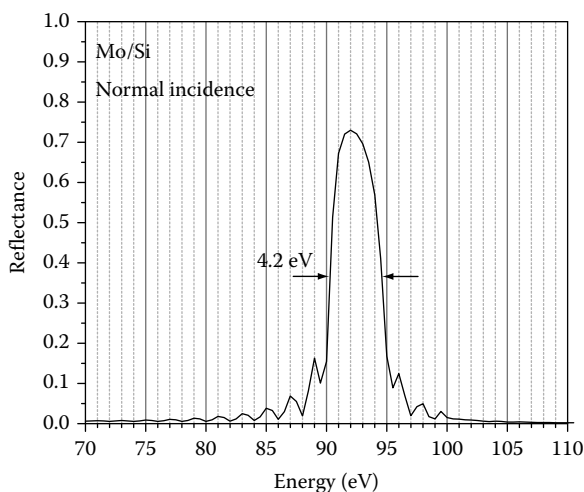


Figure 1.39 The reflectance of a standard Mo/Si mirror. (Calculated using Multilayer Reflectivity, Lawrence Berkeley National Lab, http://henke.lbl.gov/optical_constants/multi2.html, accessed April 12, 2010.)

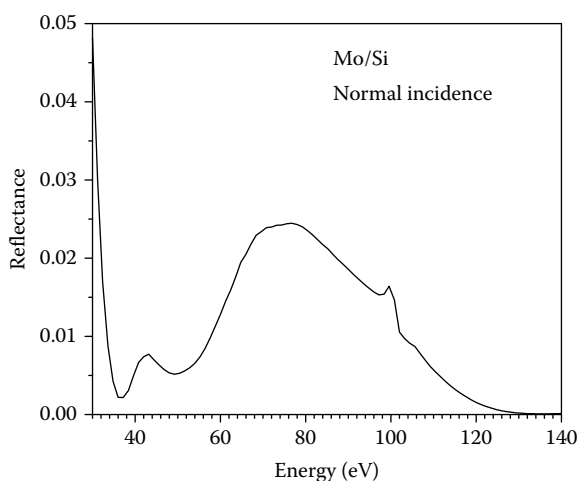


Figure 1.40 The reflectance of two periods of Mo/Si. (Calculated using Multilayer Reflectivity website, Lawrence Berkeley National Lab, http://henke.lbl.gov/optical_constants/multi2.html, accessed April 12, 2010.)

chamber where the gas target is located, and an XUV attosecond streak camera that characterizes the pulses in the time domain. The attosecond pulses are XUV or soft x-ray light that cannot propagate in air because of high absorption. The gas density in the laser interaction region is on the order of 10^{17} to 10^{18} atoms/cm³. The interaction length is typically a few mm for gas cells or gas jets.

Measurement of the optical pulse duration requires a temporal gate. For femtosecond lasers, nonlinear optics phenomena such as second harmonic generation can serve as the gating, which is the foundation of

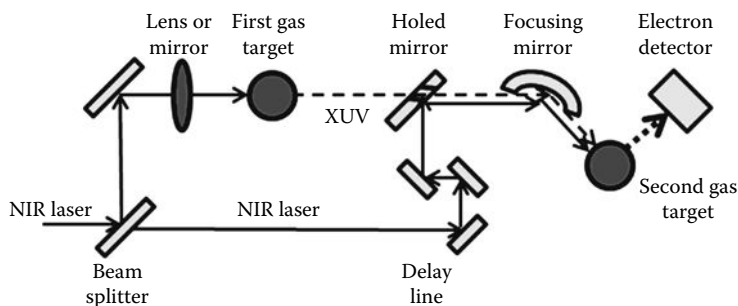


Figure 1.41 Attosecond XUV pulse generation setup.

widely implemented auto-correlation and the frequency resolved optical gating (FROG) techniques, as discussed in Chapter 2. The intensity of the attosecond pulses is not yet high enough to generate second harmonic light. Most of the methods for determining the width of the attosecond pulses require the measurements of photoelectrons or ions. The XUV beam is focused to a second gas target to generate the photoelectrons/ions. The charge particles are then detected by a TOF spectrometer. A second beam, either an XUV or an intense laser beam, is also focused to the same target, overlapping spatially and temporally with the first beam. The interaction of the two pulses in the gas serves as the temporal gate. The attosecond XUV pulses are generated in the first gas target and are measured in the second gas target. Similar apparatus has been used for studying electron dynamics in atoms, which will be discussed in Chapter 9.

An experimental setup for generating and characterizing isolated attosecond pulses in the author's lab is shown in Figure 1.42. Double optical gating (DOG) optics is for transforming a linearly polarized laser pulse into a sub-laser-cycle gating pulse, to select a single pulse from a train.

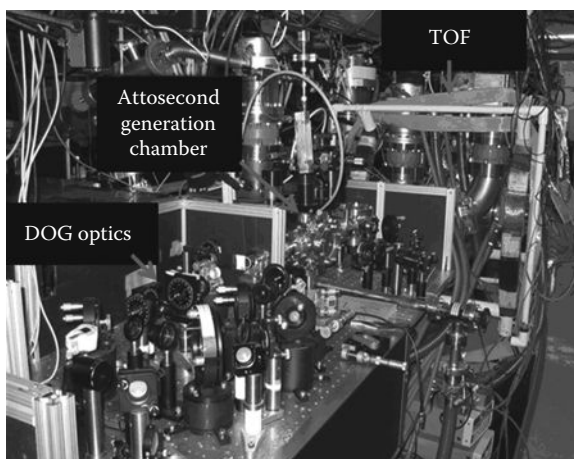


Figure 1.42 Experimental setup for an attosecond XUV pulse generation.

The TOF electron spectrometer is the key part of the attosecond streak camera. The details of the setup are explained in Chapters 7 and 8.

There are many differences between attosecond XUV optics and ultrafast optics in the visible or IR range.

1. Attosecond optics depends on high field physics. It needs ultrafast lasers that can deliver intensities higher than 10^{13} W/cm².
2. Attosecond optics relies on processes that occur in a fraction of a laser cycle, which requires the locking of the carrier-envelope phase. The precision of the time delay between the XUV pulse and the NIR laser pulse in the streak camera setup must be kept at a subfemtosecond level, which requires very good mechanical stability (such as in optical interferometric experiments), as in the setup shown in [Figure 1.42](#).
3. Attosecond pulses are XUV light. High vacuums are always needed for XUV detectors.
4. Expensive XUV optics are necessary. XUV mirrors and gratings work in glancing incidence. Special care needs to be taken to reduce aberrations.

Significant progress has been made in attosecond optics since the discovery of high harmonic generation at the end of 1980s, and the discipline continues to advance at an extremely fast pace. Two major issues that still need to be solved are: (1) achieving a photon flux high enough for conducting nonlinear optics and (2) generating pulses as short as 24 as.

Problems

- 1.1 Another pulse shape used in ultrafast optics is the square function pulse. Derive the relationship between energy, peak power, and pulse width for such pulses. Compare the results with Equation 1.2.
- 1.2 Find out the response time of the fastest photodiode and the oscilloscope on the Internet. What factors set the limits of their response time?
- 1.3 The power of lasers is always compared to that of the Hoover Dam on the border between Arizona and Nevada. What is the power of the Hoover Dam? Is it the peak power or the average power?
- 1.4 A student measures the average power of a laser system with a powermeter. The laser produces 3000 identical pulses per second. The powermeter shows 1 W.
 - a. What is the energy per pulse?
 - b. Suppose the pulse shape is Gaussian and the FWHM is 20 fs. What is the peak power of each pulse?
- 1.5 What is the definition of an electric field? What is a direct way to measure the electric field around a charged object at rest?
- 1.6 For a “flat-hat” laser beam, find out the relationship between the intensity, power, and radius. Compare with Equation 1.7.
- 1.7 Calculate the electric field amplitude of light 1 m away from a 100 W light bulb. Can it be measured directly? If the answer is yes, explain the measurement method.

- 1.8 Suppose the laser beam in Problem 1.7 is focused down to $30\text{ }\mu\text{m}$. What is the peak intensity?
- 1.9 Which atom has the smallest ionization potential? At what laser intensity is the laser field amplitude comparable to the Coulomb field experienced by the outermost electron in this atom?
- 1.10 What is the total power of sunlight on the Earth's surface? If all the power is focused by a huge lens to a grain of sand, what is the intensity on the grain? What will happen to the sand?
- 1.11 What is the speed of the fastest 100 meter runner? What time resolution is needed to take a clear picture of the runner during a race?
- 1.12 Derive Equation 1.12 using Bohr's model of the hydrogen atom. Calculate the orbiting time of the first excited state of hydrogen.
- 1.13 Derive Equation 1.20.
- 1.14 Plot the temporal phase and the instantaneous frequency of the two pulses shown in Figure 1.13.
- 1.15 Find the time-bandwidth expression for a square pulse.
- 1.16 Calculate the gas density when the vacuum is 1 torr ($\sim 1\text{ mBar}$).
- 1.17 Plot and compare the transmission of the five noble gases, Xe, Kr, Ar, Ne, and He, when the pressure is 100 torr and the length is 10 mm in the 10–300 eV range.
- 1.18 Plot and compare the transmission of the five types of filters, Al, Ag, Au, Zr, and Sn, in the 10–500 eV range. Assume the thickness is 200 nm.
- 1.19 Plot and compare the transmission of the three types of filters, Mg, Al, and Si, in the 10–100 eV range. Assume the thickness is 200 nm. What are the similarities and differences? Provide an explanation for your observations.
- 1.20 Plot the f_1 , n_R , and t_g for a 300 nm Zr filter in the 50–100 eV range.
- 1.21 Find the GVD of Sn at 130 eV.
- 1.22 How many plateau harmonics are needed to compose an attosecond pulse train in which each harmonic is 25 as wide? Assume the laser wavelength is $1.5\text{ }\mu\text{m}$.
- 1.23 Draw the potential energy diagram of molecular ion H_2^+ , indicating the ground state. When a laser field with 0.1 a.u. interacts with it, draw the potential barrier formed by the superposition between the external field and the Coulomb field. What will happen to the electron in the ground state?
- 1.24 Streaking can also be expressed in terms of kinetic energy instead of momentum. Rewrite Equation 1.59 using kinetic energy.
- 1.25 A 25 as pulse is centered at 155 eV. In order to measure the pulse duration with the streak camera, what intensity is required? The center wavelength of the laser is 750 nm.
- 1.26 Compare the reflectance of a Ti mirror in the 30–200 eV range at normal incidence to the reflectance, at 5° glancing incidence.
- 1.27 Plot the total reflection angle for Al as a function of wavelength.
- 1.28 Using Equation 1.16, prove that the linear dependence of the temporal phase is equivalent to a shift of center frequency.
- 1.29 Derive the equation $\hbar\omega_0[\text{eV}] = 1240/\lambda_0[\text{nm}]$.
- 1.30 Derive the equation $\text{GDD}[\text{fs}^2] = \text{GDD}[\text{as/eV}]/1516$.

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Femtosecond Driving Lasers

2.1 Introduction

There are several basic requirements for femtosecond driving lasers for the generation of attosecond pulses. First, the intensity at the focus must be high enough. The intensity on the gas target should be on the order of 10^{14} W/cm², which is a fraction of an atomic unit of intensity (3.55×10^{16} W/cm²). Also, the corresponding pulse energy should be 100 μ J or higher to achieve such an intensity without tight focusing.

The second requirement is that the laser pulse duration must be short enough for generating single, isolated attosecond pulses. In this case, the ionization of the target atoms by the laser field before the cycle where the attosecond pulse is generated must not completely deplete the ground state population. Depending on the generation scheme, acceptable laser pulses range from 3 to 30 fs, as is discussed in detail in Chapter 8.

A third requirement is that the carrier-envelope phase needs to be stabilized for generating isolated attosecond pulses. Since single attosecond pulses are generated in a fraction of a laser cycle, a shift in the carrier-envelope phase results in shot-to-shot variations of the attosecond pulses. More than one pulses with attosecond duration can be generated per laser shot if the carrier-envelope phase is not set correctly.

Finally, the repetition rate of the laser should be high, on the order of kilohertz. Many attosecond characterization and application schemes rely on photoelectron measurements. There is an upper limit on the number of electrons per shot in order to avoid the space-charge effect. Thus, the signal count rate is primarily determined by the repetition rate. The energy stability of high repetition rate lasers is better than those with low repetition rates.

Although pulses as short as 5 fs (centered at 800 nm) can be generated from Kerr lens mode-locked Ti:Sapphire oscillators, the pulse energy is only a few nanojoules. High-power laser pulses with durations of approximately 30 fs can be generated with chirped-pulse amplification (CPA). Pulses down to ~ 4 fs with energies of a few millijoules can be obtained by spectral broadening in hollow-core fibers filled with gases, followed by dispersion compensation using chirped mirrors or phase modulators. [Figure 2.1](#) breaks this down into a block diagram so that it is easier to visualize. We discuss the principles of such laser systems later in this chapter, while the carrier-envelope phase control is discussed in Chapter 3.

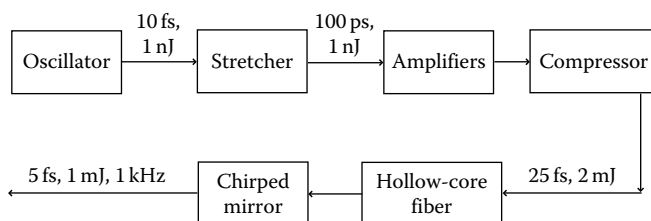


Figure 2.1 A block diagram of the laser system for attosecond pulse generation.

Ti:Sapphire is a commonly used gain medium for femtosecond lasers, primarily because of its broad gain bandwidth. Its center wavelength is approximately 800 nm, which corresponds to a 2.6 fs optical period. Femtosecond oscillators that use Ti:Sapphire as the gain medium can generate pulses with nanojoule-level energies, but direct amplification of the pulse to millijoule levels may cause damage to the laser crystal. Therefore, in a chirped amplifier, the pulses from the oscillator are stretched to hundreds of picoseconds, in order to lower the peak power. The pulses are then amplified in multipass or regenerative amplifiers, and the high-energy pulses are finally compressed to femtosecond duration. Most high-energy (>5 mJ) lasers use grating pairs to stretch and compress pulses. The schematics of a carrier-envelope phase stabilized Ti:Sapphire CPA laser system with a hollow-core fiber compressor in the author's lab is shown in Figure 2.2.

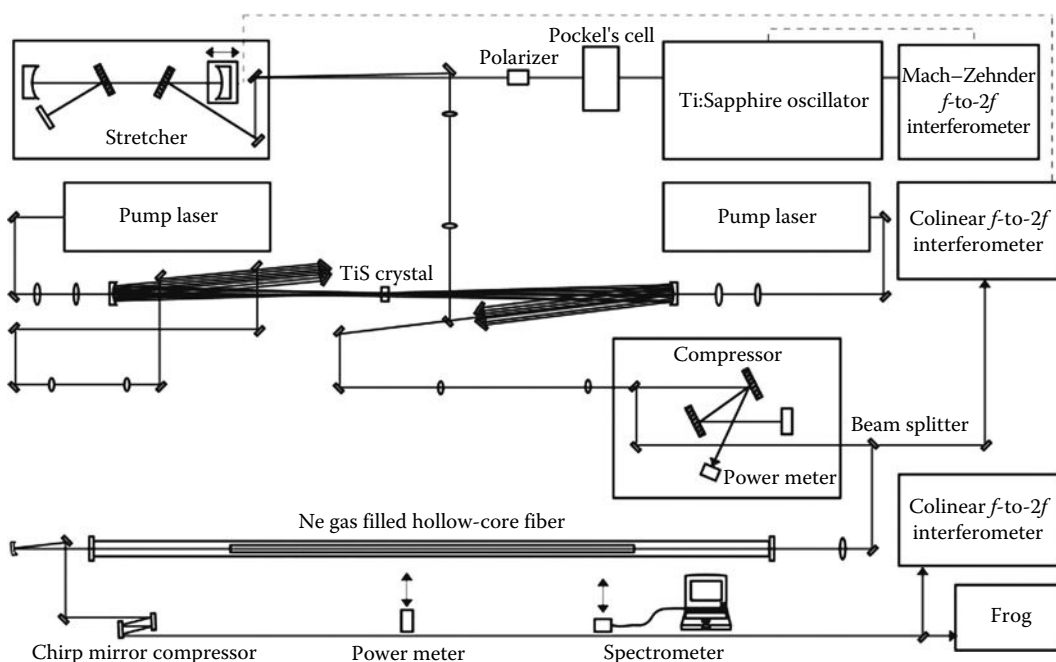


Figure 2.2 A schematic diagram of a laser system for attosecond pulse generation. (Reprinted with permission from H. Mashiko et al., Carrier-envelope phase stabilized 5.6 fs, 1.2 mJ pulses, *Appl. Phys. Lett.*, 90, 161114, 2007. Copyright 2007 American Institute of Physics.)

For the development and use of lasers, we need to know the basic properties of how the laser field changes in time and space. In principle, the variation of a laser field can be found by solving Maxwell's equations for the given initial and boundary conditions. In practice, that is mathematically too difficult. Some details of the mathematical approach are discussed in Chapter 6, where the propagation of a high harmonic field is analyzed.

It is difficult to find simple analytic equations that simultaneously describe both temporal and spatial variations of laser pulses during propagation. Thus, it is a common practice to apply the separation of variables technique and consider the spatial distribution of the field separately from the time dependence of the field. We will briefly discuss the spatial properties first, then the temporal properties. The in-depth analysis of laser propagation can be found in laser textbooks, such as Siegman's Lasers.

2.2 Laser Beam Propagation

When a laser beam propagates through a uniform isotropic medium, many of the beam parameters may change, such as the transverse beam size. We are interested in finding simple analytical expressions that allow us to calculate the beam size and other parameters at a given location.

For simplicity, we assume that the laser beam propagates along the z direction and is axially symmetric. In the cylindrical coordinate system, the electric field of a linearly polarized *monochromatic* beam can be described as

$$\varepsilon(r, z, t) = \tilde{E}(r, z)e^{i(\omega t - kz)}, \quad (2.1)$$

where

- ω and k are the angular frequency and the propagation constant, respectively
- r is the radius
- t is time

For such a beam, the time dependence is explicitly given by the exponential function $e^{i\omega t}$, and is thus known. $\tilde{E}(r, z)$ describes the transverse spatial profile (the r dependence) of the beam at a given position z , which is what we want to find. Important parameters such as beam size can be easily obtained from $\tilde{E}(r, z)$. A laser pulse is, of course, not monochromatic; however, when nonlinear effects are ignored, the theory discussed in this section can be applied to the spatial propagation of each frequency component of the laser pulse.

2.2.1 Gaussian Beam in Free Space

Under the paraxial approximation, we assume that the divergence angle of the beam is small (much smaller than 1 rad), such that it satisfies the paraxial wave equation

$$2ik \frac{\partial}{\partial z} \tilde{E}(r, z) = \frac{1}{r} \frac{\partial}{\partial r} \left[r \frac{\partial}{\partial r} \tilde{E}(r, z) \right]. \quad (2.2)$$

It indicates that the field changing rate along the z direction (the expression on the left) is caused by the field variation along the r direction (the expression on the right), which is essentially diffraction.

One of the solutions to this equation is a Gaussian function that is commonly used for describing laser beam propagation. A Gaussian function is an exponential function, which has nice mathematical properties that make the analysis simple. The complex amplitude of the Gaussian beam is

$$\tilde{E}(r,z) = \sqrt{\frac{2P}{c\epsilon_0}} \sqrt{\frac{2}{\pi}} \frac{1}{w(z)} e^{-\frac{r^2}{w^2(z)}} e^{-k\frac{r^2}{2R(z)}} e^{i\psi(z)}, \quad (2.3)$$

where

- c is the speed of light in vacuum
- ϵ_0 is the electric permittivity in vacuum
- P is the power carried by the beam
- $w(z)$ is the beam size (the $1/e^2$ radius)

The laser intensity is given by

$$I(r,z) = \frac{2}{\pi} \frac{P}{w^2(z)} e^{-\frac{2r^2}{w^2(z)}} = \frac{2}{\pi} \frac{P}{w_0^2} \frac{1}{1 + \left(\frac{z}{z_R}\right)^2} e^{-\frac{2r^2}{w^2(z)}}. \quad (2.4)$$

By introducing the Rayleigh length,

$$z_R = \frac{\pi w_0^2}{\lambda}, \quad (2.5)$$

where w_0 is the spot size at the beam waist located at $z=0$, we can describe a Gaussian beam at any position z , using the three parameters,

$$w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_R}\right)^2}, \quad (2.6)$$

$$R(z) = z + \frac{z_R^2}{z}, \quad (2.7)$$

$$\psi(z) = \tan^{-1} \left(\frac{z}{z_R} \right), \quad (2.8)$$

where $R(z)$ is the radius of curvature of the wavefront.

As an example, for a laser beam with a spot size of $30 \mu\text{m}$ and a laser wavelength of $0.8 \mu\text{m}$, which are the typical values used in attosecond pulse generations to reach the needed intensity, the Rayleigh range is 3.534 mm . The variation of beam size in the propagation is shown in [Figure 2.3](#). The intensity variation of the laser beam is also shown in the figure.

The confocal range is expressed by the parameter $b = 2z_R$, in which the beam size changes by a factor of $\sqrt{2}$ and the intensity varies by a factor of 2. The size of the gas target for attosecond generation is comparable to the Rayleigh range, so that the intensity in the whole target can reach the needed values.

$\psi(z)$ is called the Gouy phase shift, which is the phase difference between a Gaussian beam and a plane wave. This phase plays important roles in attosecond pulse generation, as is discussed in Chapter 6. The

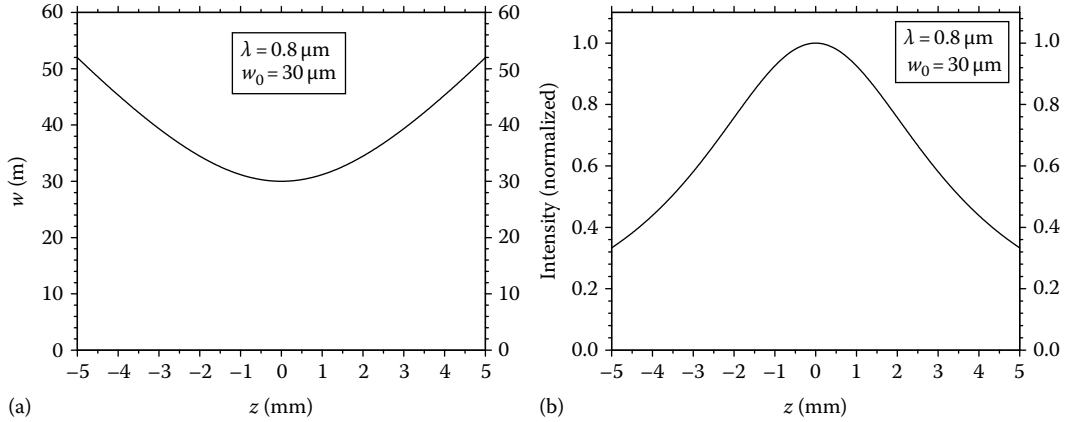


Figure 2.3 Variation of the (a) size and (b) intensity of a Gaussian beam near the beam waist.

phase value changes from $-\pi/2$ to $\pi/2$ for $z = -\infty$ to $+\infty$. The Gouy phase in the example is shown in Figure 2.4.

The divergence angle is defined as

$$\Theta = \lim_{z \rightarrow \infty} \frac{w(z)}{z} = \frac{w_0}{z_R} = \frac{\lambda}{\pi w_0}. \quad (2.9)$$

For the parameters in the example, the angle is 8.5 mrad, or 0.5° . The divergence angle of the attosecond beam is smaller than the driving laser, on the order of a few milliradians.

2.2.2 Gaussian Beam Focusing

The laser beam is always focused to a gas target to reach the required intensity for attosecond pulse generation. If a Gaussian beam is focused by

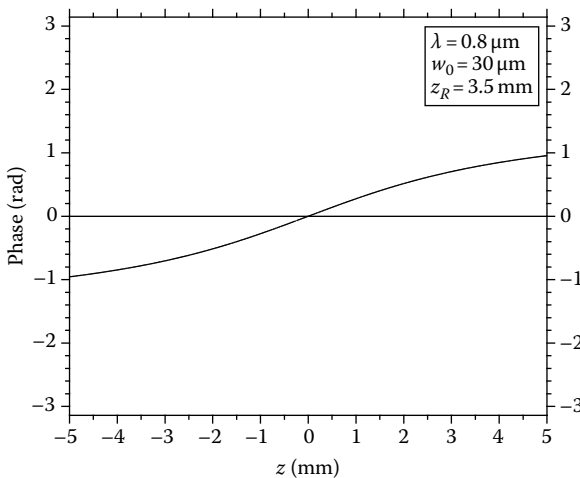


Figure 2.4 The Gouy phase in the confocal range.

an ideal lens or mirror, and if the focal length f is much longer than the confocal parameter around the focal spot, then the size of the focal spot is

$$w_f = \frac{f}{\pi w} \lambda. \quad (2.10)$$

Here, w is the radius of the beam on the lens surface, which is much smaller than the lens radius, to avoid clipping of the beam by the lens. As an example, for a laser beam with a diameter $D = \pi w = 10$ mm and a wavelength of $0.8 \mu\text{m}$, the focal spot size is $24 \mu\text{m}$ when the beam is focused by a $f = 300$ mm focal length lens. The Rayleigh range for such a system is $z_R = 2.3$ mm. These values are close to those used in the example given in [Section 2.2.1](#). The values of D and f are typical ones that have been used in attosecond pulse generation experiments when the laser energy is a few millijoules.

2.2.3 Aberration of Focusing Mirrors

Instead of lenses, mirrors are often used to focus short pulse laser beams because they do not have chromatic aberration. However, mirrors still suffer from geometrical aberrations, such as spherical aberration and astigmatism, which need to be taken into account when calculating the size and location of the focal spot.

When a collimated laser beam is focused by a spherical mirror with a radius of curvature R , the focal length f is a function of the height of a ray on the mirror, h , which is called a spherical aberration. Using the quantities defined in Figure 2.5, the focal length is

$$f(h) = \frac{R}{2} \left[2 - \frac{1}{\sqrt{1 - \left(\frac{h}{R}\right)^2}} \right]. \quad (2.11)$$

For $h/R \ll 1$, we have $f_G = R/2$, which is the focal length for paraxial rays. Paraxial optics is also called Gaussian optics. For off-axis rays, when $(h/R)^2 \ll 1$, the focal length is

$$f(h) \approx f_G \left[1 - \frac{1}{8} \left(\frac{h}{f_G} \right)^2 \right]. \quad (2.12)$$

For $h/(R/2) = 0.1$, the true focal length is different by $\sim 0.125\%$ of the paraxial focal length. As an example, for $f = 250$ mm and $h = 25$ mm, the

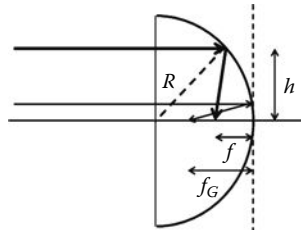


Figure 2.5 Focusing of light rays by a mirror.

error is 0.3 mm, which means the marginal rays are focused 0.3 mm away from where the central portion of the beam is focused.

In experimental setups, the reflected beam has to be deviated from the direction that the input beam comes from. Therefore, the off-axis aberrations such as astigmatism must also be considered. When the angle between the beam and the optical axis is θ , the focal length in the plane of incidence is

$$f_{\parallel}(\theta) = \frac{R}{2} \cos \theta. \quad (2.13)$$

For rays in the plane perpendicular to plane of incidence,

$$f_{\perp}(\theta) = \frac{R}{2} \frac{1}{\cos \theta}. \quad (2.14)$$

As an example, for $R/2 = 250$ mm and $\theta = 5^\circ$, $f_{\parallel} = 249$ mm, $f_{\perp} = 251$ mm. The difference (2 mm) is comparable to the Rayleigh range in a typical attosecond generation experiment, which affects the value of the peak intensity as well as the phase matching. If possible, the off-axis angle should be kept less than 2° , so that the difference is less than 0.1% of the focal length for paraxial rays, which is comparable to the spherical aberration.

Off-axis parabolic mirrors are also used in focusing lasers beams, which significantly reduces aberration. However, aspherical mirrors with good surface quality are much more expensive than spherical mirrors because they are difficult to make. It is also rather difficult to align an off-axis parabolic mirror.

2.2.4 Spherical Aberration of Focusing Lenses

When the laser pulse is long, which is the case when attosecond pulse trains are generated, lenses are also used. For simplicity, we can choose the plano-convex lens shown in Figure 2.6 as an example.

It can be shown by the third-order theory that

$$\frac{1}{f(h)} = \frac{1-n}{R} + h^2 \left[\frac{1}{2f} \left(\frac{1}{R} - \frac{1}{f} \right)^2 \right]. \quad (2.15)$$

When $h = 0$, we have the focal length of Gaussian optics,

$$\frac{1}{f_G} = \frac{1-n}{R}. \quad (2.16)$$

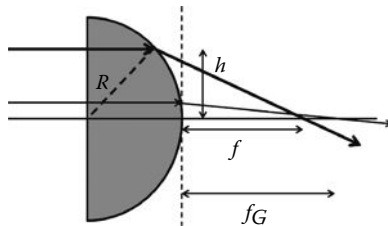


Figure 2.6 Focusing of light rays by a lens.

As an example, for $n = 1.5$, $f_G = 2R$. The third-order focal length can be rewritten as

$$\frac{1}{f(h)} \approx \frac{1}{f_G} + h^2 \left[\frac{1}{2f_G} \left(\frac{1}{R} - \frac{1-n}{R} \right)^2 \right] = \frac{1}{f_G} \left[1 + \left(\frac{h}{R} \right)^2 \frac{n^2}{2} \right]. \quad (2.17)$$

For $(h/R)^2 \ll 1$,

$$f(h) = f_G \left[1 - \left(\frac{h}{R} \right)^2 \frac{n^2}{2} \right] \approx f_G \left[1 - \left(\frac{h}{f_G} \right)^2 2n^2 \right]. \quad (2.18)$$

When comparing Equations 2.12 and 2.18, essentially comparing mirrors with lenses, we can see that the spherical aberration is $16n^2 \approx 36$ times larger for lenses than for mirrors! The aberration of a lens depends on the shape of the lens as well as the orientation. In order to minimize aberrations, the curved surface of a plano-convex lens should face the incident collimated laser beam.

2.2.5 Nonlinear Medium

2.2.5.1 Optical Kerr Effect

When the nonlinear response of the medium to the laser field is taken into account, the index of refraction of the medium is a function of the laser intensity. It increases from the field free value n_0 to a new value

$$n(I) = n_0 + n_2 I. \quad (2.19)$$

Here, n_2 is the nonlinear index of refraction. This phenomenon is called optical Kerr effect, which is related to the Kerr effect when a high voltage is applied across some optical materials (such as the highly toxic nitrobenzene). For most optical glasses, $n_2 \approx 3 \times 10^{-16} \text{ cm}^2/\text{W}$.

2.2.5.2 B Integral

The magnitude of the nonlinear phase shift caused by the optical Kerr effect is called the B integral,

$$B = \frac{2\pi}{\lambda} n_2 I L, \quad (2.20)$$

where L is the length of the medium. For many applications, $B \approx \pi$ radians is considered to be significant. For $\lambda \approx 1 \text{ }\mu\text{m}$ and $L = 1 \text{ cm}$, the intensity is on the order of 10^{12} W/cm^2 . This value can be easily attained by high-power femtosecond lasers.

2.2.5.3 Kerr Lens and Self Focusing

The intensity of a Gaussian beam at z is given by

$$I(r) = I_0 e^{-2\frac{r^2}{w^2}}. \quad (2.21)$$

The intensity distribution leads to a variation of the index of refraction in the transverse direction, as expressed by

$$n(r) = n_0 + n_2 I(r). \quad (2.22)$$

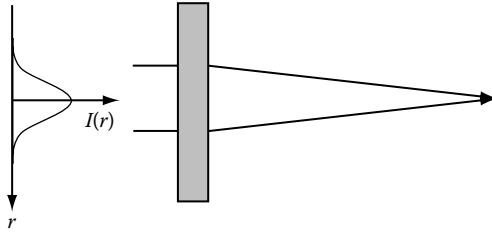


Figure 2.7 Self-focusing due to a Kerr lens.

When a laser beam propagates through a glass slab, the phase delay at the center of the beam is larger than at the edges. Consequently, the wavefront is curved, which is very similar to when a plane wave travels through a positive lens, as shown in Figure 2.7. Most femtosecond oscillators use the Kerr lens effect to lock the phase of the cavity modes, which is discussed in [Section 2.8](#).

The curvature of a wavefront for a Gaussian beam is described by the r -dependent nonlinear phase shift,

$$\phi_{nl}(r) = -\frac{2\pi}{\lambda} I_0 e^{-\frac{2r^2}{w^2}} n_2 L \approx -B \left[1 - 2 \left(\frac{r}{w} \right)^2 \right], \quad (2.23)$$

which is parabolic near the center of the beam. Here $B = (2\pi/\lambda) n_2 I_0 L$ is the nonlinear phase shift at the center of the beam.

When the laser peak power is higher than a critical power defined by

$$P_c = \pi(0.61)^2 \frac{\lambda^2}{8(n_0 n_2)}, \quad (2.24)$$

a collimated Gaussian beam will be focused to a small spot in the medium. For a model, a laser wavelength of $\lambda = 0.79 \mu\text{m}$ is chosen, and $n_0 = 1.76$ and $n_2 = 2.9 \times 10^{-16} \text{ cm}^2/\text{W}$ are the linear and nonlinear indices of refraction of the sapphire plate, respectively. The critical power, as determined by Equation 2.24, is then $P_c = 1.79 \text{ MW}$. It is important to know this value, because the material can be damaged due to the high intensity at the focal spot of the Kerr lens. Under some conditions, the self-focusing can be balanced by diffraction, which leads to the formation of single or multiple filaments in the medium, depending on the situation. The output power of the CPA lasers used is on the order of 0.1 TW , which is five orders of magnitude higher than the critical power of solids. Even a small fraction of the power can produce filaments, which is used in measuring the carrier-envelope phase of laser pulses.

2.2.5.4 Optical Damage

Straight forward amplification of femtosecond pulses from nanojoules to millijoules is difficult. In the amplifier, the laser intensity has to stay low, otherwise nonlinear effects such as optical Kerr effects become significant enough to distort the spatial and temporal properties of the pulse. Another issue associated with high power is the damage of the optics and gain materials. It is known that the damage threshold influence (J/cm^2)

increases with the laser pulse duration; thus, to avoid damage, amplifier picosecond pulses are preferred instead of femtosecond pulses. The CPA amplifier was developed to solve these problems. In a CPA laser, the femtosecond pulses from the oscillator are stretched to picoseconds, or even nanoseconds, to reduce the peak power in the amplifier. The total B integral is normally kept below π radians.

2.3 Laser Pulse Propagation

In the previous section, we discussed spatial properties of laser beams with finite transverse dimensions when it propagates in linear or nonlinear media. In this section, we discuss the temporal properties of *plane* wave (infinitely large in the transverse dimension) Gaussian pulses.

2.3.1 Wavelength Bandwidth

As discussed in Chapter 1, the frequency bandwidth of transform-limited Gaussian pulses with a FWHM of τ is

$$\Delta\omega = \frac{4 \ln 2}{\tau}. \quad (2.25)$$

For femtosecond lasers, it is more convenient to express the bandwidth in wavelength, because it can be measured directly with spectrometers. Since

$$\Delta\omega = \Delta\left(\frac{2\pi c}{\lambda}\right) = -\frac{2\pi c}{\lambda_0^2} \Delta\lambda, \quad (2.26)$$

we have

$$\Delta\lambda = \frac{2 \ln 2}{\pi c} \frac{\lambda_0^2}{\tau}, \quad (2.27)$$

where λ_0 is the center wavelength of the pulse. Table 2.1 lists the bandwidths corresponding to five different pulses widths centered at 800 nm. The spectral shapes and ranges are shown in Figure 2.8. The pulses from CPA amplifiers are approximately 25 fs, which has a bandwidth of 38 nm. For a 5 fs pulse from hollow-core fibers, the wings of the spectrum extend to 550 and 1050 nm in the short and long wavelength sides, respectively, which is almost an octave.

2.3.2 Propagation in Linear Dispersive Medium

The index of refraction of a uniform isotropic material is a function of frequency, which is called dispersion. We would like to find simple analytical equations that can be used to calculate the duration and other parameters of a laser pulse at a given location. When the absorption of the material is small, the spectral shape of the pulse does not change much

TABLE 2.1 Bandwidth of Transform-Limited Gaussian Pulses

τ (fs)	5	10	15	20	25
$\Delta\lambda$ (nm)	188.3	94.1	62.8	47.1	37.7

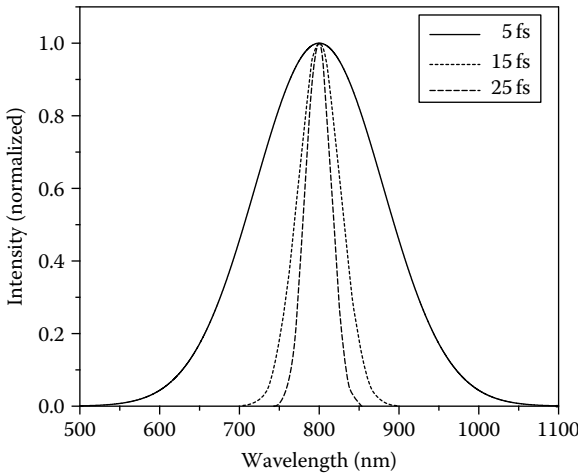


Figure 2.8 Spectra of femtosecond pulses centered at 800 nm.

during the propagation. In this case, we only need to find out the spectral phase variation as the pulse propagates.

When a laser pulse propagates through a dispersive medium, the spectral phase shift is

$$\varphi(\omega) = -\beta L = -\frac{\omega}{c} n(\omega) L, \quad (2.28)$$

where

$n(\omega)$ and L are the index of refraction and the length of the material, respectively

β is the propagation constant used in electric engineering, which is the same as k used in optics and physics

This equation is valid for both the femtosecond laser pulses in this chapter and the attosecond XUV pulses discussed in the Chapter 1.

2.3.2.1 Sellmeier Equation

In laser optics, the index of refraction is often expressed as a function of the wavelength. An analytical expression called the Sellmeier equation is used for calculating n at a given wavelength. As an example, for fused silica, the equation is

$$n^2(\lambda) = 1 + \frac{B_1 \lambda^2}{\lambda^2 - C_1} + \frac{B_2 \lambda^2}{\lambda^2 - C_2} + \frac{B_3 \lambda^2}{\lambda^2 - C_3}, \quad (2.29)$$

where the unit of the wavelength is a micrometer. The six parameters in the equation are found by fitting the measured values of the index of refraction at some discrete wavelength points for a given material. The values for fused silica are given in Table 2.2.

The index of refraction is plotted in Figure 2.9.

The spectral phase can be calculated by inserting the Sellmeier equation into Equation 2.29. However, it is difficult to calculate the pulse duration change using the complicated phase expression.

TABLE 2.2 Parameters in the Sellmeier Equation

Fused Silica	
B_1	$6.96166300 \times 10^{-1}$
B_2	$4.07942600 \times 10^{-1}$
B_3	$8.97479400 \times 10^{-1}$
C_1	$4.67914826 \times 10^{-3}$
C_2	$1.35120631 \times 10^{-2}$
C_3	9.79340025×10^1

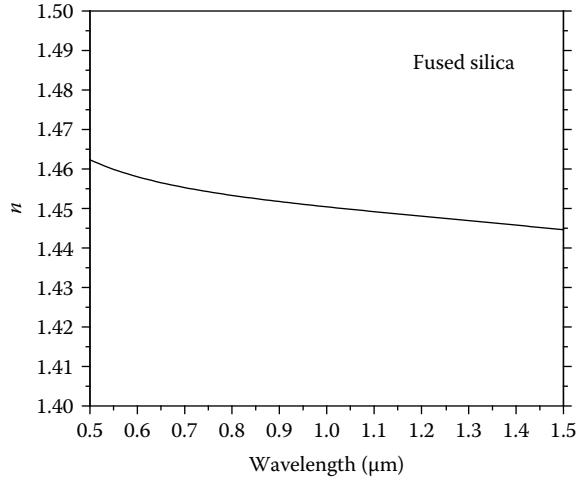


Figure 2.9 The index of refraction of fused silica.

2.3.2.2 Second-Order Approximation

When the pulse duration is much longer than the optical cycle, we can perform Taylor expansion to the spectral phase

$$-\frac{1}{L}\varphi(\omega) = \beta(\omega) = \beta(\omega_0) + \left.\frac{d\beta}{d\omega}\right|_{\omega_0}(\omega - \omega_0) + \frac{1}{2}\left.\frac{d^2\beta}{d\omega^2}\right|_{\omega_0}(\omega - \omega_0)^2 + \cdots, \quad (2.30)$$

and drop the third-order and other high-order terms. Such a simple phase expression leads to the simple expression 1.55 for calculating pulse duration variation in a dispersive medium.

2.3.2.3 Group Velocity Dispersion

The group velocity dispersion (GVD) is given by

$$GVD = \beta'' = \left.\frac{d^2\beta}{d\omega^2}\right|_{\omega_0} = \frac{d}{d\omega} \frac{1}{v_g} = -\frac{1}{v_g^2} \frac{dv_g}{d\omega}. \quad (2.31)$$

To express GVD as a function wavelength, we use the expressions

$$\frac{1}{v_g} = \frac{n}{c} - \frac{\lambda}{c} \frac{dn}{d\lambda}, \quad (2.32)$$

and

$$\frac{d\lambda}{d\omega} = -\frac{\lambda^2}{2\pi c}, \quad (2.33)$$

which implies that

$$\frac{d}{d\omega} = -\frac{\lambda^2}{2\pi c} \frac{d}{d\lambda}. \quad (2.34)$$

TABLE 2.3 Dispersion of Optical Materials

	n	GVD (fs ² /mm)	TOD (fs ³ /mm)	z_D (mm) 5 fs Pulse	10 fs Pulse
Fused silica	1.4535	36.11	27.44	0.25	1.0
BK7	1.5108	43.96	31.90	0.205	0.82
Sapphire	1.7603	58.00	42.19	0.155	0.622

Substituting the result of Equation 2.34 into Equation 2.31 gives the following result:

$$GVD = -\frac{\lambda_0^2}{2\pi c} \frac{d}{d\lambda} \left[\frac{1}{c} \left(n - \lambda \frac{dn}{d\lambda} \right) \right] \Big|_{\lambda_0} = \frac{\lambda_0^2}{2\pi c} \frac{d^2 n}{d\lambda^2} \Big|_{\lambda_0}. \quad (2.35)$$

When the center wavelength is 1.3 μm , the dispersion is zero, which is a reason that telecom industries are interested in this wavelength. At the Ti:Sapphire laser ($\lambda_0 \sim 800 \text{ nm}$) wavelength, the GVD of the fused silica is positive.

The dispersion length expression is the same as for attosecond pulses, i.e.,

$$z_D = \frac{\tau_{in}^2}{(4\ln 2)GVD}. \quad (2.36)$$

Notice that the length depends on the square of the pulse duration. The index of refraction, GVD, and third-order dispersion (TOD) of three commonly used optical materials at 800 nm are listed in Table 2.3. The dispersion lengths of 5 and 10 fs pulses are also listed.

When a transform-limited 10 fs pulse travels through 1 mm fused silica, the pulse is broadened to 14 fs and becomes positively chirped. The positive dispersion of optical materials can be compensated by using prism pairs or grating pairs, introducing negative dispersions.

2.3.2.4 High-Order Dispersions

The third-order and other high-order dispersions must be taken into account when the laser pulse duration approaches the optical cycle. The TOD can be calculated by

$$TOD = \beta''' = \frac{d\lambda}{d\omega} \frac{d}{d\lambda} (GVD) = -\frac{\lambda^2}{2\pi c} \frac{d}{d\lambda} (GVD). \quad (2.37)$$

Other high-order dispersions can be calculated from GVD in a similar way. However, no simple equations like 1.55 have been found that allow us to easily calculate the pulse duration change. For such extremely short pulses, Fourier transforms are used to convert the electric field from the frequency domain to the time domain, to find out the pulse duration.

2.4 Mirrors

Mirrors are the most commonly used optical components in laser systems and in attosecond pulse generation setups. There are two basic

requirements for mirrors used in reflecting high-power short pulses: high reflectance over a broad spectral bandwidth and high damage threshold.

2.4.1 Metal Mirrors

The reflectance of metal mirrors coated on glass substrates can be calculated using the Fresnel equations. The index of reflection of most metals is a complex number, $n_m(\lambda) = n_R(\lambda) - in_I(\lambda)$.

For normal incidence, the reflectance is

$$R(\lambda) = \left(\frac{n_m - 1}{n_m + 1} \right) \left(\frac{n_m - 1}{n_m + 1} \right)^* = \frac{(n_R - 1)^2 + n_I^2}{(n_R + 1)^2 + n_I^2}. \quad (2.38)$$

The most commonly used metal mirror for pulses as short as 5 fs centered at 800 nm is a silver mirror. Its reflection bandwidth is much broader than most ultrafast dielectric mirrors. Metal mirrors are relatively cheap. However, there is a 2%–5% loss for each reflection in the 500–1100 nm range, which is larger when many reflections are required. There is the “minimum mirror principle” in designing laser systems, which reminds people to use as few mirrors as possible to reduce power losses. The damage threshold of a metal mirror is on the order of 1×10^{12} W/cm², which is lower than most dielectric mirrors.

2.4.2 Dielectric Mirrors

Dielectric mirrors are multilayer coatings on glass substrates. The index of refraction changes periodically from layer to layer. The optical thickness of each layer is a quarter of the laser wavelength. A single stack of quarter-wave periodic structures is commonly used for reflecting femtosecond pulses. Multistack coatings do not work for pulses shorter than 100 fs, because group delay between stacks broadens the pulses. For some applications, the high loss and low damage thresholds of metal mirrors are not acceptable. The loss associated with dielectric mirrors is much lower over a narrower spectral range, which means that they can be used for longer pulses. Ultrafast dielectric mirrors can be much more expensive than metal mirrors.

2.4.2.1 High-Energy Mirrors

Commercial high-energy mirrors can withstand high fluence, but have a relatively narrow spectral range. Figure 2.10 shows the reflectance of the Ti:Sapphire laser mirror manufactured by CVI (CVI TLMB), which is frequently used to reflect the amplified pulses in CPA lasers. The damage threshold is 8 J/cm² for 300 ps pulses, which is comparable to the duration of stretched pulses in CPA amplifiers. At normal incidence, the loss is small, in the 740–860 nm range (i.e., a 140 nm bandwidth). By comparing with the pulse spectral curves in Figure 2.8, we can see that it can support 25 fs pulses. The group delay dispersion (GDD) introduced by the mirror is also shown in Figure 2.10. At 45° incidence, the bandwidth is broader for the *s* polarization (i.e., the polarization of the laser field is perpendicular to the plane of incidence) and narrower for the *p* wave (the polarization

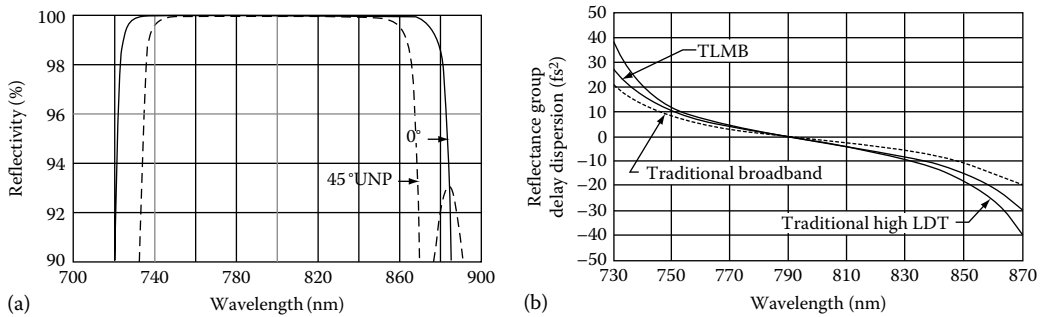


Figure 2.10 (a) Reflectance and (b) second-order phase of commercial high-energy dielectric mirrors, CVI TLMB. The graphs are from the CVI product catalog.

is in the plane of incidence), which is true for almost all dielectric mirrors. Thus, one should use *s* polarization whenever possible.

2.4.2.2 Broadband Mirrors

To reflect ~ 10 fs pulses from oscillators with nanojoule-level energy, we need broadband mirrors with low loss. For example, the measured bandwidth of a commercial broadband mirror (CVI TLM2) is approximately 200 nm for the *s*-polarized light, as shown in Figure 2.11, and can support 15 fs lasers. The damage threshold is low, approximately 100 mJ/cm^2 for 8 ns pulses, which is not sufficient for amplified pulses of a few millijoules when the beam size is close to 1 mm. Figure 2.11 shows the reflectance of commercial broadband dielectric mirrors (CVI TLM2).

2.4.2.3 Broadband High-Energy Mirrors

In 2000, Takada et al. reported the use of a broadband (200 nm bandwidth at 800 nm center wavelength) high-energy mirror, consisting of a stack of broadband $\text{TiO}_2/\text{SiO}_2$ coatings underneath another stack of high-damage-threshold coatings ($\text{ZrO}_2/\text{SiO}_2$), which decrease the intensity on the

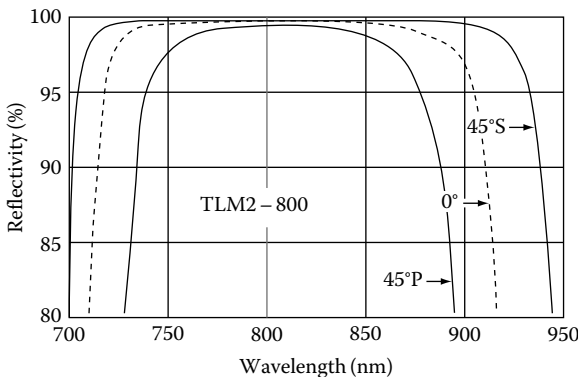


Figure 2.11 Reflectance of commercial broadband dielectric mirrors, CVI TLM2. The graphs are from the CVI product catalog.

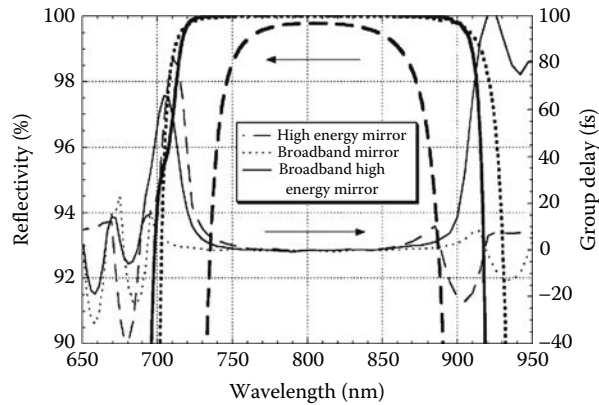


Figure 2.12 Comparison of mirror reflections. (Reprinted with kind permission from Springer Science+Business Media: *Appl. Phys. B*, Broadband high-energy mirror for ultrashort pulse amplification system, 70, 2000, 5189, H. Takada, M. Kakehata, and K. Torizuka.)

broadband coatings. The damage threshold of the broadband high-energy mirror is $>1 \text{ J/cm}^2$, which is comparable to that of the commercial high-energy mirror.

Figure 2.12 shows a comparison of measured reflectivity and delay of the broadband mirror (CVI TLM2), the high-energy mirror (CVI TLM1), and the custom-made broadband high-energy mirror (manufactured by Japan Thin Film Optics Company). This shows that the bandwidth of the broadband high-energy mirror is almost equivalent to that of the broadband mirror. The mirror provides high reflectivity, with a sufficiently broad spectral width to support $\sim 15 \text{ fs}$ pulses for s -polarized light at 45° incidence.

2.4.3 Chirped Mirrors with Negative GDD

When the mirror is designed in such way that the long wavelength light is reflected from the deeper layers of the mirror (and thus travels a longer distance than the short wavelength light that reflects off the surface layers), it introduces negative GDD. Negative GDD may be used to compensate the positive GDD introduced by the material dispersion. The GDD introduced by one pair of chirped mirrors is on the order of -30 to -100 fs^2 , which can compensate for the dispersion induced by a few millimeters of glass.

In the visible and near infrared (NIR) region, the GVD of most optical materials is positive. At least four types of devices have been invented to produce negative GDD so that the material dispersion can be compensated. The chirped mirror is one of them. The other three are prism pairs, grating pairs, and phase modulators.

2.5 Prism Pairs

So far, we have only considered the phase delay from the origin $z=0$ along the propagation direction z , which is $\varphi = -kz$. When a laser beam

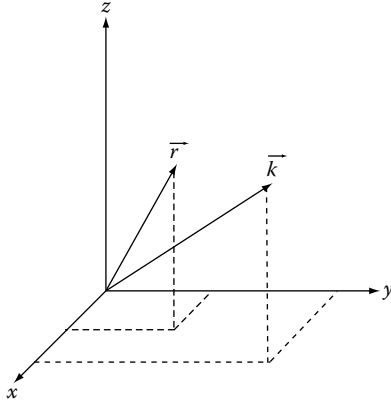


Figure 2.13 Wave vector in Cartesian coordinates

passes through a prism, each frequency component will exit at a different direction. We need to choose a common spatial point in the two-dimensional space to compare the phase between two or more frequency components. Here, we review the analysis derived by Fork et al. in 1984.

To find expressions for the GDD of prism pairs, we first consider a plane wave with frequency ω propagating in a single homogeneous material. The wave vector in the medium of index n is $\vec{k}$. The electric field can therefore be expressed as

$$\mathcal{E}(\vec{r}, t) = E_0 e^{i(\omega t - \vec{k} \cdot \vec{r})}. \quad (2.39)$$

In Cartesian coordinates, $\vec{r} = x\hat{i} + y\hat{j} + z\hat{k}$, and $\vec{k} = k_x\hat{i} + k_y\hat{j} + k_z\hat{k}$, as shown in Figure 2.13. Here, $\hat{i}$, $\hat{j}$, and $\hat{k}$ are the unit vectors in the x , y , and z directions, respectively. The propagation constant in the medium is $k = n\omega/c$. The phase delay between the origin and point at (x, y, z) is $\varphi = -\vec{k} \cdot \vec{r}$.

2.5.1 Phase Delay

For simplicity, we consider the two-dimensional case in Figure 2.14. The phase delay between points A and B is

$$\varphi = \varphi(\vec{r}_B) - \varphi(\vec{r}_A) = (-\vec{k} \cdot \vec{r}_B) - (-\vec{k} \cdot \vec{r}_A) = -\vec{k} \cdot (\vec{r}_B - \vec{r}_A). \quad (2.40)$$

The expression $-\vec{k} \cdot (\vec{r}_B - \vec{r}_A)$ can be understood as the number of wavelengths the wavefront travels between the two points, where the wavefront is perpendicular to the wave vector. For $|\vec{r}_B - \vec{r}_A| = l$, the angle between $\vec{k}$ and $\vec{r}_B - \vec{r}_A$ is θ , thus

$$\varphi(\omega) = -k(\omega)l \cos \theta = -\frac{\omega}{c}n(\omega)l \cos \theta = -\frac{\omega}{c}P(\omega). \quad (2.41)$$

Here

$$P(\omega) = -\frac{c}{\omega}\varphi(\omega) = n(\omega)l \cos \theta \quad (2.42)$$

is the equivalent optical path length.

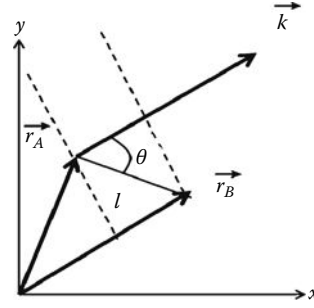


Figure 2.14 Phase delay in a two-dimensional case.

2.5.2 Group Delay Dispersion

The GDD can be expressed as

$$GDD = -\frac{d^2\varphi}{d\omega^2} = \frac{d^2}{d\omega^2} \left(\frac{\omega}{c} P \right) = \frac{\lambda^3}{2\pi c^2} \frac{d^2 P}{d\lambda^2}. \quad (2.43)$$

Therefore, the GDD can be evaluated by the second derivative of the equivalent optical path length,

$$\frac{d^2 P}{d\lambda^2} = \cos \theta \frac{d^2 n}{d\lambda^2} - 2 \sin \theta \frac{dn}{d\lambda} \frac{d\theta}{d\lambda} - n \cos \theta \left(\frac{d\theta}{d\lambda} \right)^2 - n \sin \theta \frac{d^2 \theta}{d\lambda^2}. \quad (2.44)$$

2.5.3 Single Glass Slab

We then consider the case of a pulse refracted by a glass slab of index n . For each frequency component, the wavefront is flat. It is safe to assume that the points A and B are at the input and output surfaces, respectively, as shown in Figure 2.15. We can think that all frequency components originated from point A . The direction of $\vec{k}$ is different for each frequency.

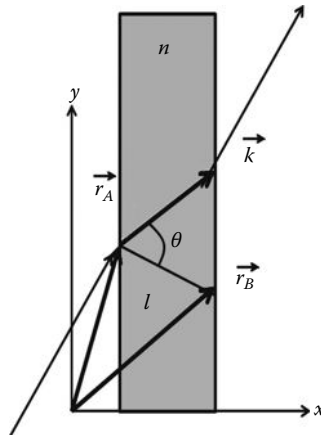


Figure 2.15 Refraction by a glass slab.

Point B is the common point for comparing the phases for different frequency components.

The GDD is usually evaluated at the central frequency. We can choose the wave vector of the center frequency in the direction from A to B , so that $\theta = 0$. In this case, Equation 2.44 can thus be simplified to

$$\frac{d^2 P}{l d\lambda^2} = \frac{d^2 n}{d\lambda^2} - n \left(\frac{d\theta}{d\lambda} \right)^2. \quad (2.45)$$

The negative sign before the term $n(d\theta/d\lambda)^2$ is important, because it means that it is possible to obtain negative GDD by angular dispersion. However, for most optical material in the visible and NIR regions, $d^2 n/d\lambda^2 > n(d\theta/d\lambda)^2$, thus the $\text{GDD} > 0$.

2.5.4 Two Slabs and Prism Pairs

To obtain $\text{GDD} < 0$, we can introduce another slab within the one we just discussed, as shown in Figure 2.16. This slab is, however, made of nothing (vacuum), thus index of refraction $n_v = 1$. The superposition of a glass slab and a vacuum slab create two identical prisms with parallel inner surfaces. The A and B points are chosen at the apices of the two prisms. Again, all frequency components originated from point A . The direction of $\vec{k}$ is different for each frequency. Point B is the common point for comparing the phases for different frequency components. In this case, the function of the second prism is just to collimate all the frequency components. It does not contribute to the phase.

In the vacuum slab, $dn_v/d\lambda = d^2 n_v/d\lambda^2 = 0$, thus

$$\frac{d^2 P}{l d\lambda^2} = -\cos \theta \left(\frac{d\theta}{d\lambda} \right)^2 - \sin \theta \frac{d^2 \theta}{d\lambda^2}. \quad (2.46)$$

If we can choose the wave vector of the center frequency in the direction from A to B , then $\theta = 0$, $\sin \theta = 0$, $\cos \theta = 1$. Finally, we have

$$\frac{d^2 P}{l d\lambda^2} = -\left(\frac{d\theta}{d\lambda} \right)^2 \quad (2.47)$$

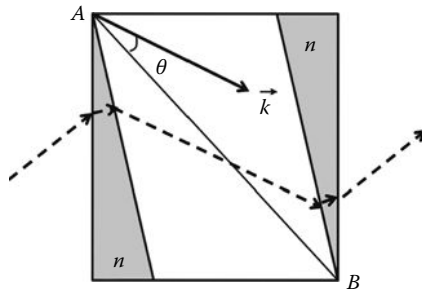


Figure 2.16 Refraction by two prisms. The wave vector of the central wavelength is indicated by the dashed lines. The thick solid line is a shift of the wave vector to show the angle.

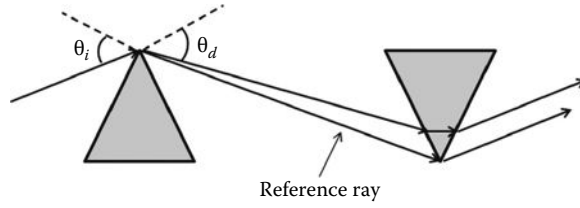


Figure 2.17 Definition of angles.

and

$$GDD = -\frac{d^2\varphi}{d\omega^2} = \frac{\lambda^3}{2\pi c^2} \frac{d^2P}{d\lambda^2} = -l \frac{\lambda^3}{2\pi c^2} \left(\frac{d\theta}{d\lambda} \right)^2. \quad (2.48)$$

The TOD can be described in a similar way, which gives

$$TOD = -\frac{d^3\varphi}{d\omega^3} = -\frac{\lambda^4}{(2\pi)^2 c^3} \left(3 \frac{d^2P}{d\lambda^2} + \lambda \frac{d^3P}{d\lambda^3} \right). \quad (2.49)$$

In some calculations, the total phase, rather than just a few orders of dispersion, needs to be considered. The phase delay in the prism pair can be expressed as

$$\varphi(\omega) = -\frac{\omega}{c} l \cos[\theta_d(\omega_{ref}) - \theta_d(\omega)]. \quad (2.50)$$

Here ω_{ref} is the reference frequency at which the wave vector passes through the apices of the two prisms. θ_d is the refraction angle from the second surface of the first prism, which can be calculated from the incident angle θ_i and the apex angle of the prism α , i.e.,

$$\theta_d(\omega) = a \sin \left\{ n(\omega) \sin \left[\alpha - a \sin \left(\frac{1}{n(\omega)} \sin \theta_i \right) \right] \right\}. \quad (2.51)$$

The angles are shown in Figure 2.17.

2.5.5 Brewster's Angle Configuration

To minimize the reflection loss, the incident angle and the diffraction angle are set to Brewster's angle. Since the two angles are the same, they also correspond to the minimum deviation angle. In this case,

$$GDD = -\frac{d^2\varphi}{d\omega^2} = -l \frac{\lambda^3}{2\pi c^2} \left(\frac{d\theta}{d\lambda} \right)^2 \approx -4l \frac{\lambda^3}{2\pi c^2} \left(\frac{dn}{d\lambda} \right)_{\lambda_0}^2. \quad (2.52)$$

Any prism in the pair can be translated in the direction perpendicular to the beam path by adding a glass length L_p to reduce the magnitude of negative GDD. The total GDD is

$$GDD = -\frac{d^2\varphi}{d\omega^2} \approx -4l \frac{\lambda_0^3}{2\pi c^2} \left(\frac{dn}{d\lambda} \right)_{\lambda_0}^2 + L_p \frac{\lambda_0^3}{2\pi c^2} \frac{d^2n}{d\lambda^2} \Big|_{\lambda_0}. \quad (2.53)$$

The derivatives $(dn/d\lambda)|_{\lambda_0}$ and $(d^2n/d\lambda^2)|_{\lambda_0}$ can be calculated by using the Sellmeier equation of the glass. As an example, for a pair of Brewster

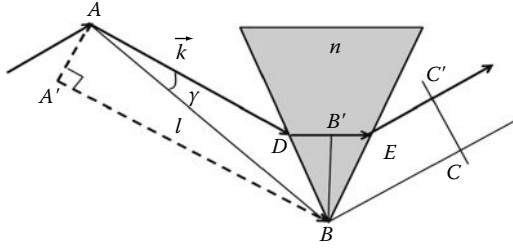


Figure 2.18 The second prism.

prisms using fused silica glass, the GDD at 800 nm is -1000 fs^2 when the distance between them is 1 m, which can compensate the positive GDD of a piece of 3 cm long SQ1 glass.

2.5.6 Effects of the Second Prism

For the prism pair, there is another way to find out the equivalent optical path length P for a particular frequency. In Figure 2.18, we choose the line AB from the apex of prism I to the apex of prism II as the reference ray with frequency ω_{ref} . The optical path length P of the ray corresponding to frequency ω is indicated by the path ADB' .

$A'B$ is parallel to AD , therefore AA' is a wavefront, because it is perpendicular to the wave vector $\vec{k}$. $B'B$ is perpendicular to DE , thus it is also a wavefront. As a result, the optical path length ADB' equals $A'B$, i.e.,

$$P = \overline{A'B} = l \cos(\theta). \quad (2.54)$$

The rays EC' and BC are parallel to each other because the second prism cancels out the first prism's effect on the ray direction. In other words, the second prism collimates rays with different frequencies. Since $C'C$ is perpendicular to EC' and BC , it is also a wavefront. Consequently, the optical path lengths $B'EC'$ and BC are equal, which makes no contribution to GDD.

2.5.7 Double Pass Configuration

When a laser pulse beam propagates through a prism pair, all the frequency components are parallel to each other in the output beam. However, they are separated in the transverse direction, which is called a spatial chirp. This problem can be solved by sending the beam back so that the beam travels through the prism pair twice, as shown in Figure 2.19. For such a double pass configuration, the GDD and high-order dispersions are also doubled. This sort of setup has been used in femtosecond pulse laser oscillators.

Prism pairs have also been used as pulse compressors in chirped pulse amplifiers because of the high throughput. However, the maximum energy that can be compressed is limited to less than 5 mJ by the nonlinear effects in the prisms, as well as by the damage to the prism glass. Grating pairs have been used to compress high-energy laser pulses instead.

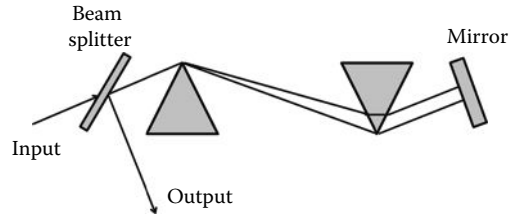


Figure 2.19 Double pass configuration.

2.6 Grating Pairs

We introduce the analysis derived by Treacy in 1969. Consider two gratings in parallel with their grooves facing each other, as illustrated in Figure 2.20. The spacing between them is G , and the grating constant is d , which gives the groove density $1/d$.

We choose B as the common point for comparing phases of different frequency components, which all originated from point A . There are two approaches to obtain the spectral phase. The first approach is the same as we used for prism pairs. The phase delay introduced by the first grating is

$$\varphi(\omega) = -\vec{k} \cdot \vec{r}_B = -\frac{2\pi}{\lambda} G \cos \theta_d(\omega). \quad (2.55)$$

The second grating collimates the rays of all the frequency components, but does not contribute to the phase difference between any two components.

θ_d is the diffraction angle, which is related to the incident angle θ_i by the grating equation

$$[\sin(\theta_d) - \sin(\theta_i)]d = m\lambda, \quad (2.56)$$

where m is the diffraction order.

The second approach is more intuitive. For a plane wave with wave vector $\vec{k}$, the phase delay between points A and B is

$$\varphi(\omega) = -\frac{\omega}{c} P'(\omega) + \varphi_c(\omega). \quad (2.57)$$

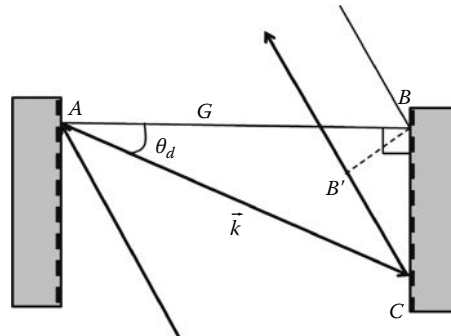


Figure 2.20 Phase delay in grating pairs.

We can define P' as the optical path length from A to B'

$$P'(\omega) = \overline{AC} + \overline{CB'}. \quad (2.58)$$

$\varphi_c(\omega)$ is a term needed for the consistent definition of the phase for the wave in the grating system. Grating diffraction may be characterized in term of phase matching by a 2π phase jump at each ruling in the first-order diffraction. Thus, one has to add 2π times the number of grating lines in the segment BB' to the phase along AC .

$$\varphi_c(\omega) = 2\pi \frac{\overline{BC}}{d} = 2\pi \frac{G \tan(\theta_d)}{d}. \quad (2.59)$$

2.6.1 Phase Matching

Phase matching can be understood using Figure 2.21. AB and $B'B$ are the wavefronts of the incident and diffracted waves. When $\overline{BC} = d$, we have the grating equation

$$\overline{A'C} + \overline{CB'} = m\lambda. \quad (2.60)$$

Here, m is the diffraction order. For the first-order diffraction $m = 1$,

$$\frac{2\pi}{\lambda} (\overline{A'C} + \overline{CB'}) = 2\pi. \quad (2.61)$$

When the phase delay introduced by the propagation in ACB' is 2π , the fields diffracted at points C and B add up constructively, which means that the two waves are phase matched. This explains the 2π phase jump per the ruling in Equation 2.61. For this reason, we argue that each groove in the grating introduced a 2π phase delay. In other words, the wave diffracted at

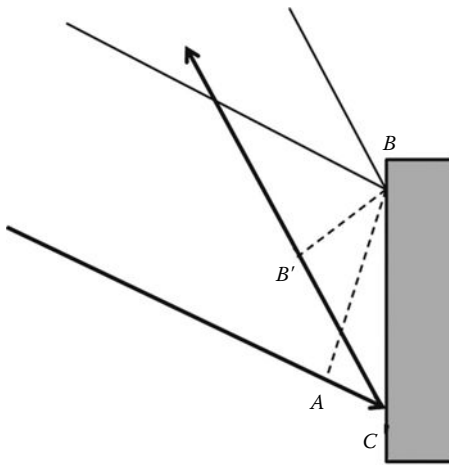


Figure 2.21 Phase matching in grating diffraction.

point B experiences a 2π phase delay with respect to point C , because B is d away from C .

For an arbitrary value of $\overline{BC}$ that contains $\overline{BC}/d$ grooves, the phase matching leads to

$$\frac{\overline{BC}}{d} 2\pi = \frac{2\pi}{\lambda} (\overline{A'C} + \overline{CB'}). \quad (2.62)$$

2.6.2 Phase

Since

$$P' = \overline{AC} + \overline{CB'} = \overline{AA'} + (\overline{A'C} + \overline{CB'}) = G \cos \theta_d + \frac{\overline{BC}}{d} \lambda. \quad (2.63)$$

Insert P' into the equation

$$\varphi(\omega) = -\frac{2\pi}{\lambda} P'(\omega) + 2\pi \frac{\overline{BC}}{d}. \quad (2.64)$$

Now we have

$$\varphi(\omega) = -\frac{2\pi}{\lambda} P(\omega) = -\frac{2\pi}{\lambda} G \cos \theta_d(\omega). \quad (2.65)$$

Here, P is the equivalent optical path length from A to B . Equation 2.65 is the same as the one for prism pairs. In both cases, the GDD is introduced by the angular dispersion.

2.6.3 Group Delay Dispersion

The GDD for a single pass grating pair is

$$GDD = -\frac{d^2 \varphi}{d\omega^2} \approx -\frac{G}{d^2} \frac{\lambda^3}{2\pi c^2} \frac{1}{\cos^2 \theta_d}. \quad (2.66)$$

As in the case of prism pairs, a spatial chirp is introduced by the grating pair. The double pass configuration is used to cancel out the spatial chirp. For example, when two gratings with $d = 1.2 \mu\text{m}$ are separated by 20 cm, the GDD can reach $-3 \times 10^6 \text{ fs}^2$, which is much larger than what other methods can provide. It is for this reason that grating pairs have been used in CPA lasers to stretch pulses to hundreds of picoseconds, or even nanoseconds. However, the throughput of grating pairs is lower compared to prism pairs. For a double pass configuration, the throughput is approximately 60%. For a double pass prism pair, it can be higher than 95%.

2.6.4 Optical Pulse Compressor

The compressor has to compensate for the positive dispersion introduced by the stretcher and the material dispersions in the amplifier such as the gain material and Pockels cell, polarizers, etc. Prism pairs have been used to compress relatively low-energy pulses, although grating pairs are commonly used because they can handle high pulse energy. There are three free parameters that can be chosen to correct the second-, third-, and

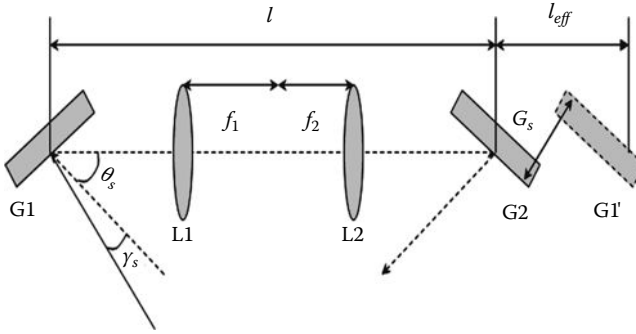


Figure 2.22 The grating stretcher. G1 and G2 are gratings. γ_s is the incidence angle on the first grating. θ_s is the angle between the diffracted beam and the incident beam. L1 and L2 are lenses for a telescope. G1' is the image of the G1 formed by the telescope. l_{eff} is the effective distance and G_s is the effective perpendicular distance between the gratings. (Reprinted from Z. Chang, Carrier envelope phase shift caused by grating-based stretchers and compressors, *Appl. Opt.*, 45, 8350, 2006. With permission of Optical Society of America.)

fourth-order dispersions. They are the separation between the gratings, the incident angle, and the groove density.

2.6.5 Optical Pulse Stretcher

When a telescope is inserted between the two prisms or gratings in the compressor, as illustrated in Figure 2.22, an image of the first grating, G1, is formed behind the second grating, G2, which is G1'. As a result, the sign of the GDD can be inverted, because the grating spacing G_s is a negative number when Equation 2.66 is used to calculate the GDD. Such configurations have been used to stretch optical pulses from the femtosecond oscillator.

For broadband laser pulses, the aberrations introduced by lenses are significant, and mirrors are commonly used instead. A special telescope design, called Offner-type, has relatively small aberrations, making it a desirable configuration.

In many CPA systems, the femtosecond pulse train from a mode-locked Ti:Sapphire laser is sent to the pulse stretcher to be stretched to 100–300 ps. The pulse compressor recompresses the chirped pulses back to tens of femtoseconds.

2.7 Laser Pulse Propagation in Nonlinear Media

2.7.1 Self-Phase Modulation

The intensity of a Gaussian pulse at z is given by

$$I(t) = I_0 e^{-4 \ln 2 \frac{t^2}{\tau_p^2}}. \quad (2.67)$$

The intensity variation in time leads to a time-dependent variation of index of refraction

$$n(t) = n_0 + n_2 I(t). \quad (2.68)$$

When a plane wave Gaussian pulse propagates through a medium with length L , a nonlinear phase shift is added to the temporal phase of the pulse. When the material dispersion is neglected,

$$\phi_{nl}(t) = -\frac{2\pi}{\lambda} I(t) n_2 L. \quad (2.69)$$

This phenomenon is called self-phase modulation (the laser pulse changes its own phase), which is the temporal correspondence of the self-focusing of a Gaussian beam.

The time-dependent nonlinear phase leads to a frequency shift

$$\Delta\omega(t) = \frac{d}{dt} \phi_{nl}(t) = -\frac{2\pi}{\lambda} n_2 L \frac{d}{dt} I(t). \quad (2.70)$$

Equations 2.69 to 2.70 are illustrated in Figure 2.23.

The up and down frequency shifts introduce new frequency components to the pulses, which broadens the bandwidth. The frequency increases near $t = 0$, which means a positive chirp is introduced to the pulse. When the chirp is compensated, shorter pulses can be generated.

For Gaussian pulses,

$$\phi_{nl}(t) = -\frac{2\pi}{\lambda} I_0 e^{-4\ln 2 \frac{t^2}{\tau^2}} n_2 L = -B e^{-4\ln 2 \frac{t^2}{\tau^2}}, \quad (2.71)$$

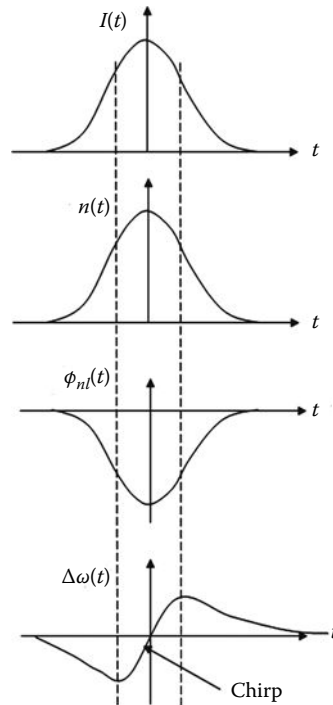


Figure 2.23 Self-phase modulation.

and for $t \ll \tau$,

$$\phi_{nl}(t) \approx -B + (4\ln 2)B \frac{t^2}{\tau^2}, \quad (2.72)$$

that is, the phase is a parabolic function of time, which corresponds to a linear chirp

$$\Delta\omega(t) \approx (8\ln 2) \frac{B}{\tau^2} t. \quad (2.73)$$

Using the chirp parameter introduced in Equation 1.21, we have

$$b = 2 \frac{B}{a}, \quad (2.74)$$

where $a = 2\ln 2/\tau^2$, as defined by Equation 1.23. According to Equation 1.29, when the input pulse is transform-limited with a bandwidth $\Delta\omega_{in}$, the output pulse bandwidth is

$$\Delta\omega_{out} = \Delta\omega_{in} \sqrt{1 + \left(\frac{b}{a}\right)^2} \approx 2B\Delta\omega_{in}. \quad (2.75)$$

A more rigorous derivation yields the width of the output spectrum

$$\Delta\lambda_{out} = 0.86B\Delta\lambda_{in}, \quad (2.76)$$

where $\Delta\lambda_{in}$ is the input spectrum width. Thus, the bandwidth broadening factor is close to the value of the B -integral.

Various fibers have been used to obtain broad spectrum through self-phase modulation. The subsequent sections discuss these types of fibers.

2.7.2 Photonic Crystal Fiber

A standard single-mode fiber guides light by total internal reflection between a core with a high refractive index and a cladding with a lower index. The zero-dispersion $\beta''(\lambda_0) = 0$, is at $\lambda_0 = 1.3 \mu\text{m}$. At the Ti:Sapphire laser's wavelength ($\lambda_0 = 0.8 \mu\text{m}$), $\beta''(\lambda_0) > 0$. As a result, the laser intensity drops during the propagating, due to the broadening of the pulse. The nonlinear effects reduce with propagation length.

In a photonic crystal fiber (PCF), the cladding is obtained by forming a matrix of air and glass, which creates a hybrid air-silica material with a refractive index lower than the core. The hybrid material can be constructed with a structure similar to that found in crystals. PCF fibers are also called microstructured or holey fibers. The zero-dispersion wavelength is engineered at the center wavelength of the laser pulses, λ_0 , by tuning the structure parameters of the fiber. Thus, the intensity is kept almost constant as the pulse propagates. The required B value can be achieved in a short piece of PCF fiber.

2.7.2.1 Highly Nonlinear Fiber

Highly nonlinear PCFs have extremely small cores ($2 \mu\text{m}$) and a cobweb-like microstructure. Because of the small core diameter and proper zero-dispersion wavelength, supercontinuum white light has been generated

using low-energy nanojoule pulses from femtosecond oscillators for locking the carrier-envelope phase, as further discussed in Chapter 3.

2.7.3 Hollow-Core Fibers

Due to material damage, the use of solid core single-mode fibers is limited to low energy (nJ) pulses. Hollow-core fibers filled with noble gases are suitable for broadening the spectrum of laser pulses with higher energies, because the ionization potentials of noble gases are much larger than that of solids. Gases can recover from the ionization when the laser pulses are gone, whereas damage to a solid by the laser is permanent. The strength of the nonlinearity can be achieved by changing the gas type and pressure. It is for these reasons that gas-filled hollow-core fibers have been widely used in few-cycle lasers for attosecond pulse generation.

Wave propagation along hollow waveguides can be thought of as occurring through grazing incidence reflections. Since the losses caused by these multiple reflections greatly discriminate against higher-order modes, only the fundamental mode can propagate in a sufficiently long fiber. The modes of hollow dielectric waveguides with diameters much larger than the wavelength were considered by Marcatili and Schmeltzer. Three types of modes propagate in a hollow waveguide: transverse electric, transverse magnetic, and hybrid modes. For fused silica hollow fibers, the lowest loss mode is the EH_{11} hybrid mode, which has a linear polarization. Its intensity profile as a function of the radial coordinate r is given by

$$I(r) = I_0 J_0^2 \left(2.405 \frac{r}{a} \right), \quad (2.77)$$

where

I_0 is the peak intensity

J_0 is the zero-order Bessel function

a is the core radius

For the same mode, the real, β (propagation constant), and imaginary, $\alpha/2$ (field attenuation coefficient), parts of the propagation constant are given by

$$\beta = \frac{2\pi}{\lambda} \left[1 - \frac{1}{2} \left(\frac{2.405\lambda}{2\pi a} \right)^2 \right], \quad (2.78)$$

$$\frac{\alpha}{2} = \left(\frac{2.405}{2\pi} \right)^2 \frac{\lambda^2}{2a^3} \frac{\nu^2 + 1}{\sqrt{\nu^2 - 1}}, \quad (2.79)$$

where

λ is the laser wavelength in the gas medium

ν is the ratio between the refractive indices of the external (fused silica) and internal (gas) media, which is approximately 1.5

For Gaussian pulses, in the absence of dispersion, the maximum broadening $\delta\omega_{max}$ at the exit of a fiber with length L is given by

$$\delta\omega_{max} = 0.86\gamma P_0 z_{eff} \frac{1}{\tau}, \quad (2.80)$$

where

$$z_{eff} = \frac{(1 - e^{-\alpha L})}{\alpha}, \quad (2.81)$$

P_0 is the pulse peak power

τ is the half-width at the $1/e$ intensity point of the pulse

The nonlinear coefficient

$$\gamma = \frac{n_2 \omega_0}{c A_{eff}}, \quad (2.82)$$

where

$A_{eff} = \pi \alpha$ is the effective mode area

ω_0 is the center frequency of the laser

For argon gas, $n_2/p = 9.8 \times 10^{-24} \text{ m}^2/\text{W atm}$, where p is the gas pressure. For a given fiber with a $140 \mu\text{m}$ diameter, $A_{eff} = 7.4 \times 10^3 \mu\text{m}^2$, one obtains from Equation 2.80 that $\delta\omega_{max} = 6.6 \times 10^{14} \text{ rad/s}$ at $p = 4 \text{ atm}$ and $P_0 = 3.5 \text{ GW}$. Although this value is approximately three times greater than that measured in the spectrum, we can attribute the difference to the neglect of the dispersion effect. Other noble gases such as neon and krypton are also options for spectrum broadening in hollow-core fibers. For krypton, $n_2/p = 2.78 \times 10^{-23} \text{ m}^2/\text{W atm}$.

2.8 Femtosecond Oscillator

Femtosecond seed pulses for the CPA amplifier are provided by a laser oscillator. The stabilization of the carrier-envelope phase of the oscillator is discussed in Chapter 3. Here, we focus on the mechanism of femtosecond pulse generation from a Kerr lens mode-locked Ti:Sapphire oscillator.

A femtosecond laser oscillator has four major components: the gain material, the pump laser, the feedback mirrors that form an optical resonant cavity, and the dispersion compensation optics. A schematic diagram of a femtosecond oscillator is shown in Figure 2.24. In this example, a pair of prisms is used to provide the needed negative GDD.

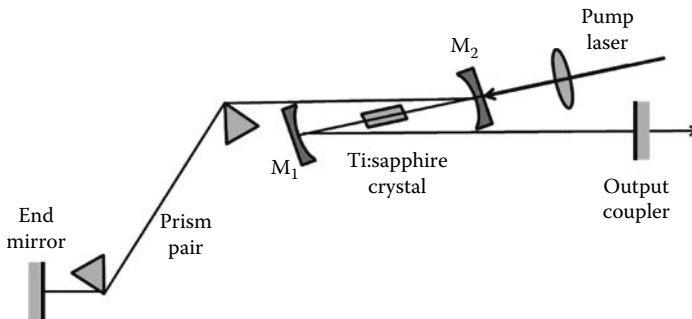


Figure 2.24 Femtosecond laser oscillator.

2.8.1 Ti:Sapphire Crystals

Ti³⁺ ion-doped sapphire crystal (Al₂O₃) is the most commonly used gain material in femtosecond lasers because of broad gain bandwidth and large gain cross sections. Sapphire is a single axis birefringent crystal with excellent optical and mechanical properties. The gain of the *e*-ray (light polarized along the optic axis) is much larger than the *o*-ray (light polarized perpendicular to the optic axis). Therefore, for laser oscillators and amplifiers, the crystal is cut in such a way that its optic axis (*c* axis) is perpendicular to the propagation direction. In many cases, the two end surfaces are cut at the Brewster angle, so that the reflection of the *e*-ray is minimized.

For a 1% atomic doping, the Ti density is $4.56 \times 10^{19} \text{ cm}^{-3}$. Up to 2.5% doping has been used to achieve high gain in a short crystal. Ti³⁺ is a four-level system. The broadband absorption cross section is peaked at approximately 500 nm, which matches the second harmonic of the Nd:YLF (526 nm), Nd:YAG (532 nm), and Nd:YOV₄ (532 nm) lasers, all commonly used as pump lasers. At the wavelength of the latter two lasers, the absorption cross section is $6 \times 10^{20} \text{ cm}^2$. The fluorescence linewidth of the Ti³⁺ is 180 nm, and centered around 790 nm at room temperature, which supports 3 fs pulses. The stimulated emission cross section is $2.8 \times 10^{-19} \text{ cm}^2$ for the light polarized along the optical axis direction.

2.8.2 Principle of Mode Locking

Femtosecond pulses from the oscillator cavity are generated by a mechanism called mode locking. The cavity shown in Figure 2.24 is a folded one. It is more intuitive to discuss mode locking for a linear cavity with a length of *L* in the *z* direction, which contains two end mirrors and the gain medium.

2.8.2.1 Longitudinal Modes

The electric field of the laser light in the linear cavity can be expressed as

$$E(z') = E(z)e^{-ikz}. \quad (2.83)$$

In the steady state operation, the electric field repeats itself after one round trip in the cavity. This requires the phase $2L/\lambda$ to be an integer value. In other words, only fields with certain frequencies can be sustained in the cavity. These angular frequencies are

$$\omega_q = q\omega_{rep}, \quad q = 0, 1, 2, \dots \quad (2.84)$$

where the frequency step is given by

$$\omega_{rep} = 2\pi f_{rep} = 2\pi \frac{c}{2L} = \frac{2\pi}{T_r}, \quad (2.85)$$

where T_r is the time that the light wave takes to make a round trip. Fields with these frequencies are called the longitudinal modes of the cavity. They are evenly distributed on the frequency axis and form a so-called frequency comb. Each mode is a standing wave in the cavity.

2.8.2.2 Mode Locking

When the light of a mode leaves the cavity through the output coupler, its electric field at a spatial point is

$$\varepsilon_q(t) = E_{q0} e^{i(q\omega_{rep}t + \varphi_q)}. \quad (2.86)$$

For simplicity, we assume that the amplitudes of all the modes are identical, $E_{q0} = E_0$. If we can somehow force the phases of all the longitudinal modes to be the same, $\varphi_q = \varphi_0$, which is called mode locking, then the superposition of N modes gives

$$\varepsilon(t) = e^{i\varphi_0} \sum_{n=0}^{N-1} E_0 e^{in\omega_{rep}t} = e^{i\varphi_0} E_0 \frac{1 - e^{iN\omega_{rep}t}}{1 - e^{i\omega_{rep}t}} e^{i\omega_0 t}. \quad (2.87)$$

The corresponding intensity is

$$I(t) \propto |\varepsilon(t)|^2 \propto \frac{\sin^2 \frac{N\omega_{rep}t}{2}}{\sin^2 \frac{\omega_{rep}t}{2}}, \quad (2.88)$$

which is a pulse train.

The duration of each pulse in the train is

$$\tau = \frac{T_{rt}}{N}. \quad (2.89)$$

To generate 10 fs pulses, the phases of $N = 10^6$ modes must be the same.

Equations 2.86 through 2.89 are very similar to the equations for discussing the formation of attosecond pulse trains in Chapter 1. In fact, the high-order harmonic spectrum in the plateau region is essentially an XUV frequency comb. The spacing between the teeth is ~ 3 eV, which corresponds to 1.3 fs. Thus, when the phases of more than two teeth are set to the same value, an attosecond pulse train can be generated.

2.8.2.3 Pulse Picker

The spacing between the pulses is the round trip time of the cavity, which is determined by the cavity length. For most laser oscillators, $T_{rt} = 10 - 20$ ns. This value is chosen so that single pulses can be switched out by a pulse picker based on polarization gating, which consists of a Pockels cell located between two crossed polarizers, as illustrated in Figure 2.25. When a 10 ns high voltage pulse is applied on an electro-optical crystal,

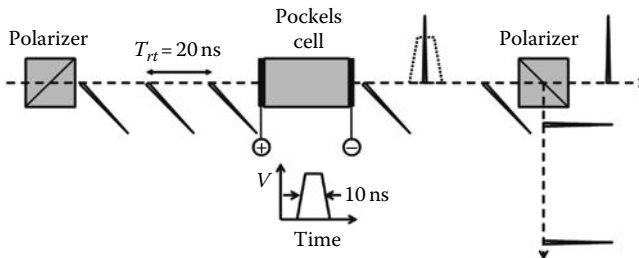


Figure 2.25 A pulse picker based on polarization gating.

the induced birefringence changes the polarization of the pulse synchronized with the gating pulse by 90° . The polarization state of other pulses does not change. As a result, the laser pulse within the voltage pulse passes through the second polarizer, while others are rejected. The minimum gating time is determined by a high voltage circuit, which is typically 1–10 ns. Apparently, the repetition rate of the pulse in the train is f_{rep} , which equals the spacing in the frequency comb. The generation of isolated attosecond pulses with a polarization gating introduced in subsequent chapters shares some similarity with the pulse picker discussed here.

2.8.3 Kerr Lens Mode Locking

When designing a femtosecond oscillator, as shown in [Figure 2.24](#), the green pump beam carrier with approximately 5 W of power is focused on the ~ 2 mm long Ti:Sapphire crystal to create the population inversion. The diameter of the 800 nm beam is reflected back from the end mirror, and the output coupler is ~ 1 mm. The identical radius of curvature is chosen for the two spherical mirrors, in the range of 10–15 cm. They focus the beam to the area in the Ti:Sapphire crystal where sufficient population inversion is created by the pump beam. The distance from M_1 to the output coupler is typically 0.5 m, which is called the short arm. The distance from M_2 to the end mirror is 1 m, which is the long arm. The total cavity length (1.5 m) sets the repetition rate to ~ 75 MHz. Thus, the spacing of the adjacent pulses is ~ 12 ns, which is large enough for pulse picking with Pockles cells.

The focal lengths of the two curved mirrors M_1 and M_2 are rather short (~ 5 cm) to achieve the high intensity in the Ti:Sapphire crystal. The focal spot size can be estimated by

$$w_f = \frac{f}{\pi w} \lambda = \frac{50 \text{ mm}}{2 \text{ mm}} 0.8 \text{ } \mu\text{m} = 20 \text{ } \mu\text{m}. \quad (2.90)$$

For a 10 nJ intracavity pulse with 10 fs duration, the peak intensity at the focal spot is

$$I_{0p} = \frac{1.88}{\pi} \frac{\mathcal{E}}{w_f^2 \tau} = 1.5 \times 10^{11} \text{ W/cm}^2. \quad (2.91)$$

Such pulsed beams induce optical Kerr lens effects in the gain medium, as in the case depicted in [Figure 2.7](#). As a result of self-focusing, the size of the pulse beam is smaller than the CW beam at the location of apertures in the cavity. To favor the pulsed mode over the CW, the cavity could be made to have low loss for the pulse mode over the CW operation. The high loss for the CW beam can be introduced by a hard aperture that cuts off more of the CW beam.

The operation of the oscillator in the pulse mode can also be accomplished by providing higher gain to the pulsed beam than to the CW beam by a better overlap between the pumped region of the gain medium and the pulsed beam. The transverse gain profile is considered as a “soft aperture,” because the gain varies continuously.

2.8.3.1 Stability Range of a Laser Cavity

The cavity can only generate a laser beam when the separations between the two curved mirrors M_1 and M_2 are within a certain range, which is called the stability range of the cavity. For the oscillator shown in [Figure 2.24](#), stable mode locking existed for only two stability regions. The stability range is given by $\delta = d - 2f$, where d is the distance between M_1 and M_2 and f is the focal length of the focusing mirrors. Experimentally, the best position of the crystal and focusing mirrors that would yield a mode-locked state was found by optimizing the CW operation to yield the highest output power. This power was generally approximately 600 mW. Then, the distance between M_1 and M_2 would be slowly increased to reduce the power to roughly 400 mW. Mode locking only begins at the edge of a stability zone. In order to start the mode locking, M_2 is rapidly moved. This motion gives the needed intensity fluctuation in the crystal, to initiate the mode locking.

2.9 Chirped Pulse Amplifiers

There are two challenges in developing femtosecond laser amplifiers that deliver high-energy pulses that are as short as possible, which are critical for attosecond pulse generation. The first challenge is to keep the material dispersion small; the second is to reduce the gain narrowing.

2.9.1 Configurations

The laser pulse energy from femtosecond oscillators is on the order of nanojoules. Therefore, for attosecond pulse generation, amplifiers are needed to boost the energy to millijoules or higher. It is difficult to achieve a gain of 10^6 in a single pass of the gain material; therefore, in most amplifiers, the laser beam travels through the gain medium 10 or more times. There are two types of amplifier designs that accomplished this goal: the multipass amplifier and the regenerative amplifier.

2.9.1.1 Multipass Amplifier

In a multipass amplifier, the laser beam of one pass is separated spatially from that of the next pass. An example of a 5-pass amplifier design is shown in [Figure 2.26](#). The beam size inside the gain material is large because there are no focusing optics. This particular design is suitable for power amplification. Multipass configuration can also be realized by focusing beams into the Ti:Sapphire crystal with two curved mirrors, as depicted in [Figure 2.2](#). It is easier to achieve high, small signal gain with such a design than with the one shown in [Figure 2.26](#).

The main advantage of the multipass design is that no extra dispersive materials are used, which is preferred for generating short pulses. However, the total number of passes is limited due to the spatial separation of beams. As a result, it is difficult to achieve high conversion efficiency from the pump energy to the laser energy.

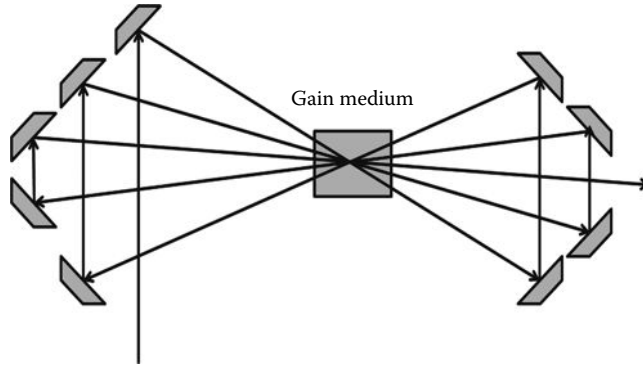


Figure 2.26 Multipass amplifier.

2.9.1.2 Regenerative Amplifier

A regenerative amplifier is similar to a laser oscillator. The gain medium is placed in a cavity. The seed beam is switched into the cavity by a Pockels cell. After traveling back and forth in the cavity for a certain number of trips, the beam is switched out by the Pockels cell. There is no spatial separation between the beams in different passes. The number of passes can be easily controlled by the delay between the switch in and switch out time. Thus, high conversion efficiency can be achieved. However, due to the large amount of material dispersion from the Pockels cell and other extra optics in the cavity, it is more difficult to produce laser pulses as short as those from multipass amplifiers.

2.9.2 Gain Narrowing

2.9.2.1 Gain Cross Section

The frequency dependence of the simulated emission cross section of femtosecond laser materials can be approximated by a Lorentzian line-shape, which is given by

$$\sigma(\omega) = \frac{\sigma_a}{1 + [2(\omega - \omega_a)/\Delta\omega_a]^2}, \quad (2.92)$$

and is shown in [Figure 2.27](#). Here ω_a is the center frequency of the gain. $\Delta\omega_a$ is the FWHM of the line profile. When $\omega - \omega_a = \Delta\omega_a/2$, the value of the cross section is one half of the peak value σ_a .

Expressed in wavelength λ , the cross section is

$$\sigma(\lambda) = \frac{\sigma_a}{1 + [2\lambda(1 - \lambda/\lambda_a)/\Delta\lambda_a]^2}, \quad (2.93)$$

where

λ_a is the wavelength at which the cross section peaks

$\Delta\lambda_a = c\Delta\omega_a/\lambda_a^2$ is the FWHM of the cross section curve

The cross section for Ti:Sapphire with $\Delta\lambda_a = 180$ nm and $\lambda_a = 790$ nm is plotted in [Figure 2.28](#). It is asymmetrical due to the frequency to wavelength conversion. The shape of the curve is significantly different from

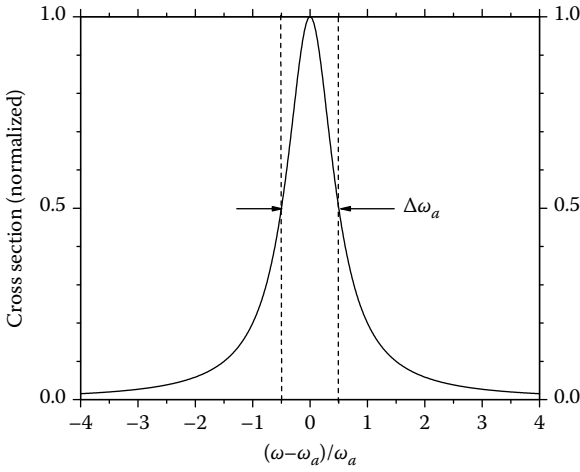


Figure 2.27 Gain cross section as function of frequency.

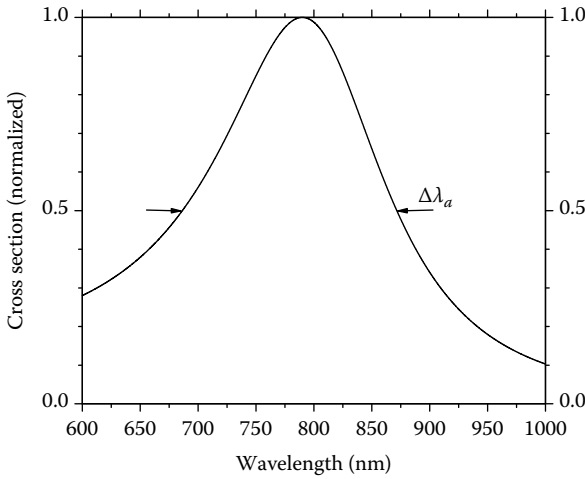


Figure 2.28 Gain cross section as function of wavelength.

the gain curve of the measured Ti:Sapphire result. However, this simple analytic expression allows us to obtain explicit results that can guide the design of the amplifier.

2.9.2.2 Gain Narrowing

The bandwidth of an amplifier is defined as the FWHM of the gain versus the frequency curve, which is also called the 3 dB width because $10 \log_{10}(1/2) = -3.1$. The bandwidth can be calculated by the equation

$$\Delta\omega = \Delta\omega_a \sqrt{\frac{3}{G_{\text{dB}}(\omega_a) - 3}}, \quad (2.94)$$

where the dB gain at the peak wavelength is

$$G_{\text{dB}}(\omega_a) = 10 \log_{10} G(\omega_a). \quad (2.95)$$

TABLE 2.4 The Output Bandwidth Limited by Gain Narrowing

$G(\lambda_a)$	10	10^2	10^3	10^4	10^5	10^6	10^7
$G_{dB}(\lambda_a)$	10	20	30	40	50	60	70
$\Delta\lambda$ (nm)	117.8	75.6	60	51.3	45.5	41.3	38.1
τ_a (fs)	7.8	12.1	15.3	17.9	20.2	22.2	24.1

Expressed in terms of the wavelength, we have

$$\Delta\lambda = \Delta\lambda_a \sqrt{\frac{3}{G_{dB}(\lambda_a) - 3}}. \quad (2.96)$$

The pulse duration supported by the gain bandwidth is

$$\tau = \frac{2\ln 2}{\pi c} \frac{\lambda_a^2}{\Delta\lambda}. \quad (2.97)$$

For a Ti:Sapphire laser, the calculated values are listed in Table 2.4.

The pulse energy from a Ti:Sapphire oscillator is on the order of 5 nJ. For attosecond and high-field experiments, the required energy is a few millijoules. Considering the losses in the chirped pulse amplifier, the net gain should be $\sim 10^7$. The gain narrowing limited bandwidth of the amplifier pulses is ~ 38 nm, which corresponds to ~ 24 fs.

2.9.2.3 Effects of the Seed Pulse Bandwidth

For an input pulse with bandwidth $\Delta\lambda_{in}$, the power spectrum of the amplified pulses is

$$P(\lambda) \propto e^{-4\ln 2 \frac{(\lambda - \lambda_a)^2}{\Delta\lambda_{in}^2}} G(\lambda), \quad (2.98)$$

where the gain is

$$G(\lambda) = G(\lambda_a) e^{-4\ln 2 \frac{(\lambda - \lambda_a)^2}{\Delta\lambda_a^2}}. \quad (2.99)$$

From these two equations, we can determine the bandwidth of the amplified pulses

$$\Delta\lambda_{out} = \Delta\lambda_{in} \frac{\Delta\lambda}{\sqrt{\Delta\lambda_{in}^2 + \Delta\lambda^2}}. \quad (2.100)$$

When the gain is 10^7 , the output pulse bandwidths and transform-limited pulse durations are given in Table 2.5.

Many Ti:Sapphire CPA systems deliver millijoule or higher energy pulses with durations that range from 25 to 40 fs.

TABLE 2.5 The Output Bandwidth Limited by Gain Narrowing and Oscillator Bandwidth

$\Delta\lambda_{osc}$ (nm)	50	75	100	125	150
$\Delta\lambda_a$ (nm)	38.1	38.1	38.1	38.1	38.1
$\Delta\lambda_{out}$ (nm)	30.3	34	35.6	36.4	36.9
τ_{out} (fs)	30.3	27	25.8	25.2	24.9

2.9.3 Gain Narrowing Compensation

Conventional Ti:Sapphire CPA lasers can generate 30 fs pulses up to the 1 J level, with a very low repetition rate. The pulses are much longer than 10 fs because of the gain narrowing effects in the Ti:Sapphire crystal. Seres et al. (Krausz group) developed a laser system to solve the gain narrowing problem, which generated 3 mJ, 10 fs pulses. The scheme can even be scaled to produce high energy. As an example, as shown in Figure 2.29, a laser system that is proposed has a power amplifier to boost the energy of the laser pulses after the hollow-core fiber compressor. Due to the small gain on the power amplifier, the gain narrowing there is reduced, which should preserve the broadband width of the pulses from the hollow-core fiber.

Consider the case where the gain of the power amplifier is on the order of 100 and the gain narrowing limited bandwidth is 76 nm. When the fiber spectrum $\Delta\lambda_{fiber} > 150$ nm, the spectral width of the amplifier output is close to 12 fs.

2.9.3.1 Spectral Shaping

The gain narrowing in the power amplifier can be reduced by introducing spectral shaping. The spectral shaping filter can be described by a spectral transmission function

$$T(\lambda) = \frac{1}{1 + \alpha} \left[1 - \alpha \cos \left(\frac{2\pi}{\Delta\lambda_m} (\lambda - \lambda_a) \right) \right], \quad (2.101)$$

where

α determines the transmission at the center wavelength

$\Delta\lambda_m$ is the peak to peak bandwidth of the filter

For example, one can chose $\Delta\lambda_m = 200$ nm, $\lambda_a = 790$ nm, and $\alpha = 0.65$. The transmission has a dip at the peak of the spectral gain curve, which counteracts the gain narrowing. The phase error introduced in the system will be compensated by an adaptive phase modulator integrated in the optical pulse stretcher. When the filter is used as shown in Figure 2.29, the final output spectrum width is ~ 90 nm. The pulse duration supported by the spectrum can be calculated by

$$\tau = \frac{2\ln 2}{\pi c} \frac{\lambda_a^2}{\Delta\lambda}, \quad (2.102)$$

which yields 10 fs.

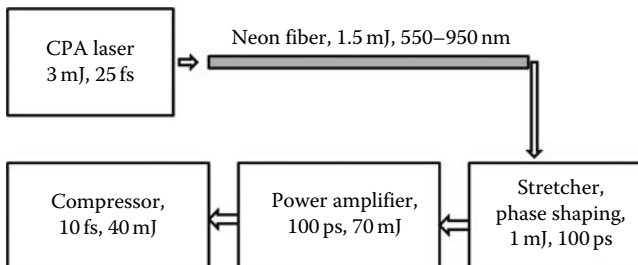


Figure 2.29 Generation of 40 mJ, 10 fs pulses centered at 790 nm with 100 Hz.

2.9.3.2 Optical Parametric Chirped Pulse Amplification

The optical parametric chirped-pulse amplification (OPCPA) technique has been developed to generate high-power sub-10 fs pulses. However, to amplify a chirped pulse with relatively high efficiency, a somewhat complex pumping system is needed, and synchronization between the pumping pulse and seed pulse is critical. A significant amount of efforts have been devoted to developing pump lasers for OPCPA lasers for attosecond pulse generation.

2.10 Pulse Characterization

Laser pulses for generating single isolated attosecond pulses are in the range of 3–30 fs. For generating an attosecond pulse train, the NIR laser can be as long as 100 fs. Linear detectors such as an optical streak camera can measure laser pulses longer than 200 fs, which is too slow. NIR pulses as short as 4 fs can now be measured by the cross-correlation method, using isolated attosecond pulses, as discussed in Chapter 9. However, such measurement is difficult to conduct. Relatively simple methods for characterizing femtosecond laser pulses, based on nonlinear optics, have been developed in the 1990s.

Since the power spectrum of optical pulses can be easily measured by grating spectrometers, one only needs to know the spectral phase to fully characterize femtosecond lasers. The two most commonly used methods for measuring the phase are frequency-resolved optical gating (FROG) and spectral phase interferometry for direct electric-field reconstruction (SPIDER). In this chapter, we only discuss FROG. This method has also been extended to measure attosecond XUV pulses, which is discussed in Chapters 7 and 8. The principle of SPIDER is similar to f -to- $2f$ interferometry for determining the carrier-envelope phase of laser pulses, which is presented in Chapter 3. Another technique called multiphoton intrapulse interference phase scan (MIIPS) is introduced here as well, which can be combined with pulse shaping.

2.10.1 FROG

FROG is an extension of the well-established autocorrelation technique developed in the picoseconds laser era.

2.10.1.1 Autocorrelators

In an autocorrelator, a laser pulse is split into two. One of the split pulses is delayed in time. The two pulses are then combined and are focused collinearly into a second harmonic generation crystal. The second harmonic signal, as a function of the time delay between the two pulses, is the autocorrelation trace, from which one can estimate the pulse duration. There are two types of autocorrelators. The scanning autocorrelator measures pulses that are reproducible, such as the pulses from mode-locked oscillators. It is common to use the Michelson interferometer to introduce the time delay in this type of autocorrelator. In a single-shot autocorrelator, two laser beams with a finite size (~ 1 cm) are crossed with an angle

on the second harmonic crystal. The time delay changes along the crystal surface, which can be measured by an imaging detector such as a CCD camera. Such a device is useful to measure high-power amplified laser pulses with low repetition rate. To estimate the laser pulse duration, a certain pulse shape, such as Gaussian, must be assumed, which is the main source of error.

2.10.1.2 FROG Trace

When the second harmonic signal from an autocorrelator is sent to a spectrometer, the frequency-resolved autocorrelation trace forms a two-dimensional FROG trace. The two axes represent the wavelength and time delay, respectively. An example of the trace obtained from a single-shot FROG setup in the author's lab is shown in Figure 2.30a. An autocorrelation trace can be obtained by integrating the signal in the vertical direction. The pulse shape and phase can be reconstructed from this trace by using phase retrieval algorithms. The result is shown in Figure 2.30c, which gives the pulse from a Ti:Sapphire CPA laser. Since one does not need to make assumptions on the pulse shape, FROG is more accurate than autocorrelators. Furthermore, FROG gives both the pulse shape and the phase.

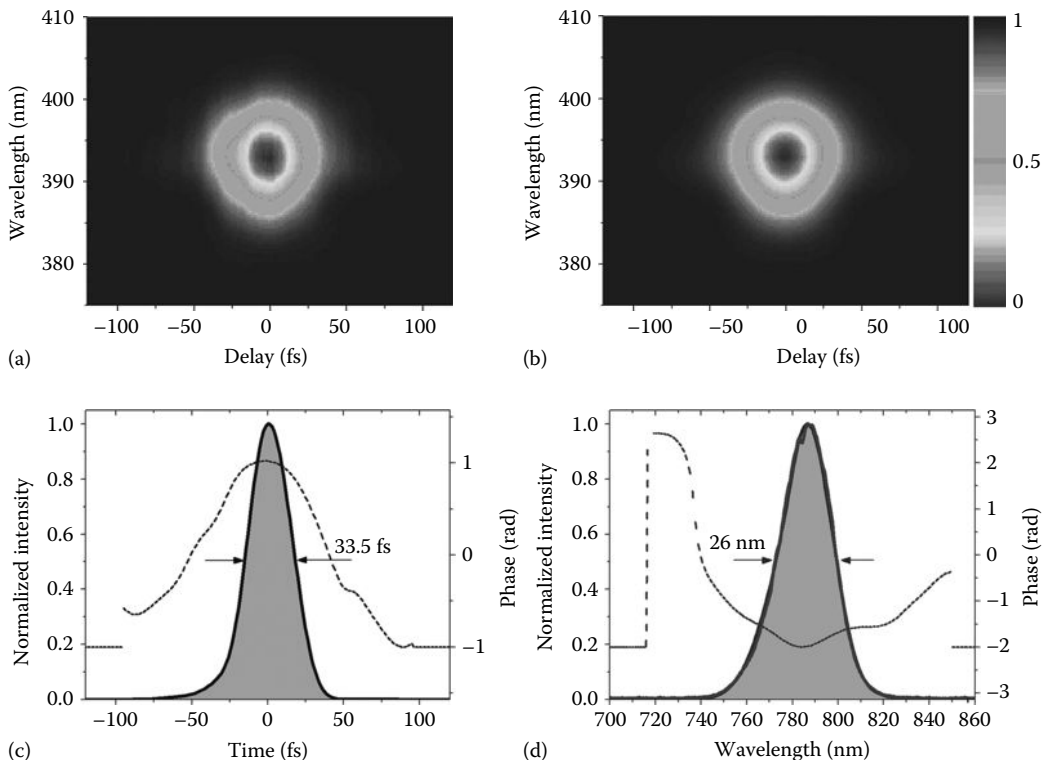


Figure 2.30 Characterization of the laser pulse from the Ti:Sapphire CPA laser in the author's lab by FROG: (a) the measured FROG trace, (b) the reconstructed trace, (c) and (d) the reconstructed pulse and phase in the time and frequency domain.

The second harmonic generation can be considered as a type of amplitude gating because the conversion efficiency increases with the laser intensity. In other words, a weaker pulse cannot pass the gate (being converted to the second harmonic light) as easily as an intense one. We can presume that one of the pulses in the autocorrelator is the one to be measured, while the other is the gating pulse. It is easy to understand this gating idea when the gating pulse is a delta function. In that case, the cross-correlation gives the pulse shape directly. One should keep in mind that the pulse duration cannot be determined by a linear autocorrelation measurement (without the second-order or other nonlinear processes), because it is equivalent to just measuring the power spectrum, as the Fourier transform theory (Wiener–Khinchine Theorem) revealed.

In general, the FROG trace can be expressed as

$$\begin{aligned} S(\omega, T_d) &= \left| \int_{-\infty}^{+\infty} \varepsilon(t) G(t + T_d) e^{-i\omega t} dt \right|^2 \\ &= \left| \int_{-\infty}^{+\infty} E(t) e^{i\phi(t)} G(t + T_d) e^{-i(\omega - \omega_0)t} dt \right|^2. \end{aligned} \quad (2.103)$$

Here $\varepsilon(t) = E(t)e^{i\phi(t)}e^{i\omega_0 t}$ is the complex laser field to be measured. $E(t)$ is the field envelope, $\phi(t)$ is the temporal phase, and $G(t)$ is the gating function, which equals $\varepsilon(t)$ for autocorrelation measurement. T_d is the time delay between the two pulses on the second harmonic crystal. The center frequency ω_0 can be determined by measuring the power spectrum. The modular square comes from the fact that photodetectors such as CCD cameras measure intensity, not electric field. It is worthy pointing out that the integral in Equation 2.103 is actually the Fourier transform of the quantity $E(t)e^{i\phi(t)}G(t + T_d)$, which can be calculated by fast Fourier transform algorithms during the phase retrieval. In [Figure 2.30a](#), the vertical axis represents wavelength instead of frequency ω .

2.10.1.3 Phase Retrieval

Extracting the laser field $\varepsilon(t)$ from the FROG trace $S(\omega, T_d)$ is a two-dimensional phase retrieval problem, and iterative algorithms are frequently used for finding the phase. The algorithm uses an initial guess for $\varepsilon(t)$ and $G(t)$ to generate a spectrogram, $S(\omega, T_d)$, using Equation 2.103. During each iteration, the magnitude of $S(\omega, T_d)$ is replaced by the square root of the experimentally measured FROG trace. The next guess for the amplitudes and phases of $\varepsilon(t)$ at each time step (which is also $G(t)$ for second harmonic FROG) is determined by the minimization of the difference between the measured and calculated FROG trace with respect to $\varepsilon(t)$. The algorithm is repeated until the error is reduced to an acceptable value. When $\varepsilon(t)$ is digitized to an array that has ~ 100 time steps, one needs to find the global minimum in a 100-dimension space! Various minimum searching methods have been tried to speed up the iterations. In the example shown in [Figure 2.30](#), the reconstructed FROG trace at the end of the iteration is shown in [Figure 2.30b](#), which looks very similar to the measured trace.

The degree of success of the phase retrieval is judged by the FROG error that is defined as the per-pixel rms error of the FROG trace, as well as by comparing the measured and reconstructed power spectra, which is called frequency marginal comparison. The marginal comparison in the given example is shown in Figure 2.30d. In this case, the phase retrieval is accurate, because the measured and reconstructed power spectra are almost indistinguishable.

2.10.1.4 Principal Component Generalized Projection Algorithm

A major drawback of many iterative phase retrieval algorithms is the slow speed. A powerful algorithm, called the principal component generalized projection algorithm (PCGPA), has been developed for fast phase retrieval. In a blind FROG retrieval algorithm, the PCGPA is started using random phases for the initial guess for $\varepsilon(t)$ and $G(t)$. A discrete complex matrix,

$$S(\omega, T_d) = \int_{-\infty}^{+\infty} \varepsilon(t)G(t + T_d)e^{-i\omega t} dt, \quad (2.104)$$

is constructed by creating an outer product of $\varepsilon(t)$ and $G(t)$ arrays.

In the second step, the magnitude of the newly constructed FROG trace is replaced by the square root of the magnitude of the experimental FROG trace. In the third step, the trace is converted to the time-domain FROG trace by use of an inverse Fourier transform by column. The final step in the first iteration is to convert the time-domain FROG trace to the outer product form. If the intensity and phase of the FROG trace are correct, this matrix would have only one non-zero eigenvalue. The eigenvector corresponding to this eigenvalue is the function $\varepsilon(t)$. The complex conjugate of the eigenvector of the transpose of the outer product matrix is the gating function, $G(t)$. If the matrix has more than one non-zero eigenvalue, only the outer product pair with the largest weighting factor (i.e., the principal component) is kept for the second iteration. This is the origin of the name PCGPA.

The second iteration is started by constructing a new FROG trace from the signal vector and the gate vector obtained from the singular value decomposition of the outer product form matrix. The process is repeated until the FROG trace error is reduced to an acceptable value. Since there is no multidimensional minimization in each iteration, PCPGA converges much faster than many other FROG algorithms; in fact, it has been used in real-time FROG. PCPGA has also been used in the characterization of attosecond XUV pulses, as discussed in Chapters 7 and 8.

2.10.2 Multiphoton Intrapulse Interference Phase Scan

In many cases, the pulses from femtosecond laser systems are not transform-limited due to spectral phase errors. This is particularly true for pulses that come out from the gas-filled hollow-core fibers. To minimize the width of the pulses, one can correct the phase errors by tuning some lasers components, such as the grating spacing in the stretchers and compressors, while measuring the spectral phase using FROG. MIIPS

provides an alternative approach for optimizing the laser system, which combines the phase measurements with phase correction.

2.10.2.1 Setup

The setup for a MIIPS measurement of the few-cycle laser pulse from the hollow-core fiber is shown in Figures 2.31 and 2.32. It consists of a spatial light modulator (SLM) for phase modulation, a second harmonic generation (SHG) crystal, and an optical spectrometer. For a laser pulse with the spectral amplitude $|\tilde{E}(\omega)| = U(\omega)$, which is known from the power spectral measurement, the spectral phase to be determined is $\varphi(\omega)$. MIIPS finds the spectral phase by adding a known chirp to the pulse while examining the second harmonic signal. The chirp is added to the laser by the phase modulator.

2.10.2.2 Principle

The second harmonic signal reaches a maximum when the added chirp cancels the chirps of the input laser pulses. Therefore, the spectral phase of



Figure 2.31 The major component of a MIIPS setup.

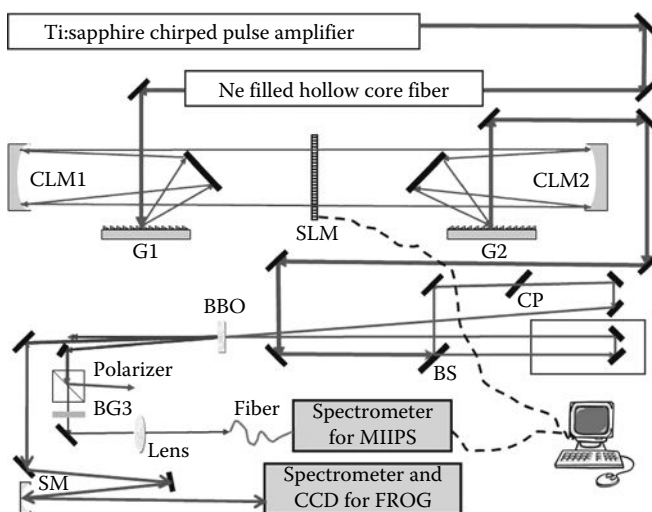


Figure 2.32 The adaptive phase modulator. After hollow-core fiber, the chirped white-light pulses were sent to the SLM through gratings (G1, G2) and cylindrical mirrors (CLM1, CLM2). The output beam was directed to the BBO. The central SH beam was used for FROG measurement, and one side SH beam was used as MIIPS feedback signal. The α -BBO polarizer and the BG3 band-pass filter worked together to eliminate the fundamental beam. The MIIPS retrieved phase was applied on SLM to compress the pulse. BS, beam splitter; CP, compensation plate; SM, spherical mirror. The dashed line represents the feedback loop. (Reprinted from H. Wang, Y. Wu, C. Li, H. Mashiko, S. Gibertson, and Z. Chang, Generation of 0.5 mJ, few-cycle laser pulses by an adaptive phase modulator, *Opt. Express*, 16, 14448, 2008. With permission of Optical Society of America.)

the laser pulse, $\varphi(\omega)$, can be obtained from the added chirp value, $\varphi_A(\omega)$, which is known.

The total phase of the pulse in the SHG crystal is

$$\varphi_T(\omega) = \varphi(\omega) + \varphi_A(\omega). \quad (2.105)$$

When the phase-matching bandwidth of a thin SHG crystal is much broader than the pulse bandwidth, the second harmonic can be written as

$$S(2\omega) \propto \left| \int d\Omega \left\{ U(\omega + \Omega) U(\omega - \Omega) e^{i[\varphi_T(\omega + \Omega) + \varphi_T(\omega - \Omega)]} \right\} \right|^2. \quad (2.106)$$

When the total spectral phase depends linearly on the frequency around a given frequency ω , that is, $\varphi_T(\omega + \Omega) = \varphi_T(\omega) + \eta\Omega$, a maximum second harmonic signal is generated at 2ω . Here η is a constant (The phase is flat when $\eta = 0$). For such phases, $\varphi_T(\omega + \Omega) + \varphi_T(\omega - \Omega) = [\varphi_T(\omega) + \eta\Omega] + [\varphi_T(\omega) - \eta\Omega] = 2\varphi_T(\omega)$, which can be taken out of the integral. As a result, all the products of spectral amplitude inside the integral add up constructively, which yields $S(2\omega) \propto \left| \int d\Omega \{ U(\omega + \Omega) U(\omega - \Omega) \} \right|^2$.

The challenge is to find the correct $\varphi_A(\omega)$. If the higher-order phase errors are ignored, $\varphi_T(\omega + \Omega) = \varphi_T(\omega) + \eta\Omega + \frac{1}{2}\varphi_T''(\omega)\Omega^2$. The second harmonic spectrum has a maximum at 2ω , when the second-order phase distortion $\varphi_T''(\omega)$ is equal to zero at ω . Thus, finding the added second-order phase (chirp) that makes $\varphi_T''(\omega)$ equal to zero at ω can be accomplished by maximizing the second harmonic signal while changing the amount of the added chirp at ω .

2.10.2.3 Experimental Approach

Intuitively, one would add a parabolic phase to introduce the chirp at ω , and then change the shape of the parabola to find the optimum chirp there. This process is repeated for all the frequencies in the spectrum to map out the spectral phase. However, this is rather time-consuming. An alternate method that is used in MIIPS is much faster. In MIIPS, varying the value of the added chirp at a given frequency is done by modulating the phases periodically

$$\varphi_A(\omega) = \alpha \cos(\gamma\omega - \delta). \quad (2.107)$$

The chirp is

$$\varphi_A''(\omega) = -\alpha\gamma^2 \cos(\gamma\omega - \delta). \quad (2.108)$$

By scanning one parameter, the phase angle δ , the chirp at all frequency points is changed simultaneously. It is possible to find the optimum added chirp at each frequency of the whole spectral range by scanning δ over 2π . $\varphi_A''(\omega)$ can cover the range from $-\alpha\gamma^2$ to $\alpha\gamma^2$, which is the range of the chirp that can be measured by the chosen MIIPS parameters.

In principle, other forms of phase function may also accomplish the same result. However, choosing function 2.107 is smart. When the spectrum of the second harmonic signal, $S(\omega, \delta)$, is measured while scanning δ , the maximum signal for different frequencies will show up at different δ positions. Thus, the second-order phase of $\varphi(\omega)$ at all frequencies can be determined simultaneously. Experimentally, δ is scanned over several π radians to make sure the patterns are repeatable.

When the laser pulse is linearly chirped, φ'' is the same for all frequencies, that is, $\varphi''(\omega) = \text{const.}$ At the maxima of second harmonic signal, $\varphi_A''(\omega)$ is equal to $-\varphi''$. As a result, the lines drawn along the second harmonic maxima obey the following equation:

$$\omega = \frac{1}{\gamma} \left[\delta + \cos^{-1} \left(-\frac{\varphi''}{\alpha\gamma^2} \right) \right]. \quad (2.109)$$

There are several lines when δ is scanned over several π radians. These lines are straight, and parallel to each other. The slope of the lines is $1/\gamma$. In general, the separations between the lines are not equal. For transform-limited laser pulses, $\varphi'' = 0$, the lines are equally spaced by π radians.

2.10.2.4 High-Order Phases

The above analysis ignores the high-order phases. In fact, the two-dimensional MIIPS patterns, $S(\omega, \delta)$, are also sensitive to the high-order phases. For the third-order phase, the phase derivative $\varphi''(\omega) = C\omega$, where C is a constant. The lines drawn along the second harmonic maxima follow

$$\alpha\gamma^2 \cos(\gamma\omega - \delta) = C\omega, \quad (2.110)$$

which are not straight lines. The total spectral phase, including the third-order and high-order phases, can be determined by iterative approaches that change the added phase until the MIIPS lines become equally spaced parallel straight lines.

2.11 Few-Cycle Pulses

An isolated attosecond pulse is generated within half an NIR driving laser cycle. Ideally, one would prefer to use a half-cycle driving laser; however, such NIR high-power pulses are extremely difficult to generate and handle. When laser pulses containing many optical cycles are used to generate single isolated attosecond pulses, the energy contained in most of the cycles of the driving lasers is wasted. To make things even worse, they can produce unwanted satellite attosecond pulses. In general, the NIR to XUV conversion efficiency and the attosecond pulse contrast can be improved by using few-cycle driving lasers.

To produce few-cycle driving laser pulses centered between 750 and 800 nm, ~ 30 fs laser pulses from Ti:Sapphire CPA lasers are sent to gas-filled hollow-core fibers to broaden the spectral bandwidth. The positive chirp of the pulses introduced by the self-phase modulation processes and the material dispersion is then removed by chirped mirrors, as shown in [Figure 2.2](#), or by other optical devices with negative GDD, such as phase modulators, as shown in [Figure 2.32](#). For lasers centered at $1.5 \mu\text{m}$ or at even longer wavelengths, one can use a fused silica plate that has negative GDD to compensate the chirp.

2.11.1 Chirped Mirror Compressor

Chirped mirrors are commonly used for compensating the positive chirp of the white-light pulses from the hollow-core fiber. The final pulse

duration is typically in the 5–10 fs range. Sub-4 fs pulses with 400 μJ energy have been generated with specially designed ultrabroadband chirped mirrors and by reducing the duration and fine-tuning the phase of the pulses seeding the hollow-core fibers. The main advantage of chirped mirrors is the high throughput and simplicity.

However, the spectral phase of the white-light pulse from the hollow fiber is very sensitive to the input laser parameters. The phase changes from day to day due to the CPA laser output variation. For a given set of chirped mirrors, the negative dispersion is fixed, and it is therefore difficult to accommodate the daily phase variations of the pulses from the hollow-core fiber. As a result, the compressed pulse duration and shape may change from day to day, which in turn affects the attosecond pulse generation process.

Furthermore, owing to the interference effects at the air/mirror interface, GDD modulation versus wavelength is found in most commercially available mirrors. Matched pairs of mirrors have been designed to overcome this problem, but not completely. Design and fabrication of chirped mirrors that can compensate fourth-order or even high-order dispersions is still in progress. These high-order phases affect both the duration and the contrast of the compressed pulses.

2.11.2 Adaptive Phase Modulator*

It has been demonstrated that the chirp of the white-light pulses from the gas-filled hollow-core fiber could be removed by using an adaptive phase modulator. As compared to the chirped mirrors, it is expected that both GDD and high-order phases can be compensated. One should be able to optimize the compensation to cope with the day-to-day variations in the fiber output. The setup in the author's lab is illustrated in [Figure 2.32](#). The adaptive phase modulator is constructed with a zero-dispersion stretcher and an SLM.

2.11.2.1 Zero-Dispersion Stretcher

The zero-dispersion stretcher consists of two reflective diffraction gratings (G1 and G2) and two telescope mirrors (CLM1 and CLM2) in the $4f$ configuration. The focal length, f , of the two mirrors is the same. The separation between the two mirrors is $2f$. The two gratings are placed symmetrically on each side of the telescope. The distance between the two gratings is four times the focal length of the mirror, hence the name “ $4f$ configuration.” The grating stretcher is similar to the one shown in [Figure 2.22](#), except that the spacing between one grating and the image of another grating, G_s , is set to zero; consequently, the GDD introduced by the gratings is also zero. This is the reason that the $4f$ configuration is also known as the zero-dispersion stretcher.

2.11.2.2 Spatial Light Modulator

A zero-dispersion stretcher is used to separate different frequency components in the plane crossing the common focal points of the two

* More information can be found in Wang, H., Y. Wu, C. Li, H. Mashiko, S. Gilbertson, and Z. Chang, Generation of 0.5 mJ, few-cycle laser pulses by an adaptive phase modulator, *Opt. Express* 16, 14448 (2008).

mirrors, which is called the Fourier plane. In the author's lab, a 640 pixels liquid crystal SLM is located at the Fourier plane. To avoid chromatic aberration when each wavelength component of the white light is focused to the Fourier plane, mirrors are used for the telescope instead of the lenses that are used in [Figure 2.22](#). Cylindrical mirrors focus each wavelength component to a line on the SLM to avoid damaging it by the high-power pulses from the hollow-core fibers. The thickness of the SLM is fixed. The index of refraction of each liquid crystal pixel can be varied independently by a voltage applied on the two surfaces of each pixel, which changes the phase of the light passing through that pixel. In this manner, the phase of the 640 frequency components can be individually and independently varied to compress the white-light pulses.

2.11.2.3 MIIPS for Compressing Pulses from Hollow-Core Fibers

Compared to chirped mirrors, phase modulation at each frequency can be conveniently set to a desired value to compensate both the low- and high-order phase errors for generating the shortest pulse possible for a given spectrum from the fiber. In addition, the phase modulator can be operated in adaptive mode to cope with the day-to-day phase variations of the white-light pulses from the fiber. The contrast of the pulse would be limited by the modulations in the white-light spectrum, instead of by the high-order phases. The spectrum phase of the white light can be determined by the FROG, SPIDER, or MIIPS methods. The setup that combines MIIPS with SLM is depicted in [Figure 2.32](#).

In an experiment done by the author's group, ~ 30 fs pulses centered at 790 nm, with a more than 2 mJ pulse from a kHz CPA laser, were coupled into a 1 m long hollow-core fiber filled with 30 psi of neon gas. The inner diameter of the fiber core was 0.4 mm. Neon gas was used because the ionization under the high input laser energy is less than that of argon or krypton. The ionization should be avoided, otherwise the plasma defocusing lens at the entrance of the hollow-core fiber may reduce the coupling efficiency and cause deterioration of the spatial mode of the beam exiting the fiber. The incapability of compressing pulses with more than 10 mJ limited the attosecond photon flux, which is a major drawback of the hollow-core fiber pulse compressor.

The spectrum of the 30 fs laser pulses from the CPA covers a narrow range, typically from 750 to 850 nm. Self-phase modulation in the fiber produced white light covering 500 to 1000 nm wavelength, as shown in [Figure 2.33](#). The pulses coming out from the fiber with 1.1 mJ were sent to the adaptive phase modulation to remove their chirp. The throughput of the whole phase modulator (including the gratings, mirrors and the SML) is $\sim 50\%$, which leads to an output pulse energy of 0.55 mJ. The overall frequency response is good enough to support sub-5 fs transform-limited pulses, which is nearly the same as what the fiber spectrum can support. Comparing to the chirped mirrors, the throughput of the phase modulator is less by a factor of two, which is its weakness.

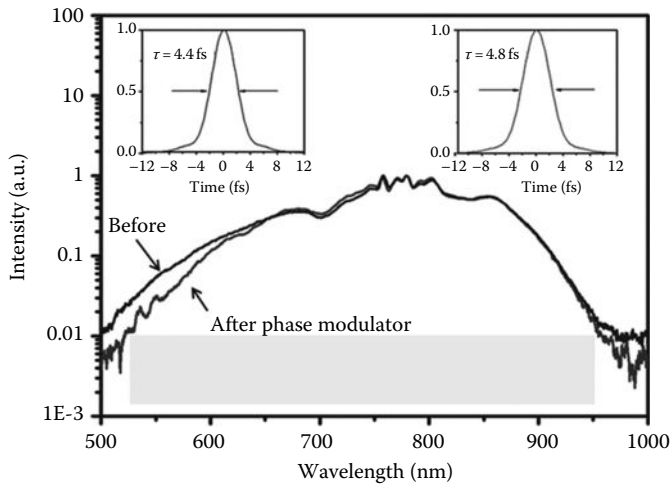


Figure 2.33 The white-light spectrum before the phase modulator and after the phase modulator. The inset shows the transform-limited pulses for both spectra. (Reprinted from H. Wang, Y. Wu, C. Li, H. Mashiko, S. Gibertson, and Z. Chang, Generation of 0.5 mJ, few-cycle laser pulses by an adaptive phase modulator, *Opt. Express*, 16, 14448, 2008. With permission of Optical Society of America.)

2.11.2.4 White-Light Chirp Compensation

A barium borate (BBO) crystal, which has higher second harmonic conversion efficiency than other crystals, is placed after the phase modulator for taking the MIIPS trace. A very thin crystal of 10 μm thickness was used for phase matching over the broad fundamental spectrum range. The pulse compression is done by modulating the spectrum phase with the SLM while simultaneously recording the second harmonic spectrum with an optical spectrometer. The second harmonic spectrum is sensitive to the spectral phase of the white light, and thus can serve as the feedback signal.

The MIIPS pattern before correcting the chirp is shown in Figure 2.34a. The curvature of the pattern indicates that high-order phases are significant. An iterative scheme is used to compensate the chirp. The chirp of the white-light spectral phase, $\varphi''(\omega)$, can be approximately determined from the MIIPS pattern, assuming that the high-order phases can be ignored, as discussed in Section 2.10.2. Next, the spectral phase, $\varphi(\omega)$, is calculated from the white-light spectral phase $\varphi''(\omega)$. To flatten the white-light phase, $-\varphi(\omega)$ is then applied on the SLM. Since high-order dispersions are neglected in this first step, the same operation is repeated for several iterations. The white-light phase becomes more and more flat as the integration number increases. The iterations are stopped when the $\varphi(\omega)$ variation is less than π radians over the whole fundamental spectrum.

The final MIIPS pattern is shown in Figure 2.34b. The evenly spaced parallel second harmonic strip distribution indicates that the compressed pulses are close to being transform-limited, as explained in Section 2.10.2. The phase $\varphi(\omega)$ obtained from the last iteration is shown in Figure 2.35. The pulse duration calculated from the measured spectrum and phase is 4.86 fs, which is a two-cycle pulse, as shown in the insert.

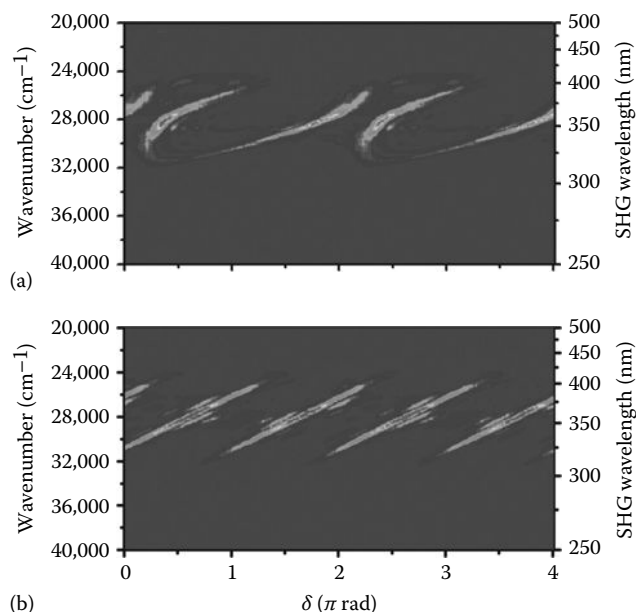


Figure 2.34 The MIIPS traces ($\alpha = 5$, $\gamma = 7$ fs): (a) from the first iteration and (b) from the last iteration. (Reprinted from H. Wang, Y. Wu, C. Li, H. Mashiko, S. Gibertson, and Z. Chang, Generation of 0.5 mJ, few-cycle laser pulses by an adaptive phase modulator, *Opt. Express*, 16, 14448, 2008. With permission of Optical Society of America.)

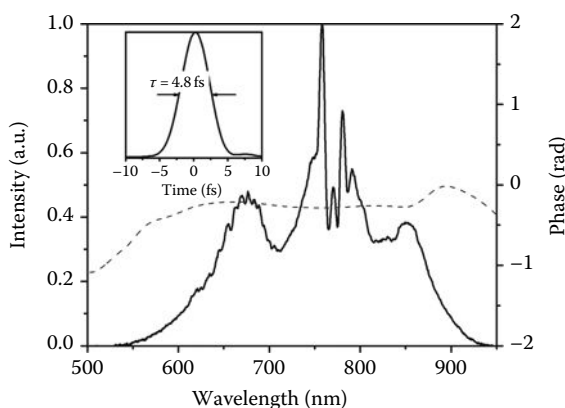


Figure 2.35 The spectral phase of the compressed laser pulse measured by MIIPS. (Adapted from H. Wang, Y. Wu, C. Li, H. Mashiko, S. Gibertson, and Z. Chang, Generation of 0.5 mJ, few-cycle laser pulses by an adaptive phase modulator, *Opt. Express*, 16, 14448, 2008. With permission of Optical Society of America.)

2.11.2.5 FROG Measurements

To confirm that the chirp of the white-light pulse is indeed well compensated by using MIIPS, the duration of the compressed pulse is measured by FROG, which is a very robust method. The MIIPS and FROG methods shared the same BBO crystal, as shown in [Figure 2.32](#). In the single-shot

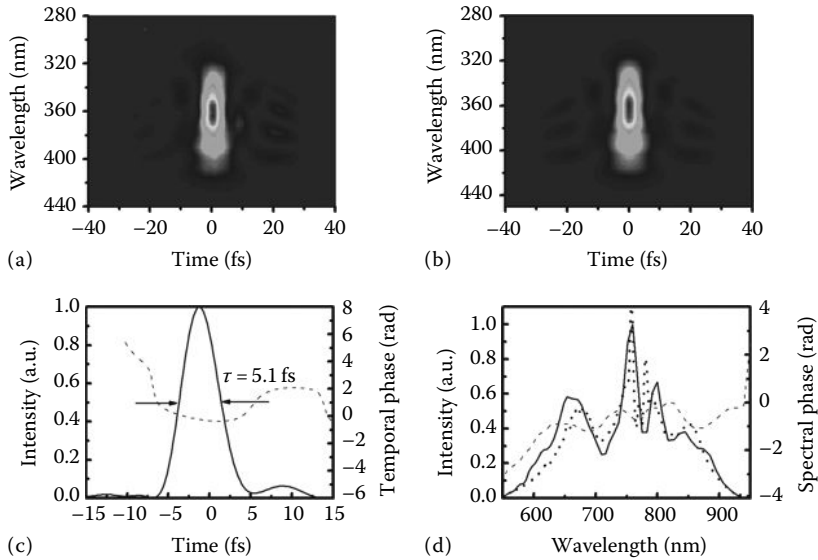


Figure 2.36 Characterization of the laser pulse by FROG: (a) the measured FROG trace, (b) the reconstructed FROG trace, (c) the retrieved pulse shape and phase (dashed curve), (d) the retrieved power spectrum and phase (dashed curve) and independently measured spectrum (dotted curve). The FROG error is 0.5%, and the trace is at 256×256 grids. (Reprinted from H. Wang, Y. Wu, C. Li, H. Mashiko, S. Gibertson, and Z. Chang, Generation of 0.5 mJ, few-cycle laser pulses by an adaptive phase modulator, *Opt. Express*, 16, 14448, 2008. With permission of Optical Society of America.)

FROG setup, three second harmonic beams exit the BBO crystal. The center beam is used by the FROG measurement because it is produced by both fundamental beams. One of the side beams that is generated from only one white-light beam is sent to the MIIPS. In such a configuration, the MIIPS and FROG methods measured the pulse at the same location, which is critical for few-cycle pulse characterization, because such pulses can be easily lengthened by the dispersion of air. The measured and reconstructed FROG patterns are shown in Figure 2.36a and b, respectively. The retrieved pulse duration is 5.1 fs, as shown in Figure 2.36c. The spectral phase is shown in Figure 2.36d. The phase is rather flat. The few-cycle duration and the high pulse energy, 0.5 mJ, make the pulse compressed by the adaptive phase modulator useful for generating isolated attosecond pulses.

2.12 Summary

Since the first demonstration of the Kerr lens mode locking of a Ti: Sapphire oscillator in Sibbett's lab in 1991, all-solid-state femtosecond oscillators and amplifier lasers have become reliable tools for ultrafast, high-field physics studies. Many of the special requirements for attosecond pulse generation—such as short pulse duration, high repetition rate, large energy, as well as stabilization of the carrier-envelope phase—have been met. Developing lasers that allow the generation of even shorter, more energetic attosecond pulses are still in progress.

Problems

- 2.1 Is the intensity of a laser beam from a laser point axially symmetric? Propose an experimental scheme to find out the axial symmetry.
- 2.2 For a laser beam carrying 1 W of power, (a) plot the intensity at $r=0$ as a function of z in the Rayleigh range and (b) plot the intensity at $z=0$ as a function of r . Assume that beam waist $w(z=0)=20\text{ }\mu\text{m}$.
- 2.3 Compare the Rayleigh ranges of 400 and 800 nm laser beams. Assume the spot size is $10\text{ }\mu\text{m}$ for both beams.
- 2.4 Compare the divergence angles of 400 and 800 nm laser beams. Assume the spot size is $10\text{ }\mu\text{m}$ for both beams.
- 2.5 Plot the radius of curvature as a function of z .
- 2.6 Compare the focal spot sizes of 400 and 800 nm laser beams. Assume the beam size at the lens is 20 mm for both beams. The focal length of the lens is 500 mm.
- 2.7 A plano-convex lens is used to focus a laser beam. Which surface should the beam incident on to have smaller spherical aberration?
- 2.8 Derive Equation 2.11, the focal length of a mirror. What is the focal length for a concave mirror with a 1 m radius of curvature?
- 2.9 Calculate the spectral bandwidth of 5 fs pulses when the center wavelength is at 400, 800, and 1600 nm.
- 2.10 What is the upper limit of the laser intensity to keep the B integral in a 10 mm long Ti:Sapphire crystal below π ?
- 2.11 A 300 ps laser pulse with 10 mJ is incident on a TLMB mirror. What is the smallest laser spot size on the mirror without damaging the mirror?
- 2.12 A 1 m long hollow-core fiber with $250\text{ }\mu\text{m}$ inner diameter is filled with air. The n_2 at 1 atmosphere pressure is $5 \times 10^{-19}\text{ cm}^2/\text{W}$. A 1 mJ, 20 fs laser pulse centered at $0.8\text{ }\mu\text{m}$ wavelength propagates through the fiber. Calculate the bandwidth (in nanometers) of the pulse after the fiber, and compare with the input bandwidth. Assume that transverse profile in the fiber is uniform.
- 2.13 To compress the pulse from the fiber in Problem 2.12, how many reflections on chirp mirrors are needed? The GDD of the mirror is -30 fs^2 per reflection. What is the pulse duration of the compressed pulse?
- 2.14 When a 2 mm long Ti:Sapphire crystal with 0.15% atomic doping is pumped by a 5 W frequency-doubled Nd:YVO₄ laser (532 nm), and the beam size in the crystal is $5\text{ }\mu\text{m}$,
 - a. How much power is absorbed by the crystal?
 - b. What is the density of the population inversion?
 - c. What is the small signal gain?
- 2.15 The Lorentzian linewidth of Ti:Sapphire is $\Delta f_a = 10^{14}\text{ Hz}$. The stimulated emission cross section is $2.8 \times 10^{-19}\text{ cm}^2$. The upper state lifetime of the Ti:Sapphire is $3.2\text{ }\mu\text{s}$. When a laser beam passes through the gain medium (Ti:Sapphire), its power is changed from 1 nW to 10 mW.
 - a. What is the power gain?
 - b. What is the gain bandwidth in nanometers?

- c. What is the gain bandwidth in nanometers when the power gain is 1?
 - d. When gain saturation changes the gain bandwidth by 30%, what is the laser beam size inside the Ti:Sapphire crystal?
- 2.16** In the first-order autocorrelation, the detector is a photodiode. The laser field to be measured is $\varepsilon(t) = |\varepsilon(t)|e^{i[\omega t + \phi(t)]}$. Prove that the output signal is $I_D(\tau) = 2 \int_{-\infty}^{+\infty} \varepsilon^2(t) dt + \int_{-\infty}^{+\infty} |\varepsilon(t)| |\varepsilon(t - \tau)| \cos[\phi(t) - \phi(t - \tau) + \omega\tau] dt$, where τ is the delay. For a 5 fs Gaussian pulse centered at 800 nm, plot $I_D(\tau)$.
- 2.17** Compare $I_D(\tau)$ in Problem 2.16 of two pulses with the same bandwidth, 20 nm, centered at 800 nm. One is transform-limited; the other has a 30 fs² chirp. Also plot their pulse shape.
- 2.18** Compare the relative SH signals of two pulses with the same bandwidth, 20 nm, centered at 800 nm. One is transform-limited; the other has a 30 fs² chirp.
- 2.19** Calculate the GDD of a single pass prism compressor at the 800 nm center wavelength. The distance between the two fused silica prisms is 1 m. The incident angle on the prism is the Brewster angle. The wavelength at the apex of the second prism is 780 nm.
- 2.20** Calculate the GDD of a single pass grating compressor at the 800 nm center wavelength. The distance between the two gratings is 1 m. The incident angle on the 1200 line/mm grating is the 60° angle.
- 2.21** A student wants to drill a 100 μm diameter hole through a piece of metal with a laser beam. The laser beam diameter πw is 10 mm. The wavelength of the CW CO₂ laser beam is 10.6 μm .
- a. Choose the focal length of the focusing lens. (Assuming the diameter of the hole equals the focal spot diameter $2w_0$.)
 - b. If the power of the laser beam before the focusing lens is 1 kW, what is the power at the focus?
 - c. What is the peak intensity of the laser on the lens? What is the peak intensity at the focus?
 - d. Is it possible to drill a 1 μm diameter hole with this laser? Give the reasons.
- 2.22** The upper state lifetime of the Ti:Sapphire crystal is 3.2 μs . A Ti:Sapphire rod is pumped by a 150 mJ, 100 ns pulse with the wavelength centered at 532 nm. Assume the pump energy is absorbed in a 1 cm long, 2 mm diameter region of the rod with a power absorption coefficient of 5/cm. The peak laser wavelength is 790 nm.
- a. How many millijoules of the pump energy transmit through the crystal?
 - b. How many millijoules of the pump energy become heat?
 - c. What is the population density of the upper level immediately after the pump pulse?
 - d. Plot the change of the population density of the upper level with time.
 - e. When the spontaneous emission from the rod is measured with a fast photodiode and an oscilloscope, what will the voltage versus time curve be?

2.23 Cauchy's equation of fused silica glass is $n(\lambda) = 1.4580 + 0.00354/\lambda^2$, where λ is in micrometers.

- a. What is the phase velocity at $0.8 \mu\text{m}$?
What is the phase velocity at $1.6 \mu\text{m}$?
- b. What is the group velocity for center wavelength at $\lambda_0 = 0.8 \mu\text{m}$?
What is the group velocity for center wavelength at $\lambda_0 = 1.6 \mu\text{m}$?
- c. What is the group velocity dispersion for center wavelength at $\lambda_0 = 0.8 \mu\text{m}$?
What is the group velocity dispersion for center wavelength at $\lambda_0 = 1.6 \mu\text{m}$?
- d. When a transform-limited 20 fs pulse centered at $\lambda_0 = 0.8 \mu\text{m}$ propagates through the glass, will the pulse duration increase, decrease, or remain unchanged?
When a transform-limited 20 fs pulse centered at $\lambda_0 = 1.6 \mu\text{m}$ propagates through the glass, will the pulse duration increase, decrease, or remain unchanged?

2.24 A 25 fs transform-limited Gaussian pulse centered at the $0.8 \mu\text{m}$ wavelength passes through a 1 mm glass window. Its linear refractive index of glass is 1.5 and its nonlinear refractive index coefficient is $n_{2I} = 1 \times 10^{-16} \text{ cm}^2/\text{W}$. The $1/e^2$ radius of the Gaussian beam is 5 mm and the pulse energy is 6.0 mJ. (The dispersion of the glass and the self-focusing can be neglected.)

- a. What is the refractive index of glass at the peak of the pulse and at the center of the beam?
- b. On the leading (or rising) edge of the pulse, will the refractive index increase, decrease, or remain unchanged with time?
On the falling edge of the pulse, will the refractive index increase, decrease, or remain unchanged with time?
- c. What is the Gaussian chirp parameter of pulse before it enters the glass and after the pulse leaves the glass?
- d. What is the B integral in the center of the beam and at the peak of the pulse?
- e. What is the bandwidth of the pulse before it enters the glass and after the pulse leaves the glass?

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Stabilization of Carrier-Envelope Phase

The concept of carrier-envelope (CE) phase is introduced in Chapter 1. For the generation of attosecond pulse trains, the CE phase of the driving lasers does not need to be stabilized. However, many gating schemes for the generation of single isolated attosecond pulses require the stabilization of CE phase, which is discussed in detail in Chapter 8.

A typical setup for generating few-cycle, high-power, CE phase stable laser pulses is shown in [Figure 3.1](#). First, the change in rate of the CE phase of the pulses from the laser oscillator is locked. Then, pulses with identical CE phase are selected by a pulse picker and sent to the chirped pulse amplifier (CPA). However, CE phase variation from pulse to pulse can be introduced by the components in the CPA amplifier and the subsequent spectral broadening and pulse compression stages. Therefore, the CE phase fluctuation of the final output pulses needs to be restrained for isolated attosecond pulse generation.

In 2003, the CE phases of amplified laser pulses were stabilized for the first time by the Krausz group. In this chapter, we discuss the CE phase properties in the chirped pulse amplifiers and the hollow-core fiber compressors. Commonly used CE phase locking techniques are also introduced.

3.1 Introduction

3.1.1 Definition of Carrier-Envelope Phase

3.1.1.1 Linearly Polarized Field

The electric field of a linearly polarized, transform-limited laser pulse at a fixed point in space can be represented as follows:

$$\varepsilon(t) = E(t) \cos(\omega_0 t + \varphi_{CE}), \quad (3.1)$$

where

$E(t)$ is the pulse envelope function

ω_0 is the carrier angular frequency

φ_{CE} is the CE phase

pulses,” which describe the field oscillation when peak of envelope is located at $t = 0$. When CE phase of the laser system shown in [Figure 3.1](#) is stabilized, the residual phase noise is about 100 mrad. At the 750 nm center wavelength, it corresponds to $0.1 \times (2500/2\pi) = 40$ as in time.

3.1.1.2 Circularly Polarized Field

Assuming that the laser beam is propagating along the z axis, the electric field of a circularly polarized Gaussian pulse can be resolved into two mutually polarized components, one along x axis and the other along y axis:

$$\varepsilon_x(t) = E_0 e^{-2\ln(2)\frac{t^2}{\tau^2}} \cos(\omega_0 t + \varphi_{CE}), \quad (3.3)$$

$$\varepsilon_y(t) = E_0 e^{-2\ln(2)\frac{t^2}{\tau^2}} \sin(\omega_0 t + \varphi_{CE}). \quad (3.4)$$

In this case, the CE phase determines the direction of the field maximum in the xy plane. For $\varphi_{CE} = 0$, the electric field reaches the maximum value, E_0 , at $t = 0$, which points to the x direction.

3.1.1.3 Elliptically Polarized Field

The field of elliptically polarized Gaussian pulses can also be resolved into two mutually polarized components, which differ only slightly from the circular case. For this circumstance,

$$\varepsilon_x(t) = E_0 e^{-2\ln(2)\frac{t^2}{\tau^2}} \cos(\omega_0 t + \varphi_{CE}), \quad (3.5)$$

and

$$\varepsilon_y(t) = \xi E_0 e^{-2\ln(2)\frac{t^2}{\tau^2}} \sin(\omega_0 t + \varphi_{CE}). \quad (3.6)$$

Here, $\xi \leq 1$ is the ellipticity. In this polarization type, the CE phase and the ellipticity together determine the direction of the field maximum rather than the CE phase alone. However, just as with circularly polarized fields, for $\varphi_{CE} = 0$, the electric field reaches the maximum value, E_0 , at $t = 0$, which points to the x direction.

3.1.2 Physics Processes Sensitive to Carrier-Envelope Phase

3.1.2.1 Sub-Cycle Field Strength Variation

As the width of linearly polarized pulse approaches a single cycle, the electric field envelope amplitude, $E(t)$, changes significantly within half of the cycle. For example, for the 5 fs cosine pulse shown in [Figure 3.2](#), the ratio between the highest electric field peak and the adjacent peak is 1:0.917. On the contrary, there is no difference in strength between the two highest peaks (one is positive and the other is negative) around $t = 0$ for the sine pulse, i.e., the ratio is 1:1. Such a ratio difference is the origin of CE phase effects in a variety of high-field processes such as above-threshold ionization (ATI), tunneling ionization, and high-harmonic/attosecond pulse generation. Ionization of atoms by a strong laser field is discussed in Chapter 4. The physics processes that are susceptible to CE phase, such as ATI, in turn can be used to measure CE phase, which are discussed further in [Section 3.10](#).

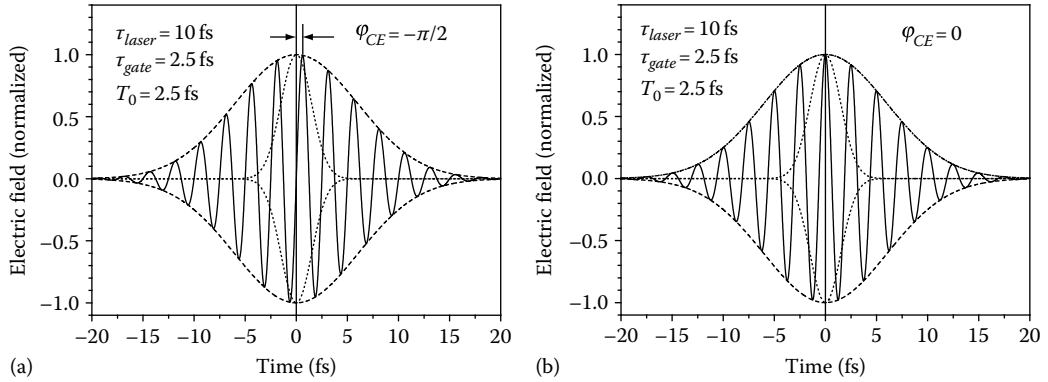


Figure 3.3 A one-cycle gating in a multi-cycle laser. Dashed line is the laser envelope whereas the solid line is the field oscillation. The dotted line is the gating function. (a) A sine pulse. (b) A cosine pulse.

3.1.2.2 Sub-Cycle Gating

For generating isolated attosecond pulses using polarization gating or double optical gating (discussed in Chapters 4 and 8), the effective electric field inside the gate can be expressed as follows:

$$\mathcal{E}_{eff}(t) = g(t) \cos(\omega_0 t + \varphi_{CE}), \quad (3.7)$$

where $g(t)$ is the gating function with a duration of a fraction of a laser cycle, and the center of the gate occurs at $t = 0$. It represents transmission of a gate to individual attosecond pulses in a train separated by half or a full optical cycle. In these cases, even if the laser pulse is many cycles long, the opening time of the gate for single attosecond pulse extraction is on the order of half to one cycle, as shown in Figure 3.3. The duration of laser pulse in Figure 3.3 is 10 fs, and the gating pulse is 2.5 fs, which is the same as the laser cycle.

Here, the envelope in the definition of CE phase should stand for the envelope of the gating function, and not the field envelope of the laser pulse. Although the ratio of the field strength between two adjacent peaks around $t = 0$ does not differ much for the two CE phase values when the laser pulse is long, CE phase effects can still be observed as long as the gating function is narrow. For some CE phase values, only one attosecond pulse can pass through the gate, whereas for other CE phases, two pulses could be generated. As the CE phase of the laser pulse from chirped pulse amplifiers changes from shot to shot, the generated attosecond pulses also vary. Thus, it is crucial to stabilize and control the CE phase for generating single isolated attosecond pulses.

3.2 Carrier-Envelope Phase and Dispersion

3.2.1 Effects of Group and Phase Velocity Difference

3.2.1.1 Group and Phase Velocity

When a linearly polarized plane wave Gaussian pulse propagates in the z direction, if we ignore the change of the pulse width during the propagation, the electric field

$$\varepsilon(z, t) = E_0 e^{-\frac{2\ln(2)}{\tau^2} \left(t - \frac{z}{v_g}\right)^2} \cos \left[\omega_0 \left(t - \frac{z}{v_p}\right) + \varphi_{CE} \right]. \quad (3.8)$$

It states that the carrier wave propagates with the phase velocity v_p , whereas the pulse envelope travels with the group velocity v_g . In the retarded frame

$$t' = t - \frac{z}{v_g}, \quad (3.9)$$

which is the frame that moves with the pulse envelope, and the field can be expressed as

$$\varepsilon(z, t') = E_0 e^{-\frac{2\ln(2)}{\tau^2} t'^2} \cos \left[\omega_0 t' + \omega_0 \left(\frac{1}{v_g} - \frac{1}{v_p} \right) z + \varphi_{CE} \right]. \quad (3.10)$$

Thus, the CE phase at position z is

$$\varphi_{CE}(z) = \omega_0 \left(\frac{1}{v_g} - \frac{1}{v_p} \right) z + \varphi_{CE}(z=0). \quad (3.11)$$

Consequently, the CE phase shift is

$$\Delta\varphi_{CE} = \varphi_{CE}(z) - \varphi_{CE}(z=0) = \omega_0 \left(\frac{1}{v_g} - \frac{1}{v_p} \right) z. \quad (3.12)$$

This indicates that the CE phase variation is due to the difference in the group and phase velocity $(1/v_g - 1/v_p)|_{\omega_0}$. As a comparison, we recall that the pulse duration change is caused by the group velocity dispersion $d/d\omega(1/v_g)|_{\omega_0} \approx 1/\delta\omega(1/v_g|_{\omega_0+\delta\omega} - 1/v_g|_{\omega_0})$. Apparently, for plane wave pulses propagating in vacuum, where $v_g = v_p = c$, the CE phase does not change with distance. Here c is the speed of light in vacuum.

3.2.1.2 Gouy Phase and Carrier-Envelope Phase

As discussed in Chapter 2, the Gouy phase is defined as phase difference between the focusing Gaussian beam and a plane wave. For a focusing Gaussian beam propagating in vacuum, the group velocity of the pulse is approximately equal to the phase velocity of a plane wave c . Consequently, the Gouy phase

$$\psi(z)|_{\omega_0} = \omega_0 \left(\frac{1}{c} - \frac{1}{v_p} \right) z = \omega_0 \left(\frac{1}{v_g} - \frac{1}{v_p} \right) z. \quad (3.13)$$

When the transverse beam profile of the Gaussian pulses is a Gaussian function, the electric field in the retarded frame can be expressed as

$$\varepsilon(r, z, t') = E_0 e^{-\frac{2\ln(2)}{\tau^2} t'^2} \frac{w_0}{w(z)} e^{-\frac{r^2}{w^2(z)}} e^{-k \frac{r^2}{2R(z)}} e^{i\psi(z)|_{\omega_0}} \cos[\omega_0 t' + \varphi_{CE}], \quad (3.14)$$

where

E_0 is the peak amplitude on axis at the beam waist where the spot size is w_0

$w(z)$ is the beam size at point z (the $1/e^2$ intensity radius)

$R(z)$ is the radius of curvature of the wavefront

Equation 3.14 can also be expressed as

$$\varepsilon(r, z, t') = E_0 e^{-\frac{2\ln(2)}{r^2} t'^2} \frac{w_0}{w(z)} e^{-\frac{r^2}{w^2(z)}} e^{-k \frac{z^2}{2R(z)}} \cos[\omega_0 t' + \psi(z)|_{\omega_0} + \varphi_{CE}]. \quad (3.15)$$

The on-axis field is

$$\varepsilon(z, t') = E_0 e^{-\frac{2\ln(2)}{r^2} t'^2} \frac{w_0}{w(z)} \cos[\omega_0 t' + \psi(z)|_{\omega_0} + \varphi_{CE}]. \quad (3.16)$$

The CE phase

$$\varphi_{CE}(z) = \psi(z)|_{\omega_0} + \varphi_{CE}(z=0) \quad (3.17)$$

or

$$\Delta\varphi_{CE} = \varphi_{CE}(z) - \varphi_{CE}(z=0) = \psi(z)|_{\omega_0} = \text{atan}\left(\frac{z}{z_R}\right), \quad (3.18)$$

where z_R is the Rayleigh range. In the confocal region where the attosecond generation target is located, the phase velocity is higher than c and the CE phase introduced by the Gouy phase shift is significant. The target length should be much smaller than the Rayleigh range; otherwise some parts of the target medium produce an isolated attosecond pulse, whereas other parts generate a pair of attosecond pulses.

3.2.1.3 Index of Refraction

For a medium, such as a glass window, with an index of refraction $n(\omega)$, the group velocity

$$\frac{1}{v_g} = \frac{d\beta}{d\omega} \Big|_{\omega_0} = \frac{d}{d\omega} \left(\frac{\omega}{c} n \right) \Big|_{\omega_0} = \frac{n}{c} + \frac{\omega_0}{c} \frac{dn}{d\omega} \Big|_{\omega_0} = \frac{1}{v_p} + \frac{\omega_0}{c} \frac{dn}{d\omega} \Big|_{\omega_0}, \quad (3.19)$$

where $\beta(\omega) = (\omega/c)n(\omega)$ is the propagation constant. The CE phase change over a distance z is

$$\Delta\varphi_{CE}(z) = \left(\frac{1}{v_g} - \frac{1}{v_p} \right) z = \frac{\omega_0^2}{c} \frac{dn}{d\omega} \Big|_{\omega_0} z. \quad (3.20)$$

Since

$$\frac{dn}{d\omega} = \frac{dn}{d\lambda} \frac{d\lambda}{d\omega} = -2\pi \frac{c}{\omega^2} \frac{dn}{d\lambda}, \quad (3.21)$$

we can also express the CE phase change by

$$\Delta\varphi_{CE}(z) = -2\pi \frac{dn}{d\lambda} \Big|_{\lambda_0} z, \quad (3.22)$$

where the derivative can be conveniently calculated by using the Sellmeier equation for a given material.

It is clear that the CE phase shift can be introduced in dispersive medium where the first derivative $dn/d\lambda \neq 0$. As a comparison, the pulse broadening is caused by $(d^2\beta/d\omega^2)|_{\omega_0} = ([\lambda_0^2/2\pi c][d^2n/d\lambda^2])|_{\lambda_0}$, which is related to the second derivative $d^2n/d\lambda^2$. For most optical

materials in the visible and near infrared region, $dn/d\lambda < 0$, thus $\varphi_{CE}(z) - \varphi_{CE}(z=0) > 0$.

For example, at the 800 nm central wavelength, $dn/d\lambda = -0.017288 \mu\text{m}^{-1}$ for fused silica, which is a commonly used window material for vacuum chambers in attosecond pulse generation setups. For roughly 58 μm of fused silica, the CE phase will shift by 2π rad. The CE phase shift is independent of the pulse duration. As a comparison, a 5 fs laser pulse will broaden to 5.13 fs when it passes through a 58 μm thick fused silica plate. For even longer pulses the relative broadening (the ratio of output pulse width to that of the input) is even smaller. In experiments, CE phase can be changed by using a pair of thin glass wedges. When one of the wedges is pulled in and out of the laser beam, the CE phase can be varied continuously.

3.2.2 Prism-Based Compressor

When chirped laser pulses are compressed with a pair of prisms, the change in prism separation affects the CE phase. The prism compressor is shown in Figure 3.4, where l is the distance between the apexes of the two prisms, and β is the angle between a ray with frequency ω and the reference ray that propagates from the first apex to the second one.

The CE phase shift expressed by Equation 3.12 can be manipulated to

$$\Delta\varphi_{CE} = \varphi_{CE}(z) - \varphi_{CE}(z=0) = \omega_0 \left(\frac{z}{v_g} - \frac{z}{v_p} \right) = \omega_0 t_g - \omega_0 t_p, \quad (3.23)$$

where t_g and t_p are the group time delay and phase time delay, respectively. If we assume that the input pulse is transform limited and the initial spectral phase is zero at ω_0 , then

$$\varphi(\omega_0) = -\omega_0 t_p \quad (3.24)$$

is the spectral phase of the output pulse. In such cases, the CE phase shift

$$\Delta\varphi_{CE} = \omega_0 t_g + \varphi(\omega_0), \quad (3.25)$$

which is valid when pulses propagate through any dispersive components, including prism pairs and grating pairs.

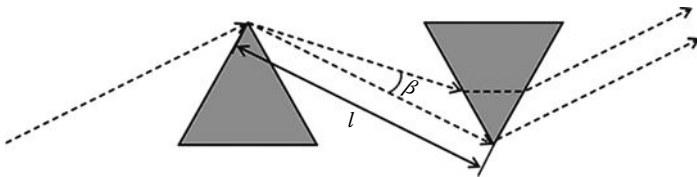


Figure 3.4 The prism compressor. l is the distance between the apexes of the prisms. The reference ray propagates from the apex of the first prism to that of the second prism. β is the angle between the ray with frequency ω and the reference ray. (Reprinted from Z. Chang, Carrier-envelope phase shift caused by grating-based stretchers and compressors, *Appl. Opt.*, 45, 8350, 2006. With permission of Optical Society of America.)

As discussed in Chapter 2, in a double-pass configuration, the spectral phase change introduced by the prism pair for the considered ray is

$$\varphi(\omega) = -2 \frac{\omega}{c} \cos \beta(\omega) l. \quad (3.26)$$

The group time delay

$$t_g = \left. \frac{d\varphi}{d\omega} \right|_{\omega_0}. \quad (3.27)$$

The CE phase shift therefore is

$$\Delta\varphi_{CE} = \omega_0 \left. \frac{d\varphi}{d\omega} \right|_{\omega_0} + \varphi(\omega_0) = 2 \frac{\omega_0^2}{c} \left. \frac{d\beta}{dn} \right|_{\omega_0} \left. \frac{dn}{d\omega} \right|_{\omega_0} \sin[\beta(\omega_0)] l, \quad (3.28)$$

where n is the index of refraction of the prism glass. For most prism compressors that are configured with minimum deviation and Brewster's angle incidence to avoid reflection loss, $d\beta/dn|_{\omega_0} = -2$. Replacing frequency with wavelength, we obtain the variation of the CE phase due to the change in prism separation:

$$\Delta\varphi_{CE} = 8\pi \sin[\beta(\lambda_0)] \left. \frac{dn}{d\lambda} \right|_{\lambda_0} \Delta l. \quad (3.29)$$

As a comparison, $GDD \approx -4l\lambda_0^3/2\pi c^2 (dn/d\lambda|_{\lambda_0})^2$. In this case, both the CE phase shift and pulse duration change are determined by the first derivative of the index of refraction. As a continuation of a previous example, for a fused silica glass prism $dn/d\lambda = -0.017288 \mu\text{m}^{-1}$ at 800 nm. The angle β is typically 10 mrad or less to avoid overfilling the second prism. For $\beta = 10$ mrad, a change of the separation by 1.45 mm introduces a 2π phase shift. Another method to change the CE phase is to introduce a different amount of material dispersion by translating the second prism in or out of the beam. In this case, the CE phase change can be calculated using Equation 3.22 where z is the length of the glass inserted.

The CE phase shift that occurs when a pulse travels through a grating pair is discussed later in this chapter.

3.3 Carrier-Envelope Phase in Laser Oscillators

A laser system that generates high-power, few-cycle lasers with stabilized CE phase is shown in Figure 3.1. For high repetition rate lasers, it is possible to use feedback control loops to stabilize the CE phase of the whole system within a certain bandwidth. We first discuss the locking of the CE phase changing rate of pulses from the oscillator that produces pulses at ~ 80 MHz repetition rate, and then study the compensation of the CE phase drift in kilohertz chirped pulse amplifiers. In such systems, it is not necessary to lock the CE phase of all the pulses from the oscillator to the same value. As long as the CE phase change rate is locked to the repetition rate, one can easily select pulses with the same CE phase to seed the amplifier.

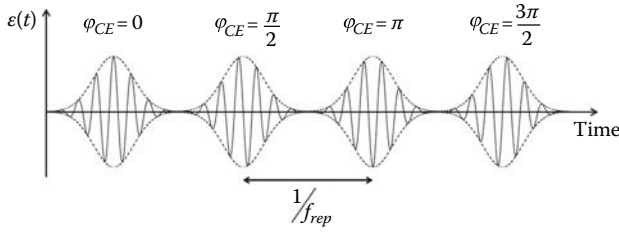


Figure 3.5 Four consecutive pulses with different CE phase values from an oscillator. (Adapted from E. Moon, PhD thesis, Kansas State University, Manhattan, KS, 2009.)

3.3.1 Carrier-Envelope Phase Offset Frequency

3.3.1.1 Carrier-Envelope Phase Change Rate

When a laser oscillator is mode locked, a femtosecond pulse travels back and forth in the cavity. Since the output coupler (OC) is partially transmissive, a portion of pulse energy leaves the cavity every time the pulse arrives there. Figure 3.5 shows the electric field for a section of a laser pulse train from a femtosecond oscillator. Here, f_{rep} is the repetition rate of the pulse train, which is the reciprocal of round trip time in the cavity, T_{rt} . The repetition rate of most femtosecond oscillators is on the order of 80 MHz. Although it is now possible to lock all CE phases of all the pulses to the same value, this is not necessary for the isolated attosecond pulse generation with amplified laser pulses. In general, it is easier to lock the CE phase change rate than locking the CE phase.

The repetition rate of the CPA amplifier is less than 1 MHz, typically between 1 and 20 kHz. As long as the change rate of the CE phase is stabilized, as is the case in Figure 3.5, pulses with identical CE phases can be selected to seed the amplifier. The CE phase in Figure 3.5 changes in steps of $\pi/2$ rad, making the rate of change

$$\frac{\Delta\varphi_{CE}}{T_{rt}} = \frac{\pi/2}{T_{rt}} = 2\pi \frac{f_{rep}}{4}. \quad (3.30)$$

Here, we consider φ_{CE} and $\varphi_{CE} + n2\pi$ being the same because the carrier wave field is a trigonometric function, where n is an integer. It is common to lock $\Delta\varphi_{CE}/2\pi T_{rt}$ to $f_{rep}/4 \approx 20$ MHz, which means that the CE phase of every fourth pulse in the train is the same. The pulse picker, located after the oscillator, operates at the rate equal to $f_{rep}/4n$. It will send the pulses with the same CE phase to the amplifier.

3.3.1.2 Carrier-Envelope Offset Frequency

When the CE phase change rate, $\Delta\varphi_{CE}/T_{rt}$, is kept as a constant, the electric field of the transform-limited pulse train at a given point can be expressed as follows:

$$\varepsilon_{PT}(t) = \sum_{j=-\infty}^{j=+\infty} E(t - jT_{rt}) e^{i[\omega_0(t-jT_{rt}) + j\Delta\varphi_{CE} + \varphi_{CE,0}]}, \quad (3.31)$$

where

j is the label of a particular pulse in the train

$\varphi_{CE,0}$ is the CE phase of the pulse labeled by $j=0$

$E(t)$ is the envelope function of a single pulse in the train

ω_0 is the carrier frequency of the pulse

We assume that the shape and width do not change from pulse to pulse.

In the frequency domain, the laser field can be obtained by performing Fourier transform to $\varepsilon_{PT}(t)$, which gives

$$\tilde{\varepsilon}_{PT}(\omega) = e^{i\varphi_{CE,0}} \sum_{j=-\infty}^{j=+\infty} e^{i[j(\Delta\varphi_{CE}-\omega_0 T_{rt})]} \int_{-\infty}^{\infty} E(t-jT_{rt}) e^{-i(\omega-\omega_0)t} dt. \quad (3.32)$$

The sum and the integral give

$$\tilde{\varepsilon}_{PT}(\omega) = e^{i\varphi_{CE,0}} \tilde{E}(\omega - \omega_0) \sum_{q=-\infty}^{q=+\infty} \delta(\omega T_{rt} - \Delta\varphi_{CE} - 2\pi q), \quad (3.33)$$

where

$$\tilde{E}(\omega) = \int_{-\infty}^{+\infty} E(t) e^{-i\omega t} dt \quad (3.34)$$

is the electric field of a single pulse in the train in the frequency domain. $|\tilde{E}(\omega)|^2$ is the power spectrum of a single pulse. When a grating spectrometer is used to measure the spectrum of the oscillator pulses, its resolution is not good enough to tell the difference between the spectrum of a single pulse and that of the pulse train, $|\tilde{\varepsilon}_{PT}(\omega)|^2$. In other words, we can consider the measured spectrum of the pulse train being the spectrum of a single pulse.

Equation 3.33 shows a frequency comb. The frequency of the q th comb tooth is given by

$$\omega_q = 2\pi \frac{q}{T_{rt}} + \frac{\Delta\varphi_{CE}}{T_{rt}}. \quad (3.35)$$

The comb is similar to the comb of the longitudinal mode discussed in Chapter 2, except for the shift introduced by the term $\Delta\varphi_{CE}/T_{rt}$. As just mentioned, the comb does not show up in a grating spectrometer because of its limited resolution.

Experimentally, frequency, instead of angular frequency, is measured. Equation 3.35 can be rewritten in terms of frequency

$$f_q = qf_{rep} + f_0, \quad (3.36)$$

where

$$f_0 = \frac{1}{2\pi} \frac{\Delta\varphi_{CE}}{T_{rt}} \quad (3.37)$$

is named the CE offset frequency. The frequency comb of a pulse laser is shown in [Figure 3.6](#). Due to limited bandwidth of the pulse, the number of teeth under the power spectrum is also finite.

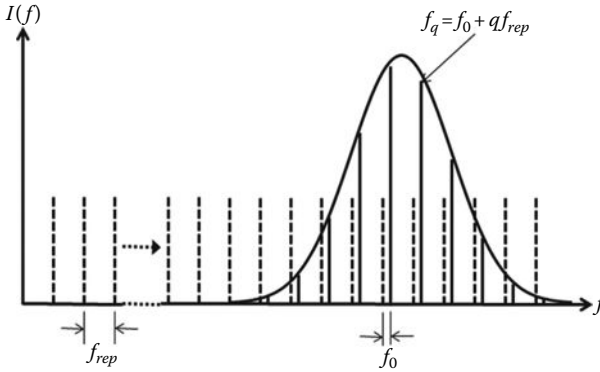


Figure 3.6 Frequency comb. The dashed lines represent the modes of the laser when the phase and group velocities of the pulse are equal. The solid lines represent the shifted modes of the laser underneath the gain spectrum in the presence of dispersion. (Adapted from E. Moon, H. Wang, S. Gilbertson, H. Mashiko, and Z. Chang: Advances in carrier-envelope phase stabilization of grating-based chirped-pulse lasers. *Laser Photon. Rev.* 2009. 4. 160. Copyright Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission.)

Another set of comb, which is the longitudinal mode of a cavity with no dispersion is also drawn as a reference. The spacing between the two adjacent teeth, f_{rep} , is the same for the two combs. The word “offset” infers the frequency offset between the two combs. For the pulse train in Figure 3.5, $\Delta\varphi_{CE}/2\pi T_r = f_{rep}/4$, the offset frequency $f_0 = f_{rep}/4$. This is the value chosen in most offset frequency stabilized oscillators.

If the changing rate varies with time, then the comb teeth position is not stable. Methods have been developed to lock the offset frequency to a preset value, in many cases to $f_{rep}/4$. The groups of Hänsch and Hall demonstrated the locking of both f_0 and f_{rep} in 2000 so that the optical comb can be used for optical frequency metrology. Both of them were awarded the Noble prize in physics in 2005. For metrology applications, the repetition rate (the cavity length) must also be stabilized, which is not a requirement for generating isolated attosecond pulses.

3.3.2 Stabilization of Offset Frequency

The CE offset frequency is stabilized by using feedback control techniques. The error signal is the change of f_0 over a certain period of time. However, historically it was a great challenge to find a method that can measure f_0 with high speed. It was only until the invention of photonic crystal fibers (PCFs), which made such measurement possible.

3.3.2.1 Measuring f_0 by f-to-2f Interferometers

Figure 3.7 displays the principle of f-to-2f interference. When the spectrum of the pulse train covers an octave, a low-frequency tooth $f_q = qf_{rep} + f_0$ is frequency doubled to $2f_q = 2(qf_{rep} + f_0)$, which interferes with a

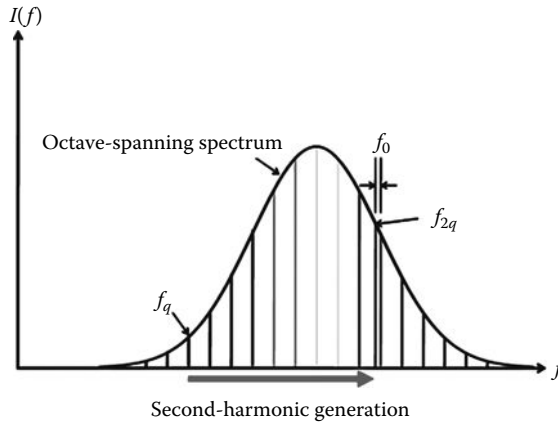


Figure 3.7 f -to- $2f$ self-referencing. (Adapted from E. Moon, H. Wang, S. Gilbertson, H. Mashiko, and Z. Chang: Advances in carrier-envelope phase stabilization of grating-based chirped-pulse lasers. *Laser Photon. Rev.* 2009. 4. 160. Copyright Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission.)

high-frequency tooth $f_{2q} = 2qf_{rep} + f_0$. The beat frequency from the inference is

$$2f_q - f_{2q} = 2(qf_{rep} + f_0) - (2qf_{rep} + f_0) = f_0. \quad (3.38)$$

Apparently, $f_0 < f_{rep}$, which is in the range of tens of megahertz for most oscillators used in CPA systems. It is easy to track f_0 using radio frequency (RF) spectrum analyzers. It is rather difficult to construct oscillators that can produce octave-spanning spectra. The broadening of a femtosecond laser spectrum to an octave in PCFs is straightforward.

The offset frequency is susceptible to pump power, temperature, pressure and, many other factors. For Ti:Sapphire laser oscillators using chirped mirrors for dispersion compensation, the offset frequency can be locked by feedback controlling the pump power.

3.4 Stabilization of the Carrier-Envelope Phase of Oscillators

3.4.1 Oscillator Configuration

The Kerr-lens mode-locked femtosecond laser oscillator in the author's lab is shown in [Figure 3.8](#), which is the modification of the product from Femtolasers Inc. The 5 W, 532 nm pump laser (Coherent Verdi) operates with a single longitudinal mode in its cavity. The pump laser beam is focused into the Ti:Sapphire crystal by a lens. The f_0 depends strongly on the pump laser power fluctuation. The noise of the single-longitudinal-mode laser is much better than the multimode one. This is the reason that most of the CE phase-stabilized oscillators are pumped by single-frequency laser.

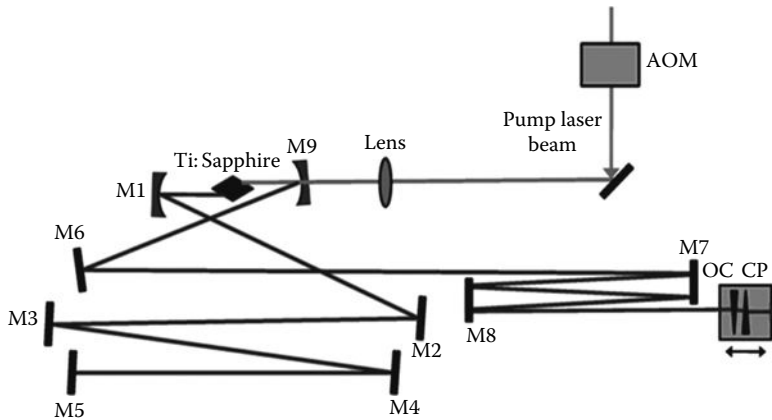


Figure 3.8 A femtosecond laser oscillator. M1–M9, cavity mirrors; AOM, acousto-optic modulator; OC, output coupler; CP, compensating plate. (Reprinted from E. Moon, PhD thesis, Kansas State University, Manhattan, KS, 2009.)

The Ti:Sapphire crystal is only about 2.3 mm long to reduce dispersions for generating short femtosecond pulses. It is cut at Brewster's angle to minimize reflection loss. The chirped cavity mirrors, M2–M8, compensate the positive chirp from the self-phase modulation and positive dispersion in the Ti:Sapphire crystal. The OC was cut at an angle of 10° to prevent reflections from the back surface into the cavity, as it would disturb the laser operation. The compensating plate (CP) was cut at the same angle and placed close to the OC in order to compensate the spatial chirp.

Once mode locked, the oscillator produces ~ 12 fs pulses with ~ 5 nJ of energy per pulse at a repetition rate f_{ref} of ~ 76 MHz. The temporal separation of pulses, T_{rt} , in the pulse train is ~ 13 ns, which can be conveniently observed by using a fast photodiode (1 ns response) and a fast oscilloscope (500 MHz bandwidth). The output average power is about 400 mW. The FWHM of the spectrum is ~ 100 nm.

An acousto-optic modulator (AOM) is added in the pump beam path, which is used to vary the pump power in order to lock the offset frequency. When the RF power fed into the acousto-optic crystal is varied, so is its optical transmission of the pump power. AOM is chosen because of its fast modulation time. The OC is mounted on a translation stage. It is used to shift the f_0 into the locking range by changing the amount of air dispersion in the cavity. Alternately, one can add a pair of glass wedges to tune f_0 , as is done in most phase-locked oscillators. f_0 can also be shifted to the locking range by controlling the temperature of the Ti:Sapphire crystal.

3.4.2 f -to- $2f$ Interferometer

3.4.2.1 White-Light Generation

The spectrum of the pulses from the oscillator shown in Figure 3.8 extends from 700 to 900 nm (100 nm FWHM), which does not span an octave

in frequency and thus is not wide enough to employ the f -to- $2f$ self-referencing method for detecting the offset frequency, f_0 . PCFs have been used to broaden the spectrum to extend from 500 to 1100 nm for covering an octave. Such fibers are discussed in Chapter 2. The nonlinear processes occurring inside the fiber preserve the comb structure of the incident laser beam so that the adjacent comb teeth are still separated by the laser repetition rate.

Difference frequency generation has also been used to measure f_0 , which needs an oscillator that is capable of generating much broader but not necessarily octave-spanning spectrum. Three teeth, $f_q = qf_{rep} + f_0$, $f_p = pf_{rep} + f_0$, and $f_r = rf_{rep} + f_0$ are involved, where the three integers satisfy $q - p = r$. The f_0 is obtained from the beating of the difference frequency signal $(f_q - f_p)$ generated in a nonlinear medium with the inferred signal at f_r , i.e., $(f_q - f_p) - f_r = f_0$. The advantage of this scheme is that the PCF is not required, which makes the alignment much easier. The CE phase locking time is also longer. However, the intensity on the Ti:Sapphire crystal is much higher to generate the broad spectrum through self-phase modulation, which often causes damages to the crystal.

3.4.2.2 Setup

The optical layout designed to obtain the offset frequency of the oscillator in Figure 3.8 is shown in Figure 3.9. The output beam from the laser oscillator was spectrally broadened to cover an octave in the PCF. When the white light from the fiber enters the interferometer, the long-wavelength portion and the short-wavelength portion are split by a dichroic beam splitter.

The long-wavelength components (1064 nm) were focused into the 5 mm long PPKTP (periodically poled potassium titanyl phosphate) crystal for generating the second harmonic light. The PPKTP crystal was used because of its high-conversion efficiency. The short-wavelength (around 532 nm) portion of the light is passed through a delay stage.

At the output of the interferometer, which is the first polarization beam splitter (PBS), the second harmonic of the 1064 nm beam and the 532 nm beam from the short-wavelength arm are spatially and temporally overlapped. The time overlap is accomplished by adjusting a delay. Due to the type I phase matching (oo-e) in the PPKTP, the second harmonic light is orthogonally polarized to the 530 nm light from the other arm. A half-wave plate at 532 nm is placed before a second PBS to balance the signals from the two arms and after the polarizing beam splitter to control their beating.

A beat signal with frequency f_0 is obtained by selecting only the wavelength components from the fundamental green light, which spectrally overlap with the frequency-doubled infrared components. The selection was accomplished by using a slit in the combined beam angularly dispersed by a grating. It is further purified by an interference filter with a 2 nm FWHM bandpass centered at 532 nm. An avalanche photodiode (APD) is used to detect the weak light signal transmitted through the slit and the filter. The number of comb teeth contributing to the beat signal is $\sim 28,000$.

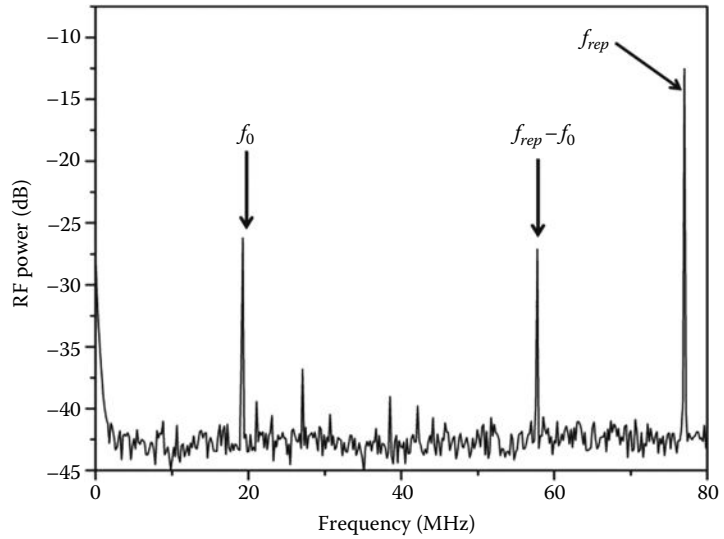


Figure 3.10 Beat signal shown on an RF spectral analyzer. (Reprinted from E. Moon, PhD thesis, Kansas State University, Manhattan, KS, 2009.)

frequency, f_0 , is sent to the other input of the phase detector. The frequency difference between the two frequencies, $f_{rep} - f_0$, is measured in terms of the time-dependent phase, which equals $(f_{rep} - f_0)t$. The phase detector measures the phase difference between the two signals to produce an error signal that is sent to the input of the proportional integral controller. The output of the proportional integral controller adjusts the acousto-optical modulator that controls the pump power to minimize the error signal, which leads to $f_0 = f_{rep}/4$.

3.4.3.2 Stability of the Locked f_0

In reality, the value of the locked f_0 does not exactly equal $f_{rep}/4$. The quality of the locking can be evaluated by measuring the width of the offset frequency with a high-resolution RF spectrum analyzer. An example of the measured linewidth of the locked f_0 is shown in Figure 3.11. It shows that the offset frequency could be locked within a 100 mHz. The criterion for CE phase locking is that the offset frequency should be locked to less than 1 Hz because that would correspond to a 1 rad phase shift in 1 s. It is worthy to point that the measurement is done inside the locking loop, which is not a true measure of the f_0 stability. The true f_0 stability is worse than the value given here.

The locking noise, as a function of frequency, is measured by using a dynamic signal analyzer. The power spectrum density (PSD) can be expressed by

$$S_{\varphi_{CE}}(f) = \lim_{T \rightarrow \infty} \frac{1}{T} \left| \int_{-\frac{T}{2}}^{\frac{T}{2}} \varphi_{CE}(t) e^{i2\pi ft} dt \right|^2, \quad (3.39)$$

where T is the observation time. The measured value is twice the value given in Equation 3.39. An example of the measured spectrum is shown in

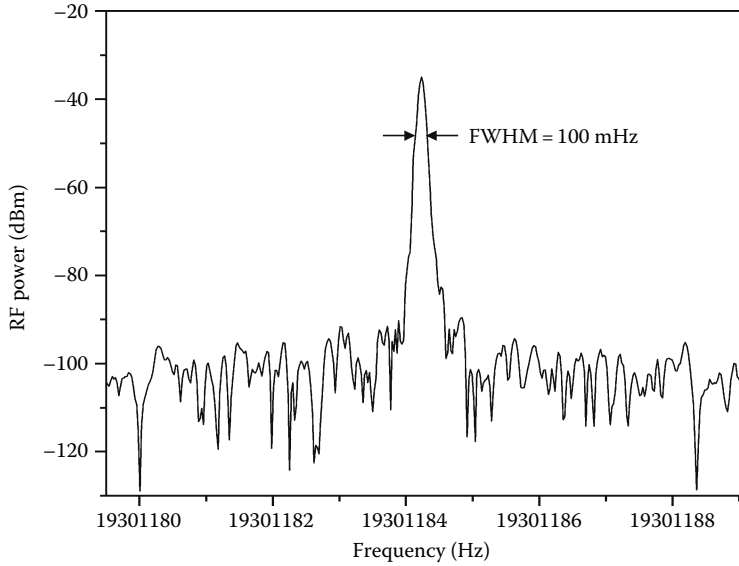


Figure 3.11 Linewidth of the offset frequency when the phase-locking loop was engaged. (Reprinted from E. Moon, PhD thesis, Kansas State University, Manhattan, KS, 2009.)

Figure 3.12. The phase error integrated over the frequency range that the measurement covers is also shown in Figure 3.12. Researchers in the field define the time of the CE phase locking to be the observation time at which the accumulated phase error is ~ 1 rad.

3.4.4 Noise of the Interferometer

3.4.4.1 Error in Measuring f_0

In a free running f -to- $2f$ interferometer, the optical path length difference ΔL in Figure 3.9 may change with time due to vibrations of the mirror mounts. As a result, the relative phase of the beams in the two arms of the interferometer becomes time dependent, which can be expressed by

$$\phi(t) = 2\pi \frac{\Delta L(t)}{\lambda}, \quad (3.40)$$

where $\lambda = 532$ nm.

We chose the short-wavelength arm of the interferometer as the reference. The electric field of the beam in this arm is given by

$$\varepsilon_1(t) = E_1(t) \cos[2\pi(qf_{rep} + f_0)t], \quad (3.41)$$

where

E_1 is the amplitude

q is the comb tooth index

The electric field of the pulse in the second harmonic arm of the interferometer is given by

$$\varepsilon_2(t) = E_2(t) \cos[2\pi(qf_{rep} + 2f_0)t + \phi(t)]. \quad (3.42)$$

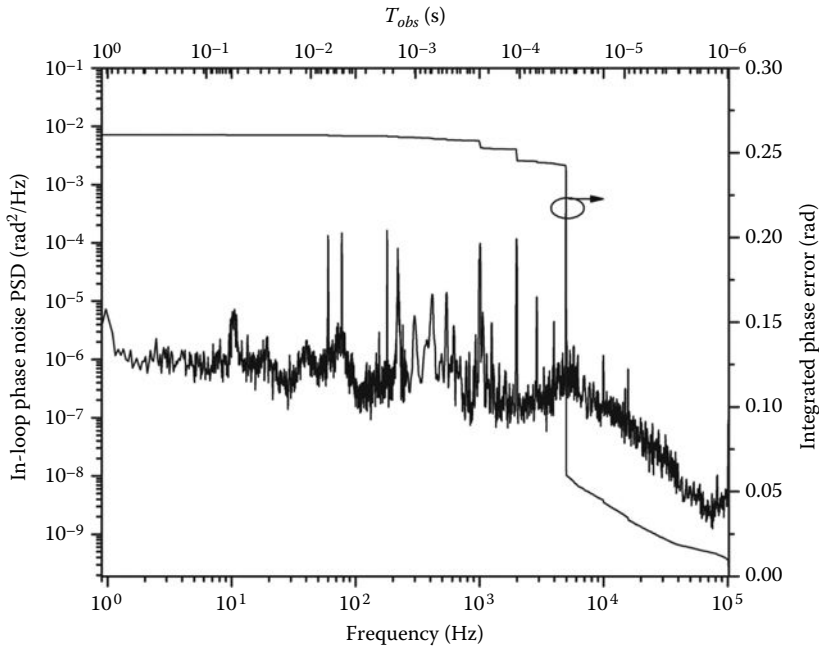


Figure 3.12 PSD of the phase detector signal and integrated phase error. (Reprinted from E. Moon, PhD thesis, Kansas State University, Manhattan, KS, 2009.)

The frequency of the detected beat signal will be deviated from its true value and will also become time dependent:

$$f(t) = f_0 + \frac{1}{2\pi} \frac{d\phi}{dt}. \quad (3.43)$$

What we want to stabilize is the offset frequency f_0 . However, the locking electronics stabilize f , without knowing the systematic error introduced by the interferometer instability. The interferometer can be locked to minimize $d\phi/dt$.

3.4.4.2 Interferometer Locking

The interferometer can be stabilized using the setup shown in [Figure 3.9](#). A Helium–Neon (HeNe) laser, operating at ~ 632 nm, co-propagates with the white light from the PCF in the f -to- $2f$ interferometer. After the HeNe beams traversed the interferometer, they were bounced off the grating and sent to two detectors. A photodiode measured the intensity of a single fringe of the interference pattern. The single fringe is obtained by using two cylindrical mirrors and a slit. The signal from the photodiode is sent to a proportional–integral–derivative (PID) controller. The PID controller provided an output voltage, which was applied to a piezoelectric transducer (PZT) attached to the mirror in the top arm of the interferometer to stabilize the path length difference.

An example of the fringes obtained from the CCD camera for cases when the interferometer is locked and unlocked is shown in [Figure 3.13](#). The jitter of fringes is easy to see when interferometer is not locked.

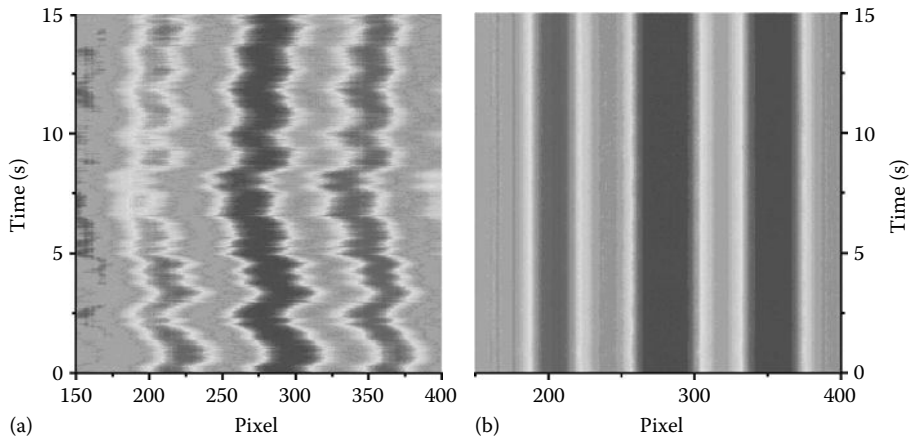


Figure 3.13 HeNe fringes: (a) unlocked and (b) locked. (Reprinted from E. Moon, C. Li, Z. Duan, J. Tackett, K.L. Corwin, B.R. Washburn, and Z. Chang, Reduction of fast carrier-envelope phase jitter in femtosecond laser amplifiers, *Opt. Express*, 14, 9758, 2006. With permission of Optical Society of America.)

3.4.4.3 Noise Spectrum

The signal from the HeNe photodiode is sent to a dynamic signal analyzer to measure the PSD. The results are shown in [Figure 3.14](#).

In the figure, the majority of the noise is present from DC to ~ 1 kHz. When the locking servo is engaged, the noise is significantly reduced within that range. The dominating noise peak at 100 Hz was reduced by almost 2 orders of magnitude. It should be noted that the optical table was not floated when the top portion of Figure 3.14 was measured.

The effect of floating and unfloating the table is shown in the bottom portion of Figure 3.14. In the measurement, the interferometer locking servo was not engaged. When the table is floated, the high-frequency (>1 kHz) noise of the interferometer is reduced by almost 2 orders of magnitude. The low-frequency noise in the range of ~ 500 mHz–100 Hz is reduced by 1 order of magnitude to 2 orders of magnitude. These results conclude that the noise mainly originated from vibrations of the optical components; thus, the optical table should be floated and the f -to- $2f$ interferometers should be locked to stabilize the CE phase of femtosecond oscillators.

3.5 Measurement of the Carrier-Envelope Phase of Amplified Pulses

The repetition rate of femtosecond oscillator is extremely high, ~ 80 MHz, which makes it almost impossible to measure the CE phase of each individual pulses. The f -to- $2f$ interferometry introduced in [Section 3.4](#) measures CE offset frequency, which is the change rate of the CE phase, not the CE phase itself.

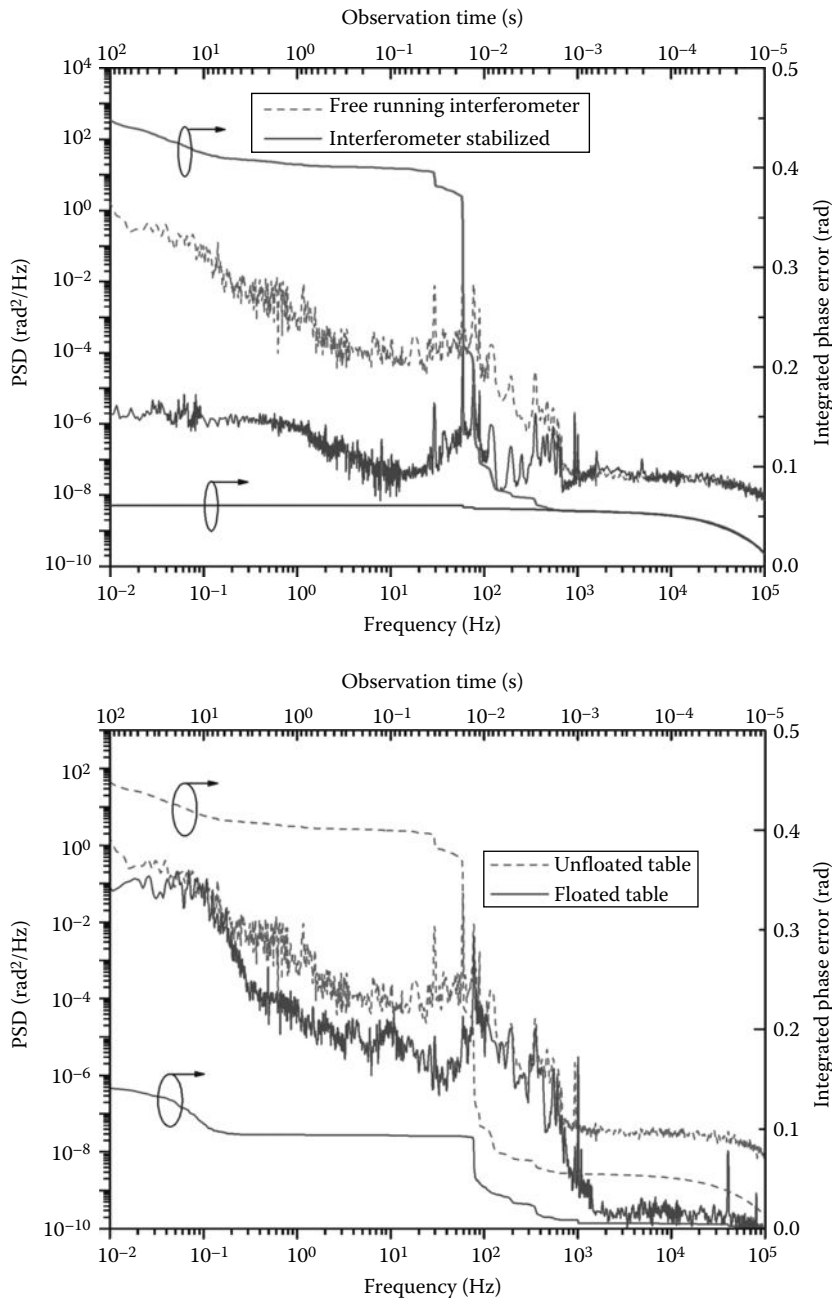


Figure 3.14 Top: The power spectrum of the interferometer phase noise and the integrated phase error. Bottom: Comparison of phase noise measurements when the optical table is floated and unfloated. (Reprinted from E. Moon, C. Li, Z. Duan, J. Tackett, K.L. Corwin, B.R. Washburn, and Z. Chang, Reduction of fast carrier-envelope phase jitter in femtosecond laser amplifiers, *Opt. Express*, 14, 9758, 2006. With permission of Optical Society of America.)

Most of the femtosecond CPA amplifiers for generating attosecond pulses operate at 1 kHz, which is slow enough for measuring the CE phase of each pulse. The principle of the f -to- $2f$ interferometry has been successfully applied to the measurement of the CE phase of the amplified pulse. Alternatively, since the field of the focused amplified beam is very strong, one can also use other physics effects such as ATI and high-harmonic generation to measure the CE phase.

3.5.1 Single Shot f -to- $2f$ Interferometry

3.5.1.1 Interferometer Setup

The spectrum of each femtosecond pulse from the amplifier is a continuous one. Unlike the pulse train from the oscillator, there is no comb structure. Strictly speaking, there is a frequency comb, but the spacing between the teeth is 1 kHz, which is not resolvable with conventional grating spectrometers. The width of a 30 fs pulse extends from 750 to 850 nm, which does not cover an octave. Consequently, f -to- $2f$ measurement cannot be done directly with the pulses from the CPA amplifiers.

Instead of using PCFs, bulk materials can be used to broaden the spectrum, which are much easier to align. An example of the setup in the author's lab is shown in Figure 3.15. A small portion of the amplified laser pulse train ($<1 \mu\text{J}$) is sent into the interferometer. The laser beam is focused into the 2.3 mm thick sapphire plate by a lens. A neutral density filter (NDF) is used to adjust the pulse energy. Sapphire is chosen because of large n_2 and high-damage threshold.

The laser peak power is higher than the critical power of the sapphire plate. As a result, a single filament is formed inside the sapphire plate.

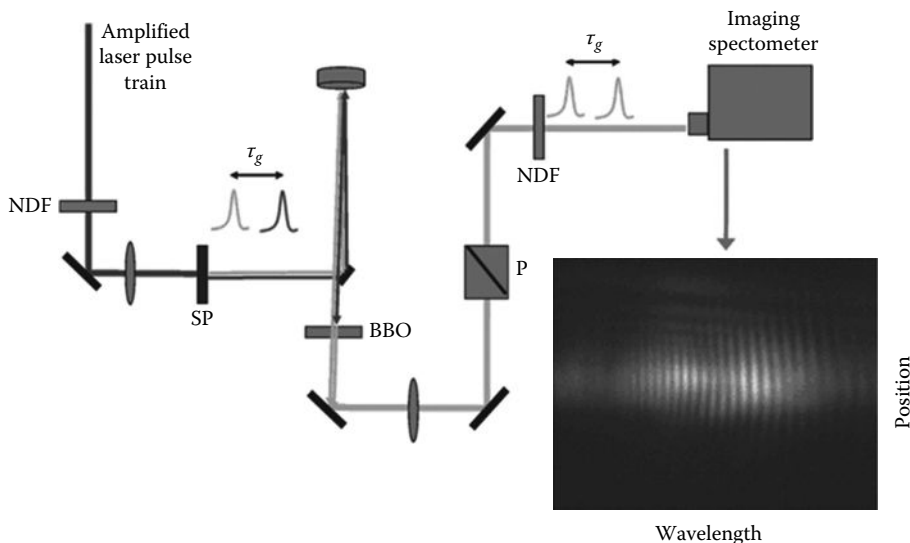


Figure 3.15 Single-shot f -to- $2f$ setup: NDF, neutral density filter; SP, sapphire plate; BBO, SHG-crystal; P, polarizer; τ_g , delay between the f and $2f$ pulses. (Reprinted from E. Moon, PhD thesis, Kansas State University, Manhattan, KS, 2009.)

Inside the filament the laser intensity is very high. The pulses undergo self-phase modulation and are broadened into white light covering an octave in the spectrum, which is necessary for the f and $2f$ components to interfere. For Ti:Sapphire lasers, it is a common practice to choose the f component centered at 1000 nm and $2f$ at 500 nm. We can imagine that the white-light pulse consists of two pulses, one centering at 1000 nm and other at 500 nm. There is time delay, τ_g , between the two due to the group velocity dispersion.

The white light is then focused into a 1 mm thick BBO crystal for converting the 1000 nm light into 500 nm. The polarization of the second harmonic pulse is perpendicular to that of the 500 nm light from the sapphire plate due to the type I phase matching in the BBO. Both the original 500 nm light and the second harmonic of 1000 nm light are sent to a spectrometer after passing through a polarizer used to select a common polarization for them to interfere. There is a spectrum phase difference between the two 500 nm pulses caused by the time delay, τ_g , between them, which leads to spectrum modulations with a period τ_g . The value of τ_g is a few hundreds of femtoseconds. The image in [Figure 3.15](#) shows a typical f -to- $2f$ interferogram, from which the CE phase can be extracted.

3.5.1.2 Fourier Transform Spectral Interferometry

In the frequency domain, the electric fields of the f and $2f$ pulses can be expressed as follows:

$$\tilde{E}_{WL}(\omega) \propto \sqrt{I_{WL}(\omega)} e^{i[\varphi_{WL}(\omega) + \varphi_{CE}]}, \quad (3.44)$$

$$\tilde{E}_{SH}(\omega) \propto \sqrt{I_{SH}(\omega)} e^{i[\varphi_{SH}(\omega) + 2\varphi_{CE} + \omega\tau_g]}, \quad (3.45)$$

where $I_{WL}(\omega)$ and $I_{SH}(\omega)$ are the intensities of the 500 nm pulse from the white light directly and the second harmonic of the 1000 nm, respectively. The bandwidth of the second harmonic pulse is typically tens of nanometer, determined by the phase-matching bandwidth of the BBO crystal. $\varphi_{WL}(\omega)$ and $\varphi_{SH}(\omega)$ are their spectral phases, respectively. τ_g is the time by which f pulse leads the $2f$ pulse due to the dispersion in the sapphire plate.

A lineout of the interference pattern between the f and $2f$ pulses can be expressed as

$$\begin{aligned} S(\omega) &\propto |\tilde{E}_{WL}(\omega) + \tilde{E}_{SH}(\omega)|^2 \\ &= I_{WL}(\omega) + I_{SH}(\omega) \\ &\quad + 2\sqrt{I_{WL}(\omega)I_{SH}(\omega)} \cos[\omega\tau_g + \varphi_{SH}(\omega) - \varphi_{WL}(\omega) + \varphi_{CE}]. \end{aligned} \quad (3.46)$$

The CE phase, φ_{CE} , contained in the interference term, can be extracted by using an algorithm called Fourier transform spectral interferometry (FTSI).

First, the contribution of the interference term in Equation 3.46 is separated from the two DC terms, $I_{WL}(\omega)$ and $I_{SH}(\omega)$, by taking the inverse Fourier transform of the spectral interferometry signal. The result is shown in [Figure 3.16](#). The two sidebands are from the interference term. Only one of them is filtered out for extracting the CE phase.

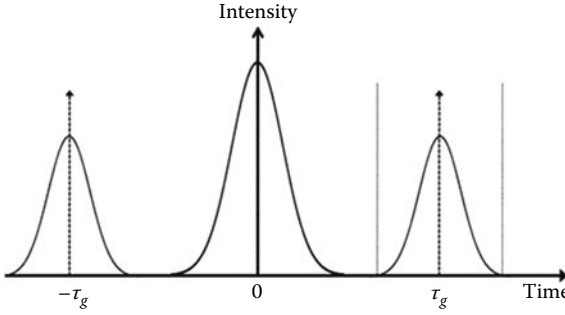


Figure 3.16 A Fourier transform peak from the spectral interferometry signal.

Second, a Fourier transform is applied to the positive delay peak. The phase from the transform can be written as follows:

$$\Phi(\omega) = \omega\tau_g + \varphi_{SH}(\omega) - \varphi_{WL}(\omega) + \varphi_{CE} = \omega\tau_g + \varphi_{CE} + \delta\varphi(\omega). \quad (3.47)$$

where $\delta\varphi(\omega) = \varphi_{SH}(\omega) - \varphi_{WL}(\omega)$. If the shot to shot variation of $\delta\varphi(\omega)$ and delay τ_g is negligible, the change of CE phase, $\Delta\varphi_{CE}$, can be determined from the measured shift of the total phase, $\Delta\Phi(\omega)$, between two measurements, i.e.,

$$\Delta\varphi_{CE} = \Delta\Phi(\omega). \quad (3.48)$$

The resultant $\Delta\Phi(\omega)$ can serve as the error signal for stabilizing the CE phase of the amplifier through feedback control.

3.5.2 Precisions of the Carrier-Envelope Phase Measurement

The accuracy of the f -to- $2f$ measurements of the CE phase can be affected by many factors. Fluctuation of the laser pulse energy is a major factor. The energy of the laser pulses from CPA systems, ε , changes from shot to shot due to the pump laser fluctuation and other reasons. The relative energy fluctuation, $\Delta\varepsilon/\varepsilon$, is on the order of 1% for kilohertz lasers equipped with diode-pumped solid-state pump lasers. As a result, $\delta\varphi(\omega)$ and delay τ_g also vary from shot to shot, which affects the precision of the $\Delta\varphi_{CE}$ measurement.

3.5.2.1 Experimental Determination of the Carrier-Envelope Phase–Energy Coupling Coefficient

The coupling between the error of the CE phase measurement, $\Delta\varphi_{CE}$, and the relative laser energy change, $\Delta\varepsilon/\varepsilon$, can be expressed by the coefficient

$$C_{PE} = \frac{\Delta\varphi_{CE}}{(\Delta\varepsilon/\varepsilon)}, \quad (3.49)$$

which has been investigated experimentally in the author's lab. The setup is shown in [Figure 3.17](#).

Two f -to- $2f$ interferometers were used in the experiment: one of them was inside the CE phase stabilization loop, whereas the other was out of

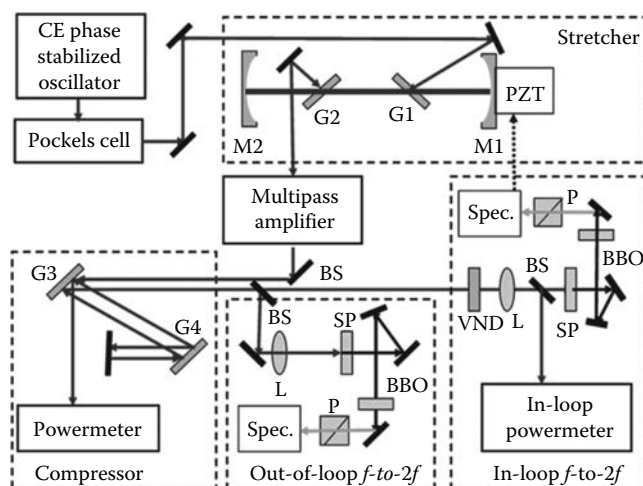


Figure 3.17 Setup for measuring the CE phase–energy coupling coefficient. (Reprinted from C. Li, E. Moon, H. Wang, H. Mashiko, C.M. Nakamura, J. Tackett, and Z. Chang, Determining the phase–energy coupling coefficient in carrier-envelope phase measurements, *Opt. Lett.*, 32, 796, 2007. With permission of Optical Society of America.)

the loop, which was used to independently measure the CE phase drift. A variable NDF before the sapphire plate in the in-loop was used to change the laser energy in a triangle fashion to introduce $\Delta\epsilon/\epsilon$, as shown in Figure 3.18a. The laser power in the out-of-loop interferometer was kept constant. The difference between the φ_{CE} values given by the two interferometers is the $\Delta\varphi_{CE}$.

The in-loop f -to- $2f$ was used to lock the total phase $\Phi(\omega)$. As the laser energy was changed between 0.3 and 0.4 μJ in the in-loop f -to- $2f$, it would measure the $\Phi(\omega)$ drift and lock the phase of the laser to whatever value its measurement shows. As a result, the CE phase displayed by the in-loop f -to- $2f$ is a constant as shown in Figure 3.18c assuming $\varphi_{CE} = \Phi(\omega)$, although the real CE phase may not be stabilized. The CE phase value given by the out-of-loop interferometer is the true CE phase that varies with the laser energy in the in-loop f -to- $2f$, as shown in Figure 3.18b. The results clearly show how the measured out-of-loop phase modulated as the energy in the in-loop was modulated.

The CE phase measured by the out-of-loop interferometer as a function of pulse energy in the in-loop interferometer is shown in Figure 3.19a. The CE phase–energy coupling coefficient can be obtained from the slope of a line fitted to the measured data. The slope in this figure corresponded to a coupling coefficient value of $C_{PE} = 160$ mrad per 1% change in laser energy. It is worthy to point that the coefficient changes with the laser parameters and the setup; thus, the number given here should be used with caution. The energy stability of a typical kilohertz Ti:Sapphire oscillator is 1% or more, which may introduce a CE phase measurement error. The stability of many CE phase–stabilized lasers is in the range of 100–200 mrad; thus, the energy stability of the amplified laser pulses is key to the quality of the phase stabilization.

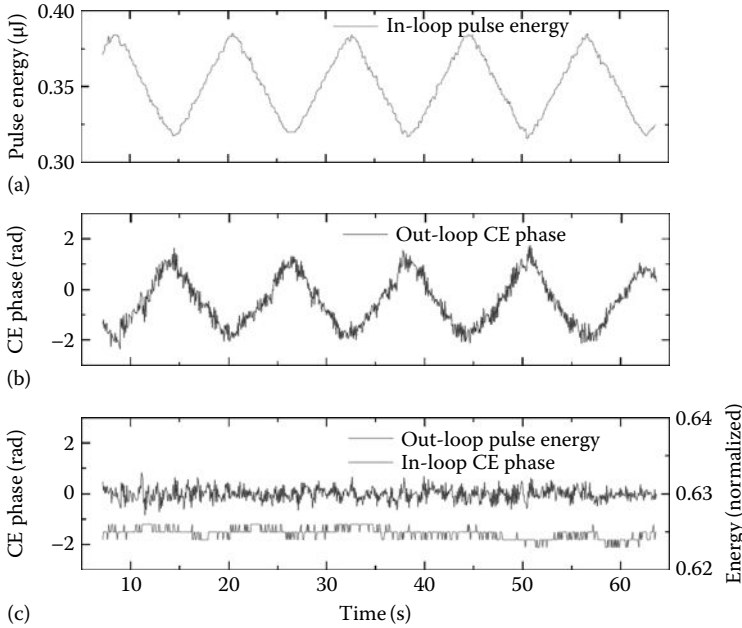


Figure 3.18 Measured phases by two interferometers when the laser energy in the in-loop interferometers is modulated: (a) modulated in-loop pulse energy, (b) measured out-of-loop phase, and (c) the in-loop phase and out-of-loop pulse energy. (Reprinted from C. Li, E. Moon, H. Wang, H. Mashiko, C.M. Nakamura, J. Tackett, and Z. Chang, Determining the phase-energy coupling coefficient in carrier-envelope phase measurements, *Opt. Lett.*, 32, 796, 2007. With permission of Optical Society of America.)

3.5.2.2 Explanation of the Carrier-Envelope Phase–Energy Coupling

The CE phase measurement error introduced by the energy fluctuation can be explained by considering the measured total phase $\Phi(\omega) = \omega\tau_g + \varphi_{CE} + \delta\varphi(\omega)$ in Equation 3.47. For the CE phase stabilization procedure, it is the total phase that is directly measured and stabilized. However, the terms in the total phase, except for the CE phase φ_{CE} , are energy dependent.

The real CE phase shift, then, is given by

$$\Delta\varphi_{CE} = \Delta\Phi(\omega) - \Delta[\varphi_{SH}(\omega) - \varphi_{WL}(\omega) + \omega\tau_g], \quad (3.50)$$

where $\Delta\Phi(\omega)$ is the change of the total phase and is set to zero when the CE phase–stabilization procedure is implemented.

Equation 3.47 can be rewritten as

$$\Phi(\omega) = \omega\tau_g + [\varphi_{SH}(\omega) - \varphi_{WL}(\omega) + \varphi_{CE}]. \quad (3.51)$$

It is found experimentally that the $\Phi(\omega)$ versus ω plot is close to a straight line. The delay time in the results given in Figure 3.19b was determined by the slope. The intercept of the line to vertical axis shown in Figure 3.19c originates from the term $\varphi_{SH}(\omega) - \varphi_{WL}(\omega) + \varphi_{CE}$. It is evident that both the delay and the intercept are functions of the laser energy, which cause the error in the CE phase measurements.

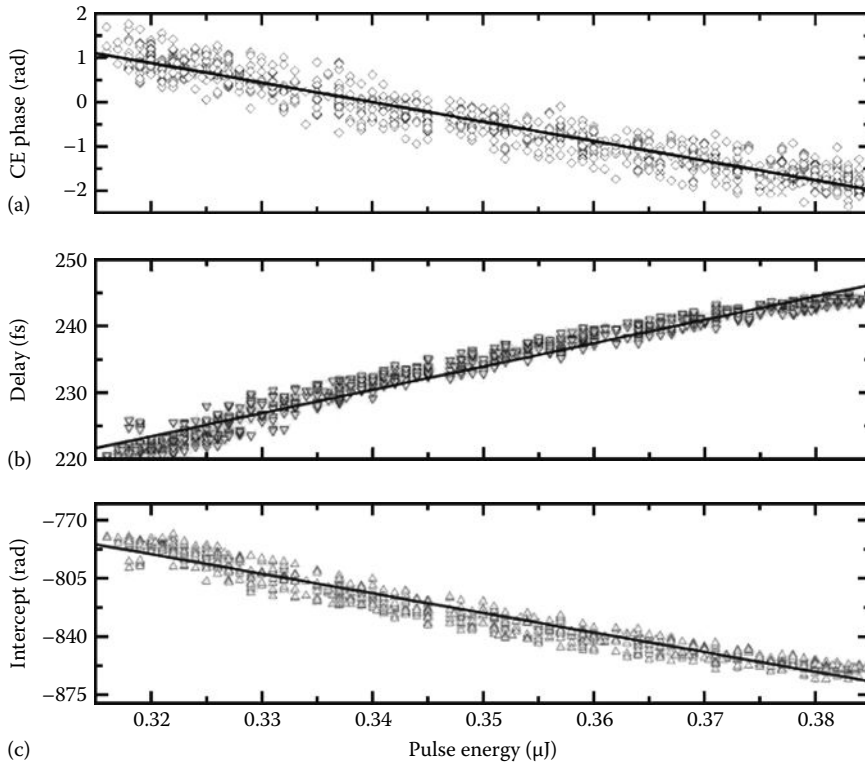


Figure 3.19 Experimental results versus the laser energy: (a) relative CE phase, (b) delay time, and (c) the residual intercept after the subtraction of the CE phase. (Reprinted from C. Li, E. Moon, H. Wang, H. Mashiko, C.M. Nakamura, J. Tackett, and Z. Chang, Determining the phase-energy coupling coefficient in carrier-envelope phase measurements, *Opt. Lett.*, 32, 796, 2007. With permission of Optical Society of America.)

3.5.3 Two-Step Model*

A simple two-step model has been proposed to explain the coupling between the laser energy fluctuation and the measured CE phase shift, i.e., to quantitatively evaluate the dependence of the quantity $\Delta[\varphi_{SH}(\omega) - \varphi_{WL}(\omega) + \omega\tau_g]$ on laser energy. Figure 3.20 shows the basic premise of the model.

3.5.3.1 Filamentation in Sapphire Plate

In the f -to- $2f$ interferometer considered here, a femtosecond laser beam with diameter D propagating along the z direction is focused on a sapphire plate by a lens with focal length f . The focal spot radius at the input of the sapphire plate is given by $w_0 = \lambda_0(f/D)$. Here, we assume the laser center wavelength $\lambda_0 = 0.79 \mu\text{m}$.

* This section is adapted from Li, C., E. Moon, H. Mashiko, H. Wang, C.M. Nakamura, J. Tackett, and Z. Chang, Mechanism of phase-energy coupling in f -to- $2f$ interferometry, *Appl. Opt.* 48, 1303 (2009).

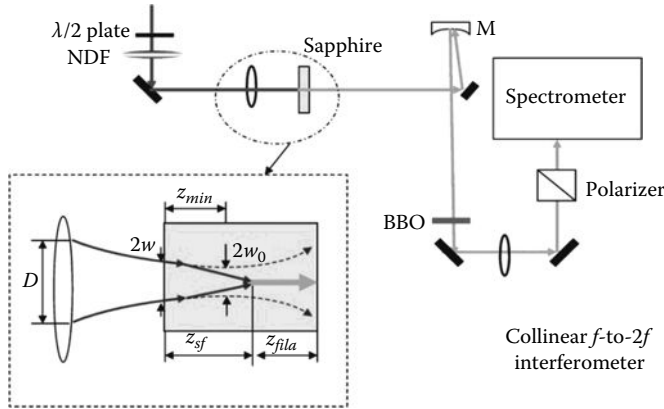


Figure 3.20 Collinear f -to- $2f$ interferometer with a sapphire plate. Inset: formation of a single filament by self-focusing. D , diameter of the laser beam; f , focal length of the focusing lens; w_0 , radius of the focal spot; z_{fil} , length of the filament. (Adapted from C. Li, E. Moon, H. Mashiko, H. Wang, C.M. Nakamura, J. Tackett, and Z. Chang, Mechanism of phase-energy coupling in f -to- $2f$ interferometry, *Appl. Opt.*, 48, 1303, 2009 With permission of Optical Society of America.)

As mentioned in Chapter 2, when the laser peak power is higher than the critical power, $P_c = \pi(0.61)^2 \lambda_0^2 / 8(n_0 n_2)$, a filament is formed inside the sapphire plate. The filament is the balance between the convergence as a result of Kerr-lens effect and the divergence due to the diffraction of the small beam at the focus as well as the plasma-lens effect. The linear and nonlinear indices of refraction of the sapphire plate are $n_0 = 1.76$ and $n_2 \approx 2.9 \times 10^{-16} \text{ cm}^2/\text{W}$ at the laser center wavelength, respectively, which yields a critical power $P_c = 1.79 \text{ MW}$. Actually, sapphire is a birefringence material. A half-wave plate sets the polarization direction to where n_2 is maximized in the experiments.

We choose the input surface of the sapphire plate as $z = 0$. Suppose the filament starts at the location $z = z_{sf}$, then it has been found that the self-focusing distance as a function of laser energy can be expressed as follows:

$$z_{sf}(\mathcal{E}) = \frac{2n_0 w_0^2}{\lambda_0} \frac{1}{\sqrt{\frac{P}{P_c} - 1}}, \quad (3.52)$$

where

the peak power $P = \mathcal{E}/\tau_p$

τ_p is the laser pulse duration at the input of the sapphire plate

$\mathcal{E}$ is the laser pulse energy

In the model, the pulse duration is 35 fs, the input beam diameter is $D = 5 \text{ mm}$, and the focal length is $f = 75 \text{ mm}$. The calculated self-focusing distance is shown in Figure 3.21, which decreases with increasing pulse energy.

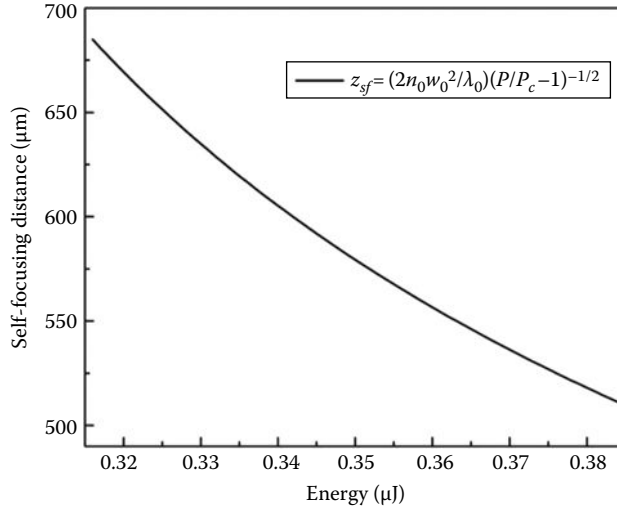


Figure 3.21 Calculated dependence of the self-focusing distance on the pulse energy. (Adapted from C. Li, E. Moon, H. Mashiko, H. Wang, C.M. Nakamura, J. Tackett, and Z. Chang, Mechanism of phase-energy coupling in f -to- $2f$ interferometry, *Appl. Opt.*, 48, 1303, 2009 With permission of Optical Society of America.)

The filament length is

$$z_{\text{fila}}(\varepsilon) = L - z_{\text{sf}}(\varepsilon), \quad (3.53)$$

where L is the thickness of the sapphire plate, which is 2.3 mm thick in the model.

3.5.3.2 White-Light Generation

As the beam contracts to a filament, self-phase modulation occurs along with other nonlinear processes such as plasma formation. Those processes broaden the spectrum of the input laser pulse so that it covers an octave at the exit of the sapphire plate for the f -to- $2f$ measurements. In the model, the FWHM spectral width of the input laser is $\Delta\lambda_0 \approx 35$ nm, which is typical for Ti:Sapphire CPA lasers. The spectral broadening can be estimated as $\Delta\lambda \approx \phi_{\text{spm}} \Delta\lambda_0$, where ϕ_{spm} is the nonlinear phase introduced by the self-phase modulation as explained in Chapter 2. The maximum nonlinear phase shift is given by

$$\begin{aligned} \phi_{\text{spm}} &= - \int_0^{z_{\text{sf}}} \frac{2\pi}{\lambda_0} n_2 I(z) dz = - \int_0^{z_{\text{sf}}} \frac{2\pi}{\lambda_0} n_2 \frac{2\varepsilon}{\tau_p \pi w^2(z)} dz \\ &\approx - \frac{2\pi^2}{\lambda_0^2} \frac{n_2 \varepsilon}{\tau_p}. \end{aligned} \quad (3.54)$$

Using the parameters in the model, the nonlinear phase shift is $\phi_{\text{spm}} \approx 10$ rad, which would broaden the spectrum from 35 to 350 nm FWHM. The wings of such a broad spectrum would cover an octave.

3.5.3.3 Frequency Phase of White Light, Nonlinear Phase, and Carrier-Envelope Phase

Accounting for linear and nonlinear dispersions, the spectral phases of the green (500 nm, the f pulse) and IR (1000 nm, for generating the $2f$ pulse) pulses at the start of the filament are given by

$$\varphi_{G,sf}(\omega) = \varphi_{CE} + \Delta\varphi_{n_0} + \Delta\varphi_{n_2} + \phi_{spm} \approx \phi_{G,sf}(\omega_G), \quad (3.55)$$

$$\varphi_{IR,sf}(\omega) = \varphi_{CE} + \Delta\varphi_{n_0} + \Delta\varphi_{n_2} + \phi_{spm} \approx \phi_{IR,sf}(\omega_{IR}), \quad (3.56)$$

where

ω_{IR} and $2\omega_G = 2\omega_{IR}$ are the center angular frequencies of the IR and green pulses, respectively

φ_{CE} is the CE phase at the input of the sapphire plate

$\Delta\varphi_{n_0}$ is the CE phase shift caused by linear dispersion, which is equal to $\omega_0\Delta\tau_0$

where $\Delta\tau_0$ is the difference between the group and phase delay at the input carrier frequency ω_0

The nonlinear contribution to the CE phase shift is given by

$$\Delta\varphi_{n_2} = -\phi_{spm}\omega_0 \left. \frac{dn_2}{d\omega} \right|_{\omega_0} \frac{1}{n_2}, \quad (3.57)$$

where $\omega_0(dn_2/d\omega)|_{\omega_0} = 8 \times 10^{-17} \text{ cm}^2/\text{W}$, $n_2 \approx 2.9 \times 10^{-16} \text{ cm}^2/\text{W}$. Thus, $\Delta\varphi_{n_2} \approx -0.28\phi_{spm}$.

It is assumed that the spectral phase difference between the green pulse and the IR pulse is only affected by the linear dispersion during the propagation in the filament. The justification is that the laser peak power decreases as the pulse duration increases during the propagation, making the nonlinear contributions negligible. Then, including the linear dispersion in the filament, the spectral phases become

$$\varphi_G(\omega) = \varphi_{G,sf}(\omega) - [\beta_G + \beta'_G(\omega - \omega_G)]z_{fila}, \quad (3.58)$$

$$\varphi_{IR}(\omega) = \varphi_{IR,sf}(\omega) - [\beta_{IR} + \beta'_{IR}(\omega - \omega_{IR})]z_{fila}, \quad (3.59)$$

where

β_G and β_{IR} are the propagation constant of sapphire at ω_G and ω_{IR} , respectively

β'_G and β'_{IR} are the first derivatives of β at the center wavelengths of the green and IR pulses

The phase delay of the IR pulse and the green pulse are given by $\beta_{IR}z_{fila} = (\omega_{IR}/c)n(\omega_{IR})z_{fila}$ and $\beta_Gz_{fila} = (\omega_G/c)n(\omega_G)z_{fila}$, respectively. The group delay of the IR and the green pulses are given by $\beta'_{IR}z_{fila} = (d\beta/d\omega)|_{\omega_{IR}}z_{fila}$ and $\beta'_Gz_{fila} = (d\beta/d\omega)|_{\omega_G}z_{fila}$, respectively.

If the phase matching of the second harmonic generation of the IR pulses is assumed to be perfect, the spectral phase of the second harmonic then becomes

$$\varphi_{SH}(\omega) = 2\varphi_{IR,sf}(\omega_{IR}) - 2[\beta_{IR} + \beta'_{IR}(\omega - \omega_G)]z_{fila}. \quad (3.60)$$

The total phase then becomes

$$\Phi(\omega) = \varphi_{CE} + \omega\tau_g - \omega_G\tau_g + (\Delta\varphi_{n_2} + \phi_{spm} + \omega_0\Delta\tau_0 + \omega_G\tau_{ph}), \quad (3.61)$$

where the phase delay between the second harmonic ($2f$) and green (f) pulses is

$$\tau_{ph} = z_{fila} \left(\frac{\beta_G}{\omega_G} - \frac{\beta_{IR}}{\omega_{IR}} \right) \quad (3.62)$$

and the group delay is given by

$$\tau_g = z_{fila} (\beta'_G - \beta'_{IR}). \quad (3.63)$$

The total phase expressed by (3.61) is equivalent to that expressed by (3.51), i.e., $\Phi(\omega) = \varphi_{CE} + \omega\tau_g + [\varphi_{SH}(\omega) - \varphi_{WL}(\omega)]$. Here $\varphi_{WL}(\omega) = \varphi_G(\omega)$.

3.5.3.4 Group Delay

It was found that the measured $\Phi(\omega)$ versus ω plot at a given laser intensity is almost a straight line as depicted by the solid section of the lines in Figure 3.22a. The frequency range of the solid section is what can be measured. The slope of the line should be equal to the group delay τ_g as Equation 3.61 suggests. This slope obtained by fitting the measured $\Phi(\omega)$ line increases with laser energy, as shown in the Figure 3.22b, where the change of the slope is plotted.

The $\Phi(\omega)$ line can be extrapolated to $\omega = 0$ as the dashed section of the lines shown in Figure 3.22a. When the total phase $\Phi(\omega)$ at ω_G is locked to zero in the experiments, the measured intercept of the $\Phi(\omega)$ versus ω line with the vertical axis decreases with the pulse energy, as shown in the Figure 3.22c. In this case, $\Phi(\omega) = \omega\tau_g - \omega_G\tau_g$ because $\varphi_{CE} + (\Delta\varphi_{n_2} + \phi_{spm} + \omega_0\Delta\tau_0 + \omega_G\tau_{ph}) = 0$. In other words, the measured intercept should correspond to $\Phi(0) = -\omega_G\tau_g$. The calculated $-\omega_G\tau_g$ also decreases with laser energy, as shown in the figure.

Two fitting parameters are used in the calculations of τ_g with Equation 3.63 for the plots. First, the power was chosen to be 32% of the peak power of the input pulse. Second, the spot size was fitted as $9.68 \pm 0.4 \mu\text{m}$.

3.5.3.5 Carrier-Envelope Phase Measurement Error

For the CE phase measurement, the total phase $\Phi(\omega)$ was measured at $\omega \approx \omega_G$, which leads to $(\omega_G - \omega)\tau_g \approx 0$. Therefore, the group delay fluctuation does not directly affect the CE phase measurement. Also, the product $\omega_G\tau_g$ agrees with the intercept of the measured $\Phi(\omega)$ versus ω plot, when $\Phi(\omega_G)$ is stabilized for CE phase locking.

At $\omega = \omega_G$, $\Phi(\omega) \approx \varphi_{CE} + (0.7\phi_{spm} + \omega_0\Delta\tau_0 + \omega_G\tau_{ph})$. It is clear that the measurement error is caused by the pulse-to-pulse variation of the quantity $\Delta\varphi_{err} = \Delta(0.7\phi_{spm} + \omega_0\Delta\tau_0 + \omega_G\tau_{ph})$ due to laser intensity fluctuations.

z_{sf} decreases as the laser energy increases, which leads to an increase of the filament length z_{fila} . Consequently, $\omega_G\tau_{ph}$ also increases with the pulse energy. On the contrary, $\omega_0\Delta\tau_0$ decreases with the shortening of z_{sf} which cancels the effects of $\omega_G\tau_{ph}$ to a large degree, as the calculation shows. The nonlinear term, $0.7\phi_{spm}$, also counters the effects of $\omega_G\tau_{ph}$. However, the overall result is that $\Delta\varphi_{err}$ increases with laser energy. This explains

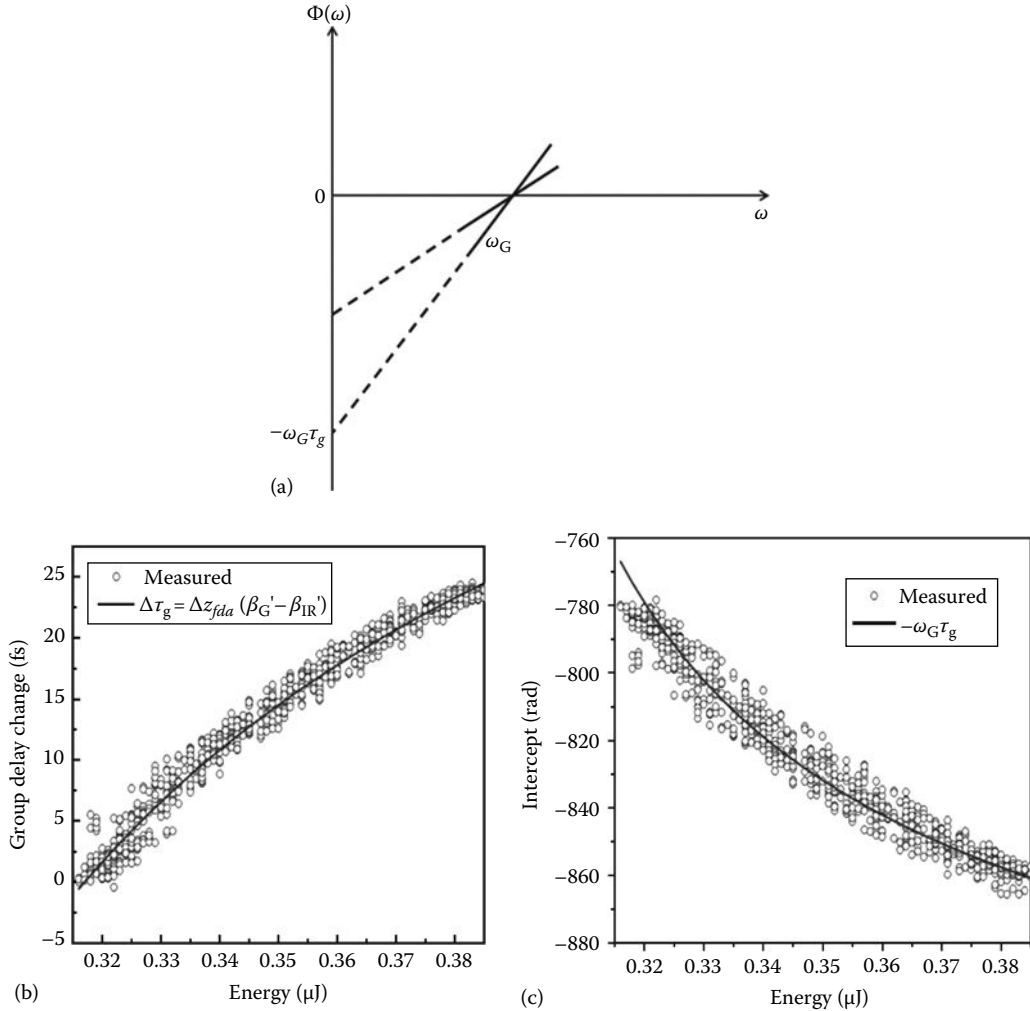


Figure 3.22 (a) The total phase. (b) The change of group delay between green and IR pulses as a function of laser energy. (c) Intercept of total phase as a function of laser energy. (Adapted from C. Li, E. Moon, H. Mashiko, H. Wang, C.M. Nakamura, J. Tackett, and Z. Chang, Mechanism of phase-energy coupling in f -to- $2f$ interferometry, *Appl. Opt.*, 48, 1303, 2009. With permission of Optical Society of America.)

the measured decrease of $\Delta \varphi_{CE} = -\Delta \varphi_{err}$ with laser energy when $\Phi(\omega_G)$ was locked to zero. The calculated phase error as a function of laser energy is plotted in Figure 3.23, which agrees well with the measured value. The center wavelength in $\omega_0 \Delta \tau_0$ for generating the above plot was chosen as 750 nm, which is another fitting parameter.

The formation of a filament in solid with femtosecond lasers is a complicated nonlinear process. One often needs to solve the nonlinear Schrödinger equation to obtain quantitative results. The simple model presented here should be considered as a semiquantitative one that reveals the major factors contributing to the CE phase measurements. When the laser pulse energy changes, the length of the filament varies. The CE

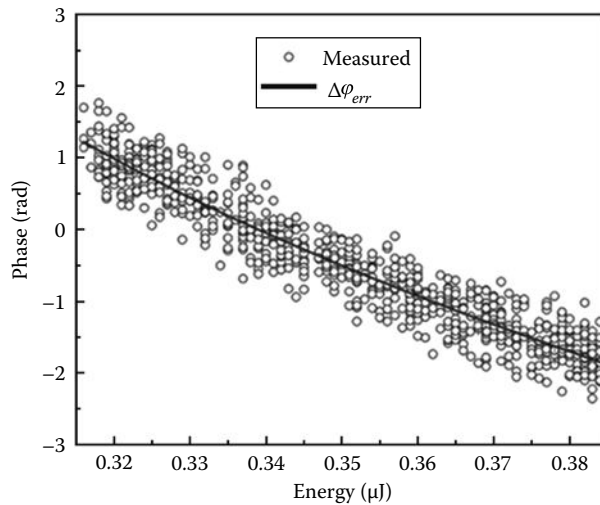


Figure 3.23 Comparison between the experimental and calculated results of the CE phase shift due to laser energy change. (Adapted from C. Li, E. Moon, H. Mashiko, H. Wang, C.M. Nakamura, J. Tackett, and Z. Chang, Mechanism of phase-energy coupling in f -to- $2f$ interferometry, *Appl. Opt.*, 48, 1303, 2009. With permission of Optical Society of America.)

phase given by the f -to- $2f$ method depends on the filament length. That is why it fluctuates with the laser power.

3.6 Carrier-Envelope Phase Shift in Stretchers and Compressors

3.6.1 Carrier-Envelope Phase Shift Introduced by Grating-Based Compressors

We analyze the compressor first because it has less optical components. In femtosecond CPA laser systems, the laser beam travelling through a grating pair in the compressor is reflected back so that the beam passes through the gratings for a second time. The purpose of doing so is to remove the spatial chirp of the beam. The configuration of the double-pass grating compressor is shown in [Figure 3.24](#), where M is the mirror that reflects the beam. The reflected beam is not shown. We first discuss the CE phase shift after a single pass.

3.6.1.1 Carrier-Envelope Phase

In CPA lasers, the pulses that enter the compressor are positively chirped. For simplicity, the input pulse here is assumed to be transform limited and linearly polarized, which can be expressed as follows:

$$\varepsilon(t) = E(t)e^{i(\omega_0 + \varphi_{CE})}, \quad (3.64)$$

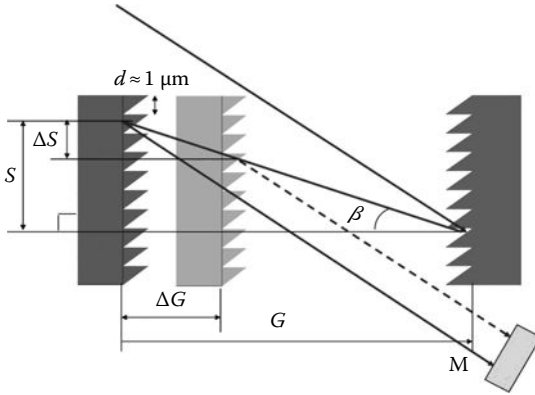


Figure 3.24 Configuration of a double-pass grating compressor. G , grating separation; d , grating constant; β , diffraction angle; M , retro-reflection mirror. (Reprinted from C. Li, E. Moon, H. Mashiko, C. Nakamura, P. Ranitovic, C.L. Cocke, Z. Chang, and G.G. Paulus, Precision control of carrier-envelope phase in grating based chirped pulse amplifiers, *Opt. Express*, 14, 11468, 2006. With permission of Optical Society of America.)

where

$E(t)$ is the envelope of the laser pulse

ω_0 is the carrier frequency

φ_{CE} is the CE phase

In this case, the grating pairs stretch the pulses. However, the conclusions reached with the assumption are valid for the conventional case where the incident pulses are chirped.

In the frequency domain, the electric field of the input pulse is

$$\tilde{E}(\omega) = U(\omega)e^{i[\varphi_{CE} + \varphi(\omega)]}, \quad (3.65)$$

where

$U^2(\omega)$ is the power spectrum

$\varphi(\omega)$ is the spectral phase, which is set to zero for the transform-limited input pulse

As discussed in Chapter 2, when the pulse propagates through the grating compressor, its spectral phase becomes

$$\varphi(\omega) = -\frac{\omega}{c}P'(\omega) + \varphi_c(\omega), \quad (3.66)$$

where P' is the optical path length, and

$$\varphi_c(\omega) = 2\pi \frac{G}{d} \tan[\beta(\omega)], \quad (3.67)$$

where

G is the perpendicular distance between the gratings

d is the grating constant

$\beta(\omega)$ is the diffraction angle, as shown in [Figure 3.24](#)

It can be rewritten as

$$\varphi(\omega) = -\omega\tau(\omega) + 2\pi \frac{G}{d} \tan[\beta(\omega)]. \quad (3.68)$$

The group delay

$$\tau(\omega) = \frac{P'}{c}. \quad (3.69)$$

The difference of the group delay, $-\omega\tau(\omega)$, and phase delay, $\varphi(\omega)$, at the center frequency is the CE phase shift for a single pass through the grating pairs, which is

$$\varphi'_{CE} - \varphi_{CE} = \varphi(\omega_0) - [-\omega_0\tau(\omega_0)] = 2\pi \frac{G}{d} \tan[\beta(\omega_0)], \quad (3.70)$$

where φ'_{CE} is the CE phase at the exit. For a given grating, d is a constant. The CE phase shift can be introduced by the variation of either G or β .

3.6.1.2 Beam Pointing

The direction of a laser beam changes with time due to the thermal effects of the gain crystal, the vibration of mirror mounts, etc. It has been studied that the variation of $\beta(\omega_0)$ due to laser beam pointing can change φ'_{CE} . In a double-pass configuration, the increase of incident angle in the first pass leads to the decrease of the incident angle in the second pass as a result of the reflection on the mirror M. Thus, the net effects of the incident angle variation on the CE phase are small. The CE phase shift due to beam pointing is proportional to the absolute value of the grating spacing G . Thus, small values of G are preferred. However, it reduces the duration of the stretched pulses in the CPA. In CPA lasers, the values of G and d are chosen to compress ~ 100 ps chirped pulses.

3.6.1.3 Grating Separation

When the separation between the gratings is changed by an amount ΔG due to a mechanical vibration (fast variation) or thermal drift (slow variation), the subsequent CE phase variation for a single pass is

$$\Delta\varphi_{CE} = 2\pi \frac{\Delta G}{d} \tan[\beta(\omega_0)]. \quad (3.71)$$

When the incident angle γ is near the Littrow angle, i.e., the incident and diffraction angles are almost the same, at which the gratings are most efficient, Equation 3.71 can be expressed in terms of incident angle

$$\Delta\varphi_{CE} \approx 2\pi \tan(\gamma) \frac{\Delta G}{d}. \quad (3.72)$$

It is a common practice to use gratings with $d^{-1} = 1200$ lines/mm because high-diffraction efficiency can be achieved at 800 nm. If the grating mounts are not interferometrically stable, the magnitude of ΔG can reach a fraction of a laser wavelength, which is on the order of half the grating constant $d/2 \approx 0.4 \mu\text{m}$. In this case, $\Delta\varphi_{CE} \approx 2\pi \tan(\gamma)$, which is on the order of 2π radians since $\gamma > 45^\circ$ for most compressors. This simple estimate reveals the significance of stabilization of the grating separation in the compressor of the chirped pulse amplification systems.

Equation 3.71 can also be written as

$$\Delta\varphi_{CE} = 2\pi \frac{\Delta S}{d}, \quad (3.73)$$

where ΔS is displacement of the laser beam on the second grating (the one on the left), as defined in Figure 3.24. This equation can be explained by the fact that each groove introduces a 2π phase shift, which originates from the definition of phase for the wave in the grating system. The CE phase change for each pass is equal to the number of grooves covered by S defined in Figure 3.24 multiplied by 2π .

3.6.2 Carrier-Envelope Phase Shift Introduced by Grating-Based Stretcher

In Figure 3.25, two grating-based stretcher configurations are shown. The top one shows a double-pass stretcher utilizing mirrors in the telescope arrangement. For femtosecond CPA lasers, mirrors are commonly used in the telescope to avoid chromatic aberrations. However, it is easier to understand the stretching principle with the bottom one with lenses as the telescope. The two are optically identical when aberrations are ignored.

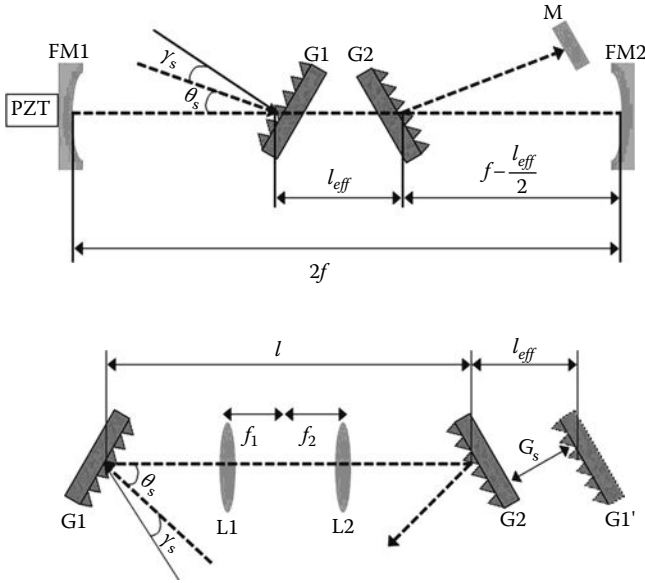


Figure 3.25 Grating-based stretchers for CE phase stabilization of the amplified laser pulses: γ_s , angle of incidence; θ_s , angle between the incident and diffracted rays; l_{eff} , effective grating separation; f , focal length; FM1 and FM2 are the focusing mirrors; M, retro-reflecting mirror; PZT, piezoelectronic transducer; G_s , perpendicular distance between the gratings; G1 and G2, the gratings; G1', image of G1; l , the distance between the gratings. (Reprinted from C. Li, E. Moon, and Z. Chang, Carrier-envelope phase shift caused by variation of grating separation, *Opt. Lett.*, 31, 3113, 2006. With permission of Optical Society of America.)

The CE phase shift after a single-pass grating stretcher can also be expressed by Equation 3.71 except that the distance between the gratings, G , needs to be replaced by the effective perpendicular distance

$$G_s = l_{eff} \cos(\gamma_s + \theta_s), \quad (3.74)$$

where

γ_s is the incident angle

θ_s is the angle between the incident and diffracted rays, as shown in Figure 3.25

l_{eff} is the effective linear distance between the gratings, which is the distance between the second grating and the image of the first grating formed by the telescope.

When the two lenses are confocal, the image position of the G1 can be found by using geometric optics, which gives

$$l_{eff} = [l - 2(f_1 + f_2)] \left(\frac{f_1}{f_2} \right)^2, \quad (3.75)$$

where

f_1 and f_2 are the focal lengths of the lenses (mirrors) that form the telescope between the gratings

l is the geometrical distance between the two gratings

For most stretchers, $f_1 = f_2 = f$, then Equation 3.75 can be simplified to $l_{eff} = l - 4f$.

When $\theta_s \approx 0$, the amount of single-pass CE phase errors introduced by the variation of the effective grating separation, Δl_{eff} , is

$$\Delta \varphi_{CE} \approx 2\pi \tan(\gamma_s) \frac{\Delta l_{eff} \cos(\gamma_s)}{d} = 2\pi \sin(\gamma_s) \frac{\Delta l_{eff}}{d_s}. \quad (3.76)$$

The incident angle, γ_s , and grating constant, d_s , of the stretcher can be different from those of the compressor. The change of the l_{eff} value can originate from the motion of either the lenses or the gratings.

Equation 3.76 can be further simplified if two approximations are made: when the incident angle is close to the Littrow angle and when $d_s \approx \lambda_0$. In this case, Equation 3.76 simplifies to

$$\frac{\Delta \varphi_{CE}}{\Delta l_{eff}} = 2\pi \frac{\lambda_0}{d_s^2} \approx \frac{2\pi}{\lambda_0}, \quad (3.77)$$

where λ_0 is the center wavelength of the laser. Equation 3.77 shows that a variation in the effective grating separation on the order of a wavelength will impart a significant CE phase shift to a laser pulse, like in the case of grating compressor.

3.6.2.1 Pulse Duration

The compressed pulse duration, τ_p , does not change much when $\Delta l_{eff} \approx \lambda_0$. It can be shown that

$$(\tau'_p)^2 = \tau_p^2 + \left(\frac{\tau_s}{l_{eff}} \Delta l_{eff} \right)^2, \quad (3.78)$$

where

τ'_p is the compressed pulsed duration when the grating separation is changed

τ_s is the stretched pulse duration

For $\tau_p \approx 20$ fs, $\tau_s/l_{eff} \approx 1$ fs/ $1 \mu\text{m}$, and $\tau'_p = 20.025$ fs when $\Delta l_{eff} = 1 \mu\text{m}$. In other words, a change of grating separation on the order of a laser wavelength only causes an increase of small fraction of a femtosecond in laser pulse duration. Thus, for the CE phase-stabilized amplifiers the requirement of mechanical stability is much stricter than for conventional chirped pulse amplification laser systems.

3.7 Stabilization of the Carrier-Envelope Phase in CPA

Even if the CE phase of pulses from the oscillator is perfectly stabilized, the phase at the output of a chirped pulse amplifier can still deviate from a constant value. Many factors can cause the CE phase to fluctuate, such as the temperature variation of the gain crystal, the change of the separation of the grating pairs in the stretcher, and compressor due to vibration, etc. To compensate the CE phase variation caused by the components after the oscillator, we can take advantage of the fact that the CE phase is susceptible to the grating separation in the stretcher or the compressor. The CE phase can be stabilized by feedback controlling the separation.

3.7.1 Using the Compressor

The CE phase of a CPA system with two multipass amplifiers in the author's lab was used for the demonstration.* The experimental layout is shown in Figure 3.26. The CE offset frequency of the pulses from the laser oscillator is stabilized by controlling the pump power. Pulses with the same CE phase are selected by a Pockels cell and sent to the stretcher of the Ti:Sapphire CPA with a repetition rate of 1 kHz. The slow CE phase drift in the amplifier is corrected by feedback controlling the grating separation in the compressor.

To lock the CE phase, one of the gratings in the compressor is mounted on a PZT stage to change the grating spacing. A PZT is a disk or a cylinder. The length of it can be changed by a voltage applied on the two surfaces. PZT, instead of a motor, is used because it can move the grating with high speed, which is important for achieving a broad feedback bandwidth. The range of motion is only a few micrometers, which is within the capability of the PZT.

A small portion of the laser pulse energy, about $1 \mu\text{J}$, from the chirped pulse amplifier is directed to a single shot f -to- $2f$ interferometer in order to

* The text in this section is adapted from Li, C., H. Mashiko, H. Wang, E. Moon, S. Gilbertson, and Z. Chang, Carrier-envelope phase stabilization by controlling compressor grating separation, *Appl. Phys. Lett.* 92, 191114 (2008).

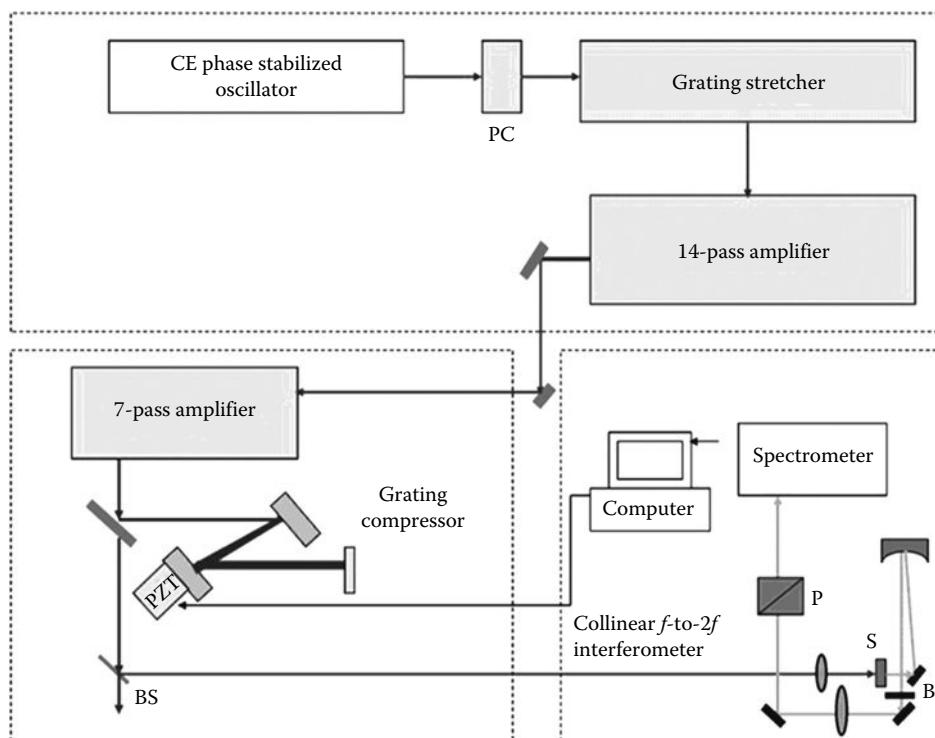


Figure 3.26 Experimental setup for controlling the CE phase using the grating compressor. PC, Pockels cell; BS, beam splitter. (Adapted from C. Li, E. Moon, H. Mashiko, H. Wang, C.M. Nakamura, J. Tackett, and Z. Chang, Mechanism of phase-energy coupling in f -to- $2f$ interferometry, *Appl. Opt.*, 48, 1303, 2009. With permission of Optical Society of America.)

measure the CE phase drift. The measured CE phase is compared to a preset value to yield an error signal. The grating spacing is changed by a software PID controller to minimize the error signal.

3.7.1.1 Frequency Response of the PZT Mount

The CE phase drift caused by temperature variation or air flow in the laser container is slow. On the contrary, CE phase noise introduced by vibration and acoustic disturbance is fast. Ideally, one would like to suppress all the phase variation. In reality, we can only reduce the relatively slow phase changes. The frequency range over which the CE phase noise can be effectively suppressed is determined by the bandwidth of the feedback control loop.

One of the major factors limiting the bandwidth is the response time of the PZT loaded with the grating, i.e., by its resonance frequency. For example, the frequency response of a grating used in the author's lab is measured with a Michelson interferometer with a CW laser. The grating attached to a PZT replaces a reflecting mirror in one of the arms. The zero-order refraction of the grating serves as a mirror. A fast photodiode measures the laser at the exit of the interferometer. When the arm length is changed by the PZT, the signal on the photodiode also changes due to the interference of the laser beams from the two arms. When a sinusoidal

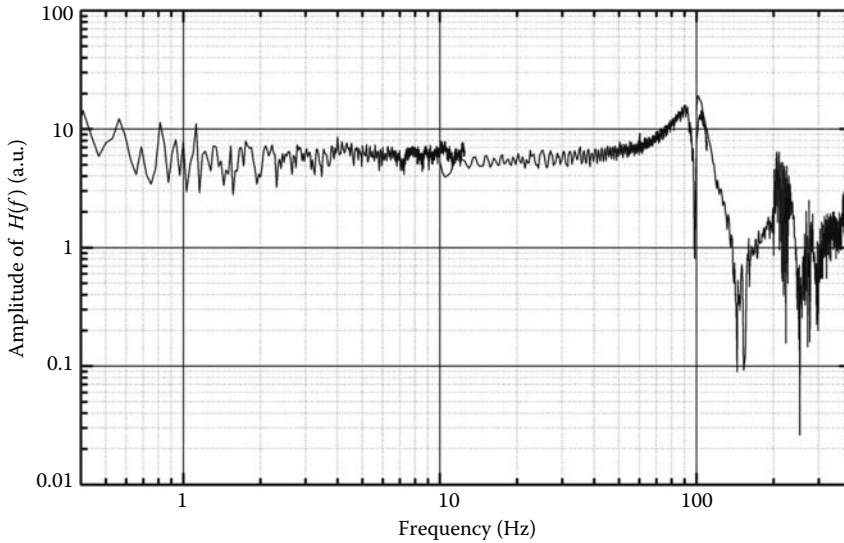


Figure 3.27 The frequency response of the PZT mounted grating. (Adapted from C. Li, E. Moon, H. Mashiko, H. Wang, C.M. Nakamura, J. Tackett, and Z. Chang, Mechanism of phase-energy coupling in f -to- $2f$ interferometry, *Appl. Opt.*, 48, 1303, 2009. With permission of Optical Society of America.)

voltage is applied on the PZT, the photodiode signal is also a sinusoidal function. A scan of the voltage frequency allows the measurement of the Fourier transfer function (frequency response function), which represents the response time of the grating and its mount.

The output of a photodiode is sent to a dynamic signal analyzer. The signal can be expressed as

$$S(f) = H(f)V(f), \quad (3.79)$$

where

$H(f)$ is the frequency response function

$V(f)$ is a sinusoidal voltage applied on the PZT generated by the dynamic signal analyzer

The result is shown in Figure 3.27 when the frequency, f , is scanned. The bandwidth is about 90 Hz, which is limited by the resonance frequency found near 100 Hz determined primarily by the mass and the spring constant of the grating mount. Apparently, the weight of the mount and grating should be as small as possible to increase the cutoff frequency. For a kilohertz laser, the CE phase noise spectrum can reach to the repetition rate of the laser. Due to the resonance of the grating mount, the phase noise above 100 Hz cannot be suppressed by the feedback control.

3.7.1.2 Frequency Response of the f -to- $2f$ Interferometer and of the PZT

Other factors such as the integration time of the CCD camera in the spectrometer of the f -to- $2f$ interferometer also limit the bandwidth of the feedback control loop. The response of the whole loop is measured by applying a sinusoidal voltage $V(f)$ to the PZT while measuring the CE

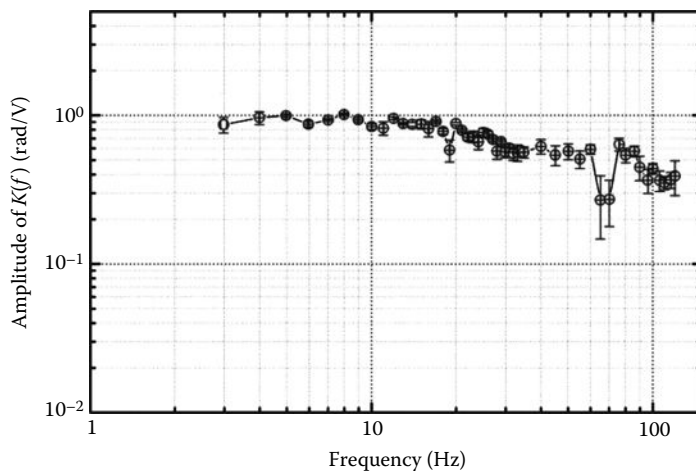


Figure 3.28 The frequency response of the retrieved CE phase control. (Adapted from C. Li, E. Moon, H. Mashiko, H. Wang, C.M. Nakamura, J. Tackett, and Z. Chang, Mechanism of phase-energy coupling in f -to- $2f$ interferometry, *Appl. Opt.*, 48, 1303, 2009. With permission of Optical Society of America.)

phase with an f -to- $2f$ interferometer. When the frequency of the sinusoidal wave is scanned, the measured CE phase can be expressed as follows:

$$|\Delta\varphi_{CE}(f)| = K(f)|V(f)|, \quad (3.80)$$

where $K(f)$ is the frequency response of the f -to- $2f$ interferometer together with that of the grating and PZT stage.

The result of the measurement is shown in Figure 3.28. The resonant frequency of the feedback control system was found to be near 60 Hz, which implies that CE phase drift lower than 60 Hz could be corrected by moving the grating using this particular system. The frequency is much lower than the laser repetition rate (1 kHz), which is the limitation of the feedback scheme.

3.7.1.3 Carrier-Envelope Phase Locking

The experimental results are shown in Figure 3.29. In the top graph, the dotted line shows the free running CE phase drift (feedback was turned off). The solid line is the CE phase when the feedback was switched on. The CE phase was stabilized over 270 s with a 230 mrad RMS error. The bottom plot of Figure 3.29 shows the Fourier transform spectra of the locked and unlocked cases. The graph shows that the slow CE phase drift (<4 Hz) is well corrected. Although the high-frequency CE phase noise is not reduced, the quality of the CE phase stability is good enough for many applications, including the generation of single isolated attosecond pulses.

3.7.2 Using the Stretcher

The gratings in the compressor of a CPA laser can be larger and heavier than the ones used in the stretcher. This is because the laser beam diameter

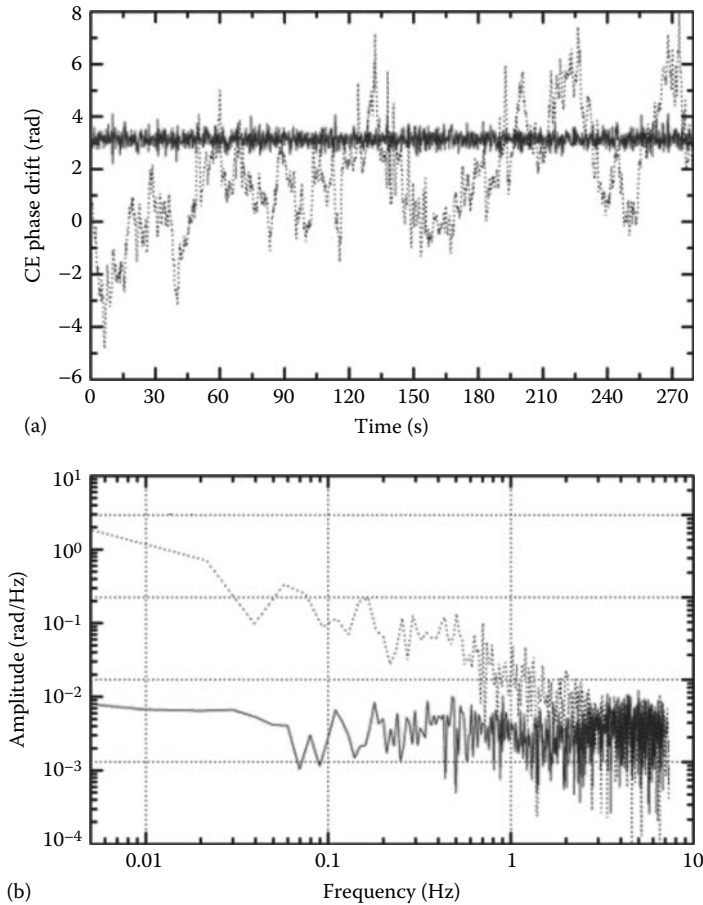


Figure 3.29 (a) The evolution of the free drifted (dotted line) and stabilized (solid line) CE phase drift. (b) The fast Fourier transform of the CE phase drift under the free running (dotted line) and stabilized conditions (solid line). (Adapted from C. Li, E. Moon, H. Mashiko, H. Wang, C.M. Nakamura, J. Tackett, and Z. Chang, Mechanism of phase-energy coupling in f -to- $2f$ interferometry, *Appl. Opt.*, 48, 1303, 2009. With permission of Optical Society of America.)

is larger to avoid damage of the grating by the amplified laser beam. The resonance frequency of the grating and its mount is inversely proportional to the square root of their mass; thus, one may choose to control the lighter gratings or other equivalent optical components in the stretcher to stabilize the CE phase or the amplified pulses, which has been demonstrated in the author's lab in 2006, which is the first time that the CE phase of a grating-based CPA is actively stabilized.

The setup for stabilizing the CE phase of the amplified laser pulses from a CPA laser with a single stage multipass amplifier is shown in [Figure 3.30](#), which was accomplished by feedback controlling the effective grating separation in the stretcher. In this case, the phase error signal is sent to the PZT that carries one of the telescope mirrors in the stretcher. In this example, the integration time of the spectrometer's CCD camera in the f -to- $2f$ is 50 ms.

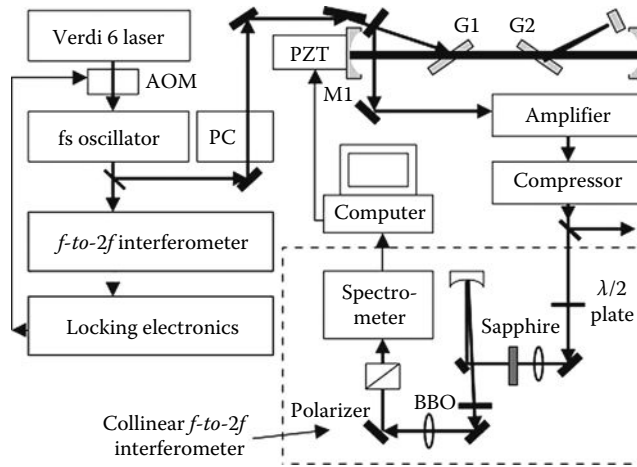


Figure 3.30 CE phase stabilization using stretcher.

3.7.2.1 Dependence of Carrier-Envelope Phase on the Effective Grating Separation

The dependence of CE phase on the grating separation is measured experimentally. Instead of moving the grating directly, one of the telescope mirrors is moved with a PZT stage. It changes the effective grating separation by shifting the image location of the first grating. The first mirror, M1, was chosen because of its small size as compared to both gratings and the other telescope mirror. Its high-resonance frequency is good for eliminating the phase noise over a broad frequency range.

In the setup, the groove density of the two gratings is $1/d = 1200$ lines/mm. The focal length of the two telescope mirrors is $f = 250$ mm. The effective linear distance between the gratings becomes $l_{eff} = 130$ mm, which is the distance between the second grating and the image of the first grating through the telescope.

When M1 moves away from the first grating G1, by Δ , the change of l_{eff} is

$$\Delta l_{eff} = -\Delta - \left(\frac{l_{eff}}{2f} \right)^2 \Delta \approx -\Delta. \quad (3.81)$$

The first term is from the increase of the distance between the first grating to the first mirror, while the second term originates from the increase of the mirror separation. Since $l_{eff} < 2f$, the contribution from the latter is small, which can be neglected. It means that moving the mirror is equivalent to displacing a grating.

The CE phase shift $\Delta\varphi_{CE}$ introduced by the mirror displacement Δ satisfies the equation for the double-pass stretcher,

$$\frac{\Delta\varphi_{CE}}{\Delta} = 4\pi \sin(\gamma_s + \theta_s) \frac{1}{d_s}. \quad (3.82)$$

For the laser used in the experiments, the incident angle of the laser beam on the first grating, G1, is $\gamma_s = 33.5^\circ$ and the diffraction angle

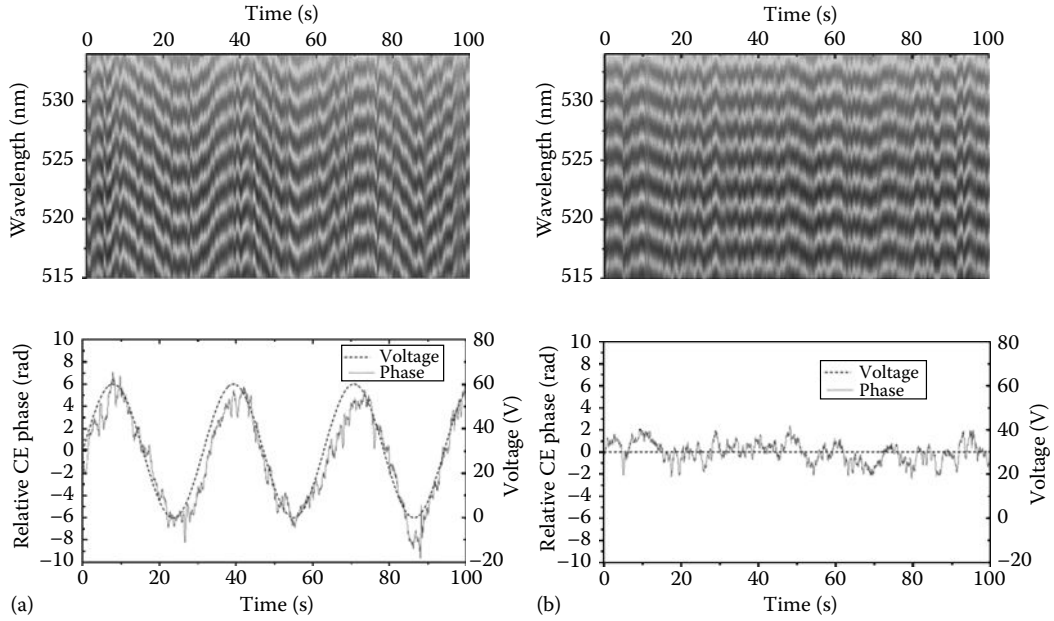


Figure 3.31 Effect of changing the grating separation on the CE phase drift: (a) 60 V_{p-p} voltage applied and (b) DC voltage applied. (Reprinted from C. Li, E. Moon, and Z. Chang, Carrier-envelope phase shift caused by variation of grating separation, *Opt. Lett.*, 31, 3113, 2006. With permission of Optical Society of America.)

$(\gamma_s + \theta_s) = 23.3^\circ$. Using these parameters, it was estimated that $\Delta\varphi_{CE}/\Delta \approx 6 \text{ rad}/\mu\text{m}$.

When a sinusoidal voltage is applied to the PZT, the mirror moves back and forth with a $3.6 \mu\text{m}$ displacement amplitude. The measured CE phase oscillator is shown in Figure 3.31a. The interferogram yielded $\Delta\varphi_{CE}/\Delta \approx \Delta\varphi_{CE}/\Delta l_{eff} \approx 6 \text{ rad}/\mu\text{m}$, which agreed with the calculated results. The CE phase oscillation disappears when the voltage is set to a constant value, as shown in Figure 3.31b. Since the gratings in the stretchers and compressor are not interferometrically stable, their vibration and thermal drift introduce CE phase deviations from a straight line.

3.7.2.2 Compensation of Slow Carrier-Envelope Phase Drift

The results in Figure 3.31 were taken when the feedback control was turned off. In that case, the CE phase fluctuates over π rad. The fluctuation was strongly suppressed when the feedback was turned on. Figure 3.32 shows how the feedback control of the effective grating separation in the stretcher corrected the slow CE phase drift of the amplified laser pulses. In the figure, the CE phase was locked in 200 s. The RMS error of the CE phase drift was 179 mrad, which is a typical value.

3.7.2.3 Effects of the Oscillator f-to-2f Stability

The effects of locking the optical path length difference in the oscillator f -to- $2f$ interferometer on the CE phase stability of the pulses from the amplifier have also been studied. In the experiment, the CE offset

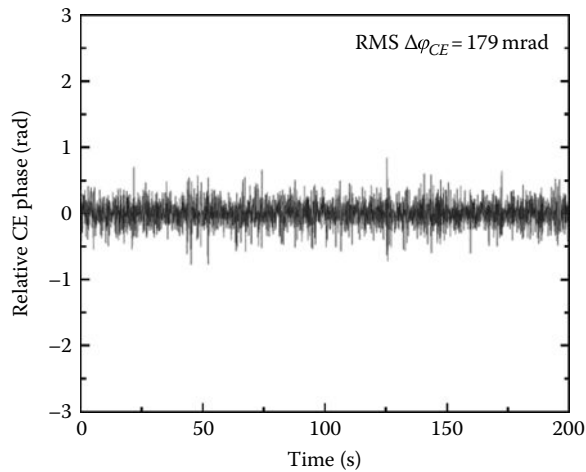


Figure 3.32 Stabilized CE phase drift. (Reprinted from C. Li, E. Moon, and Z. Chang, Carrier-envelope phase shift caused by variation of grating separation, *Opt. Lett.*, 31, 3113, 2006. With permission of Optical Society of America.)

frequency of the oscillator was stabilized by controlling the pump power. The slow drift of the CE phase of the amplified laser pulses was corrected by controlling the effective grating separation in the stretcher. The results are shown in Figure 3.33. The image on the left in this figure shows the CE phase stability of the system when the optical path length difference oscillator f -to- $2f$ interferometer was locked and unlocked. In the graph on the right, the fast CE phase noise was investigated by applying a high-pass (>3 Hz) filter to the measurements in the left figure. The results show that the high-frequency CE phase noise of the amplified laser

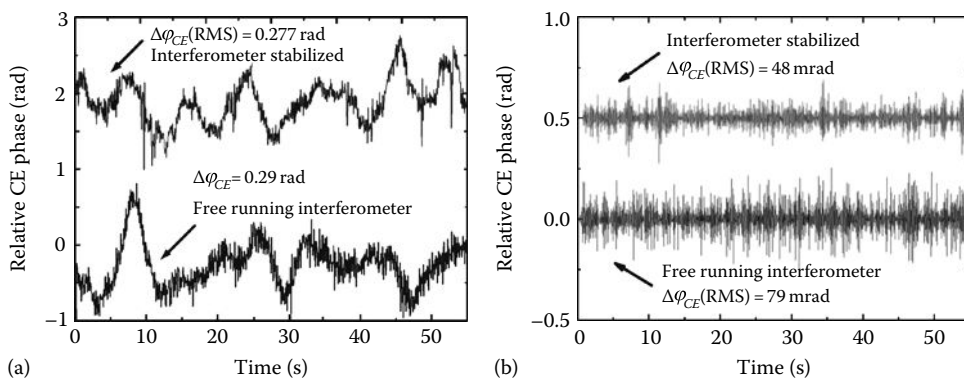


Figure 3.33 (a) The relative CE phase measured by the collinear f -to- $2f$ interferometer. (b) The fast jitter of the CE phase obtained by applying a high-pass filter to the spectra in (a). (Reprinted from E. Moon, C. Li, Z. Duan, J. Tackett, K.L. Corwin, B.R. Washburn, and Z. Chang, Reduction of fast carrier-envelope phase jitter in femtosecond laser amplifiers, *Opt. Express*, 14, 9758, 2006. With permission of Optical Society of America.)

pulses was suppressed by 40%. Thus, by locking the interferometer, the high-frequency CE phase noise, which could not be corrected by the amplifier feedback loop, was suppressed.

3.8 Controlling of the Stabilized Carrier-Envelope Phase

To study the dependence of a physics process, such as the attosecond spectral shape, on the CE phase of the driving laser, one would need to set the stabilized CE phase to any desired values. One method to vary the CE phase is to add a pair of thin glass wedges in the laser beam before it enters the interaction chamber. The CE phase can be continually changed by varying the total thickness of the wedges. It has been demonstrated that the value of the stabilized CE phase can also be changed by controlling the gratings in the stretcher or compressor of the CPAs.

3.8.1 Carrier-Envelope Phase Staircase

In some experimental situations, it might be desirable to sit at a fixed CE phase for a period of time to accumulate the signals (electron, ions, XUV photon) from the process being studied and then move to another point. Controlling the grating separation could not only be used to stabilize the CE phase drift but also to change the CE phase in steps. This can be accomplished by changing the set point for stabilization. An example is shown in [Figure 3.34](#).

The top graph of [Figure 3.34](#) shows the fringes obtained by using an f -to- $2f$ interferometer. The bottom plot shows the relative CE phase as the set point was varied. The CE phase was locked at each set point for 1 min and shifted to the next value in an increment of 0.2π . It can be seen that the amount of fringe shift is consistent with the set-point change.

3.8.2 Phase Sweeping

In some other circumstances, continuous variation of the CE phase might be desirable. An example of CE phase scan obtained experimentally is shown in [Figure 3.35](#). The figure shows a triangular modulation to the relative CE phase over $\sim 2.45\pi$.

The CE phase error and displacement of the PZT during the experiment are shown in [Figure 3.36](#). The RMS error during the experiment was 171 mrad. Obviously, there is a correlation between the PZT motion and the CE phase variation.

The ability to use grating separation to change the CE phase during an experiment obviates the need for wedge pairs. When wedge pairs are used to shift the CE phase by changing the amount of glass materials in the optical path, their positive GVD must be compensated by extra chirped mirrors. In laser systems, one should not add any component unless when it is really necessary.

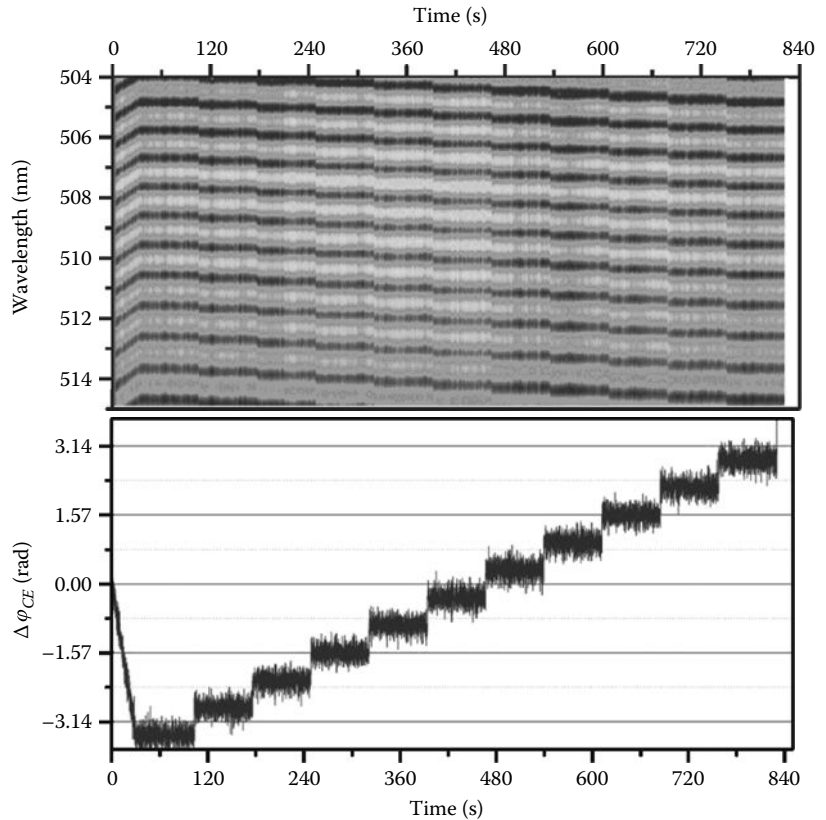


Figure 3.34 Precisely controlling the CE phase of the amplified laser pulses. Top graph: The temporal evolution of the interference fringes. Bottom graph: The effect of changing the locking set point on the measured relative CE phase. (Reprinted from C. Li, E. Moon, H. Mashiko, C. Nakamura, P. Ranitovic, C.L. Cocke, Z. Chang, and G.G. Paulus, Precision control of carrier-envelope phase in grating based chirped pulse amplifiers, *Opt. Express*, 14, 11468, 2006. With permission of Optical Society of America.)

3.9 Carrier-Envelope Phase Measurements after Hollow-Core Fibers*

As discussed in Chapter 2, gas-filled hollow-core fibers are commonly used to broaden the spectrum for compressing millijoule level 30 fs laser pulses from the chirped pulse amplifier to a few femtoseconds. Even if the CE phase of the CPA is locked, CE phase shift may still be introduced by the hollow-core fiber.

In some cases, the fiber output spectrum covers an octave; thus, it can be used for measuring CE phase shift with the f -to- $2f$ method for locking the CE phase of the white-light pulses from the fiber. In this case, the

* This section is adapted from Wang, H., E. Moon, M. Chini, H. Mashiko, C. Li, and Z. Chang, Coupling between energy and carrier-envelope phase in hollow-core fiber based f -to- $2f$ interferometers, *Opt. Express* 17, 12082 (2009).

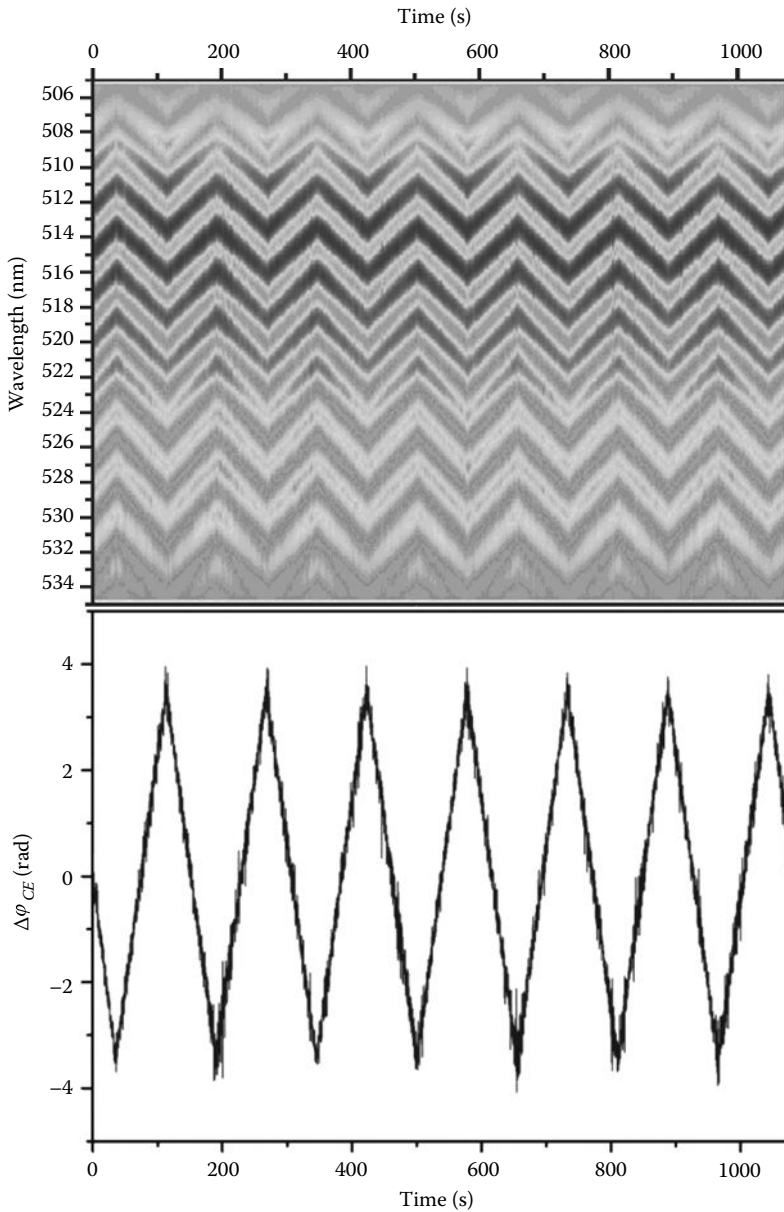


Figure 3.35 Top: Temporal evolution of the interference fringes. Bottom: Swept relative CE phase versus time.

phase locking accuracy is affected by the input pulse energy fluctuation. The laser energy to CE phase coupling in the hollow fiber has been measured in a similar way as that for the sapphire plate.

3.9.1 Experimental Setup

The experiment was carried out in the author's lab with a Ti:Sapphire chirped pulse amplification laser system equipped with a grating-based

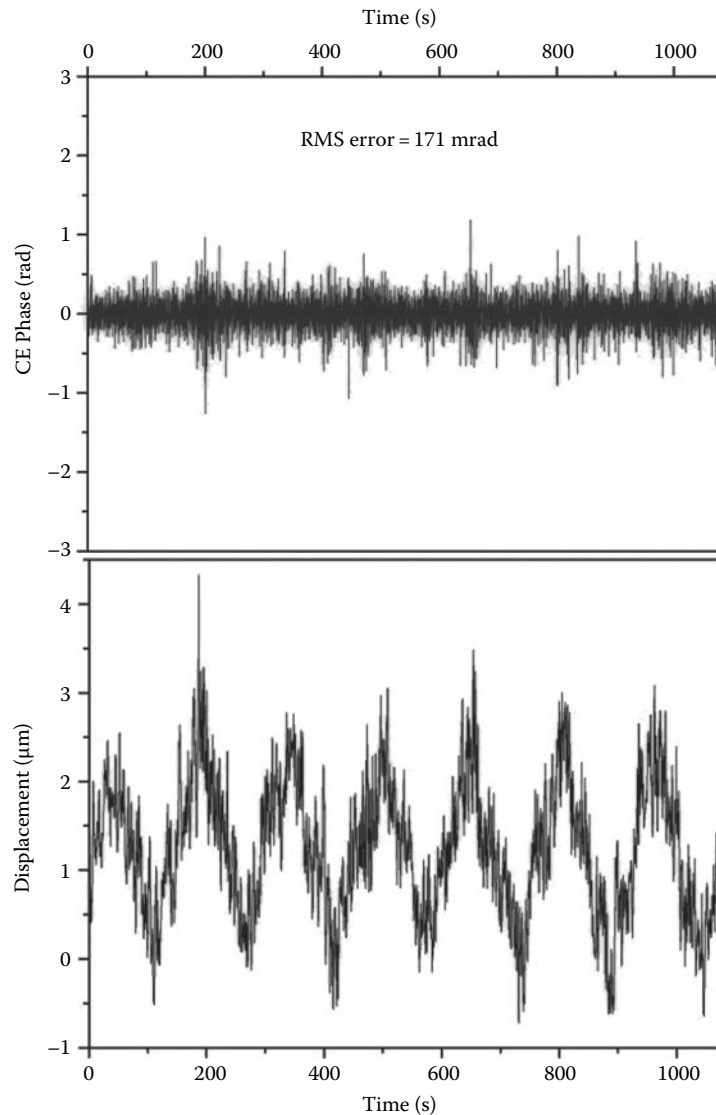


Figure 3.36 Top: CE phase error during the modulation. Bottom: Displacement of the PZT.

stretcher and compressor operating at 1 kHz, as shown in [Figure 3.37](#). The oscillator's CE offset frequency, f_0 , was stabilized. The output power of the CPA was stabilized to around 0.5% RMS, in order to improve the CE phase measurement accuracy and stability.

The 30 fs pulses with more than 2 mJ energy and with the beam diameter of 1 cm from the CPA laser were focused into a 0.9 m long, 400 μm inner core diameter hollow-core fiber filled with 2 bar of neon gas. Strong self-phase modulation produced white-light pulses with 1.2 mJ energy. The spectrum covered the 400–1000 nm range, which is more than one octave, as shown in [Figure 3.38](#). When the 900 nm light is frequency doubled,

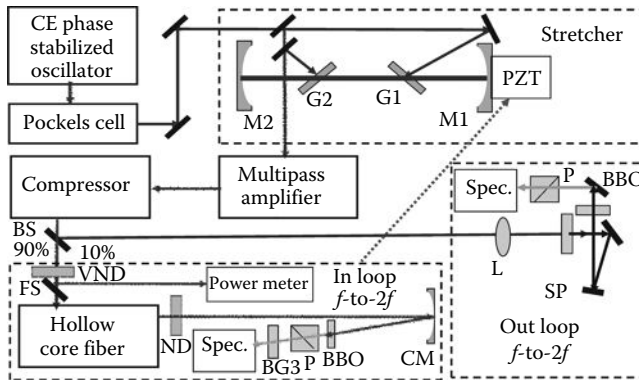


Figure 3.37 Experimental setup for determining the energy to CE phase coupling. VND, variable neutral density filter; L, focusing lens; SP, sapphire plate; BBO, frequency doubling crystal; P, polarizers; FS, fused silica; Spec., spectrometer and computer. In stretcher, G1 and G2, gratings; PZT, piezoelectric transducer; M1 and M2, mirrors; BS, beam splitter. (Reprinted from H. Wang, E. Moon, M. Chini, H. Mashiko, C. Li, and Z. Chang, Coupling between energy and carrier-envelope phase in hollow-core fiber based f -to- $2f$ interferometers, *Opt. Express*, 17, 12082, 2009. With permission of Optical Society of America.)

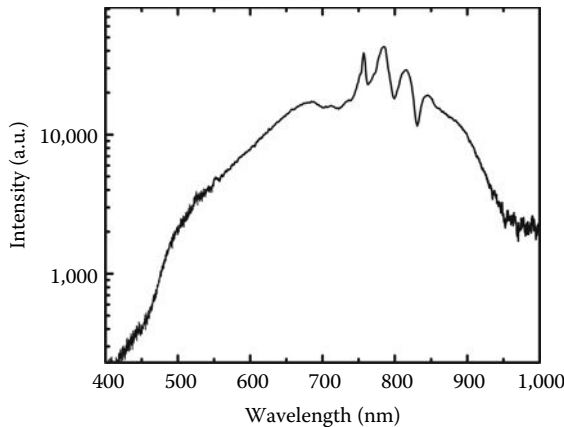


Figure 3.38 The output spectrum of the octave-spanning white light from the hollow-core fiber with 2 mJ input and 2 bar Ne pressure. (Reprinted from H. Wang, E. Moon, M. Chini, H. Mashiko, C. Li, and Z. Chang, Coupling between energy and carrier-envelope phase in hollow-core fiber based f -to- $2f$ interferometers, *Opt. Express*, 17, 12082, 2009. With permission of Optical Society of America.)

the interference of the second harmonic light with the 450 nm light in the white light gives the CE phase of the white-light pulse.

The effect of the input laser energy stability on the CE phase of the pulses from a hollow-core fiber was studied using two f -to- $2f$ interferometers.

The CE phase in the in-loop f -to- $2f$ interferometer was measured with the octave-spanning white-light spectrum from the hollow-core fiber. The

1 cm diameter beam was focused into a 100- μm -thick barium borate crystal (BBO) for second harmonic generation at 900 nm. The second harmonic field and the fundamental wave are orthogonally polarized as a result of the type I phase matching. A polarizer was used to project the second harmonic of the 900 nm light and the fundamental field at 450 nm onto the same polarization direction to facilitate interference. A spectrometer with a resolution of 0.11 nm was used to record the interference fringes over a 50 ms exposure time. The CE phase drift was extracted from the fringes by the standard algorithm of FTSI.

The out-of-loop f -to- $2f$ interferometer was based on white-light generation in a sapphire plate, which is located between the grating compressor and the hollow-core fiber.

3.9.2 Carrier-Envelope Phase Stability

The CE phase of the pulse from the hollow-core fiber was locked by feedback controlling the grating-based stretcher in the chirped pulse amplifier. The CE phase shift for the feedback was measured with the in-loop f -to- $2f$ interferometer. As shown in Figure 3.39, the CE phase after the fiber was locked within an in-loop accuracy of 94 mrad RMS. At the same time, the out-of-loop f -to- $2f$ interferometer measurement showed a CE phase fluctuation of 144 mrad before the fiber. In other words, the CE phase right after the CPA pulse was also locked although the feedback signal was obtained after the hollow-core fiber.

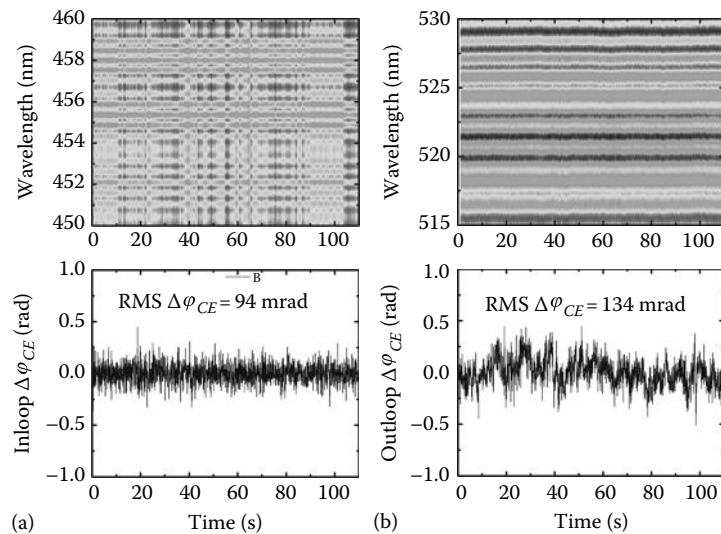


Figure 3.39 In-loop CE phase stabilized by a hollow-core fiber based f -to- $2f$ interferometer (left) and out-of-loop CE phase measured by a sapphire plate based f -to- $2f$ interferometer (right). (Reprinted from H. Wang, E. Moon, M. Chini, H. Mashiko, C. Li, and Z. Chang, Coupling between energy and carrier-envelope phase in hollow-core fiber based f -to- $2f$ interferometers, *Opt. Express*, 17, 12082, 2009. With permission of Optical Society of America.)

3.9.3 Energy to Carrier-Envelope Phase Coupling Coefficient

To quantitatively measure the laser energy to CE phase coupling coefficient for the hollow-core fiber-based f -to- $2f$ interferometer, a variable reflective fused silica NDF driven by an electric motor was placed before the focusing mirror of the hollow-core fiber to modulate the input power, as shown in Figure 3.37.

When the NDF wheel was rotated periodically within the range of 5° , the power was modulated within the range of 10%. The hollow-core fiber f -to- $2f$ interferometer was used to stabilize the CE phase after the fiber with a 107 mrad RMS, as shown in Figure 3.40a. Meanwhile, the sapphire plate f -to- $2f$ interferometer was used to measure the out-of-loop CE phase where the laser power was not modulated.

Figure 3.40b shows the anticorrelation between the in-loop power modulations and out-of-loop CE phase measurement. A least-square linear fit in Figure 3.40c shows that the 1% power fluctuation introduces a 128 mrad CE phase error, which is smaller than the 160 mrad for sapphire plate-based f -to- $2f$ interferometers. It is worthy to point that the factor is for the setup and the laser used in the experiments. It could be different for other laser systems. Obviously, the CPA output should be as stable as

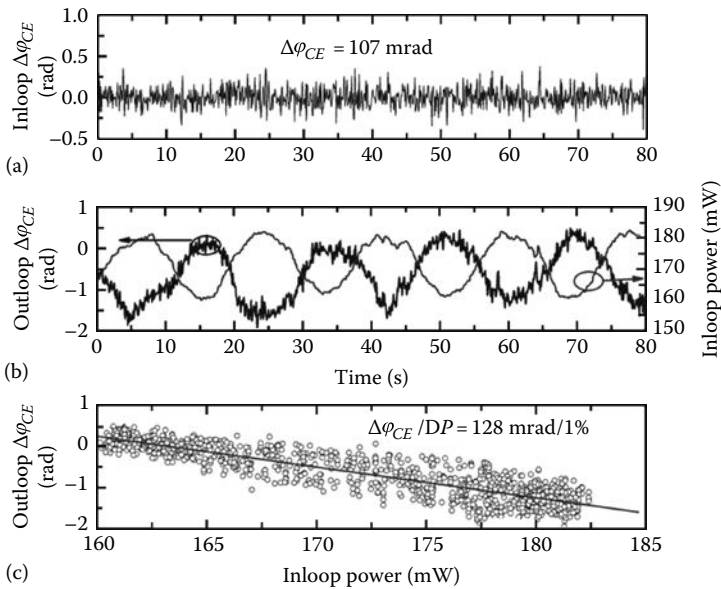


Figure 3.40 (a) In-loop CE phase locked by a hollow-core fiber based f -to- $2f$ interferometer. (b) Out-of-loop CE phase measured by a sapphire plate based f -to- $2f$ interferometer and the in-loop power modulation. (c) CE phase change to laser power coupling coefficient by a least-square fitting. (Reprinted from H. Wang, E. Moon, M. Chini, H. Mashiko, C. Li, and Z. Chang, Coupling between energy and carrier-envelope phase in hollow-core fiber based f -to- $2f$ interferometers, *Opt. Express*, 17, 12082, 2009. With permission of Optical Society of America.)

possible to improve the CE phase locking accuracy when the feedback signal is sampled after the hollow-core fiber.

3.10 Stabilizing Carrier-Envelope Phase of Pulses from Adaptive Phase Modulators*

As discussed in Chapter 2, few-cycle, high-power laser pulses can be generated by compressing the white-light from the hollow-core fiber with adaptive phase modulators. The positive chirp of the white-light pulses is removed by the phase modulators. Compared to chirped mirrors, adaptive phase modulators have high flexibility of phase control and can be adjusted to cope with the day-to-day phase variation of the white-light pulses. It has been demonstrated that the CE phase pulses from the phase modulator can also be stabilized.

3.10.1 Carrier-Envelope Phase Stability

A demonstration of the CE phase stabilization was carried out with the laser system in the author's lab, as shown in Figure 3.41, which consists of a Ti:Sapphire CPA with a grating-based stretcher and compressor,

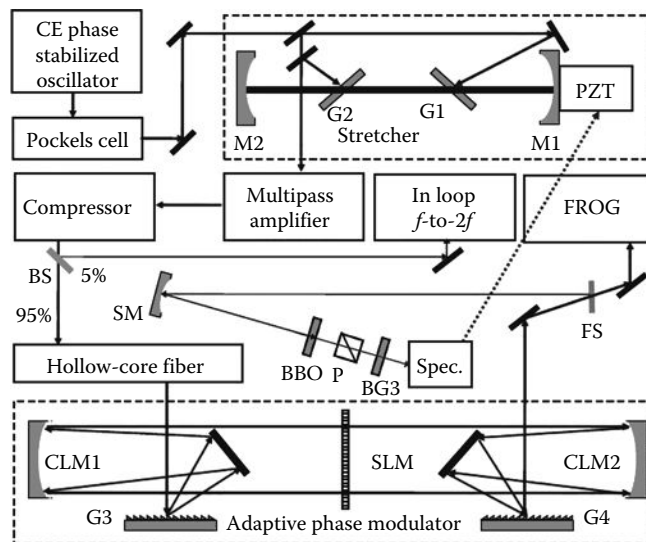


Figure 3.41 Experimental setup for generation of CE phase controllable 5 fs pulses. G1–G4, gratings; BS, beam splitter; FS, fused silica; CLM, cylindrical mirrors; SLM, liquid crystal spatial light modulator; SM, spherical mirror; P, polarizer. (Reprinted with kind permission from Springer Science+Business Media: *Appl. Phys. B*, Carrier-envelope phase stabilization of 5-fs, 0.5-mJ pulses from adaptive phase modulator, 98, 2010, 291, H. Wang et al.)

* The work is published in Wang, H., M. Chini, Y. Wu, E. Moon, H. Mashiko, and Z. Chang, Carrier-envelope phase stabilization of 5 fs, 0.5 mJ, pulses from adaptive phase modulators, *Appl. Phys. B* 98, 291–294 (2010).

a hollow-core fiber, and an adaptive phase modulator. During the experiment, the offset frequency f_0 of the femtosecond oscillator CE was stabilized first. After amplification, the pulses were compressed to 30 fs. A fraction of the output beam ($<1 \mu\text{J}$) was used to measure the relative CE phase of the amplified pulses with a sapphire plate-based f -to- $2f$ interferometer. The input pulse energy to the hollow-core fiber is 2 mJ. The spectrum of the 1.2 mJ white-light pulses comes out from the neon-filled hollow-core fiber, covered 400–1000 nm wavelength range. An adaptive phase modulator introduced in Chapter 2 was placed after the hollow-core fiber to compress the pulses. The output pulse energy from the phase modulator was 0.5 mJ.

The CE phase of the white-light pulses after the phase correction was measured by an f -to- $2f$ interferometer that took advantage of the $4f$ system of the phase modulator. The modulator was designed to perform spectral phase correction in the range of 500–1000 nm. However, the light around 460 nm can still go through the $4f$ system by the second order diffraction off of the grating, which overlapped with the fundamental component of 920 nm, as shown in Figure 3.41. It allowed the f -to- $2f$ measurement using the light at 460 and 920 nm.

In the second f -to- $2f$ interferometer, a 0.5 mm thick fused silica plate reflects about 3% of the output beam after adaptive phase modulator. A spherical mirror was used to focus both the 460 and 920 nm light into a $100 \mu\text{m}$ BBO to achieve high intensity there. The phase matching angle of the BBO was tuned to generate the second harmonic of the 920 nm light. A polarizer was used to project the second harmonic and the fundamental 460 nm light polarization onto the same axis, to facilitate interference. A spectrometer with a resolution of 0.11 nm was used to record the fringes at 50 ms exposure time. The voltage on spatial light modulator (SLM) above 910 nm was set to zero to obtain f -to- $2f$ fringes with good quality. With such an f -to- $2f$ interferometer, the CE phase after the adaptive phase modulator could be measured.

As shown in Figure 3.42, when the CE phase of the CPA was stabilized, the in-loop CE phase error before the hollow-core fiber was 171 mrad RMS. The CE phase noise measured after the adaptive phase modulator was 301 mrad, regardless of whether the SLM was turned on or off.

By comparing the phase error with and without the phase modulator, one can say that the extra CE phase drift is caused by the phase modulator itself. Furthermore, since the phase noise is almost the same when the SLM was on or off, the noise is likely introduced by the mechanical vibrations of the $4f$ zero dispersion compressor.

3.10.2 Carrier-Envelope Phase Error Introduced by the Zero-Dispersion Stretcher

Since the adaptive phase modulator is a grating-based zero dispersion $4f$ system, the effective distance between the two gratings was set to zero and the role of pointing fluctuation on CE phase can be ignored. However, the CE phase drift caused by a small fluctuation of the effective distance

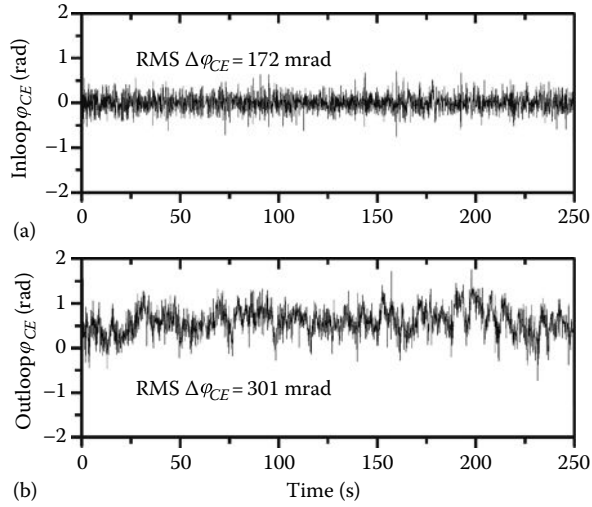


Figure 3.42 CE phase drift introduced by the adaptive phase modulator: (a) CE phase stabilized before the hollow-core fiber using sapphire-based in-loop f -to- $2f$ interferometer and (b) CE phase measured after the adaptive phase modulator by using the hollow-core fiber white light. (Reprinted with kind permission from Springer Science+Business Media: *Appl. Phys. B*, Carrier-envelope phase stabilization of 5-fs, 0.5-mJ pulses from adaptive phase modulator, 98, 2010, 291, H. Wang et al.)

between the two gratings can be expressed in the same way as regular stretcher in CPA lasers

$$\Delta\varphi_{CE} = 2\pi \frac{\Delta G}{d} \tan[\beta(\omega_0)], \quad (3.83)$$

where

ω_0 is the carrier frequency of the pulse

ΔG is small change in effective distance between the two gratings

d is the grating constant

β is the diffraction angle

It is estimated that a ΔG of $23 \mu\text{m}$ introduces a 2π CE phase drift for setup used in the demonstration. Thus, mechanical vibration of gratings and cavity mirrors leads to CE phase instability. Lower density gratings (large d) as compared to the CPA gratings are used in the adaptive phase modulator. Consequently, the CE phase is less susceptible to vibrations.

3.10.3 Compensate the Carrier-Envelope Phase Shift Introduced by the $4f$ System

The measured CE phase after the phase modulator was used to feedback control the grating separation of the CPA stretcher to compensate the total slow CE phase drift that was introduced by the CPA, hollow fiber, and phase modulator. As shown in [Figure 3.43](#), the CE phase after the phase

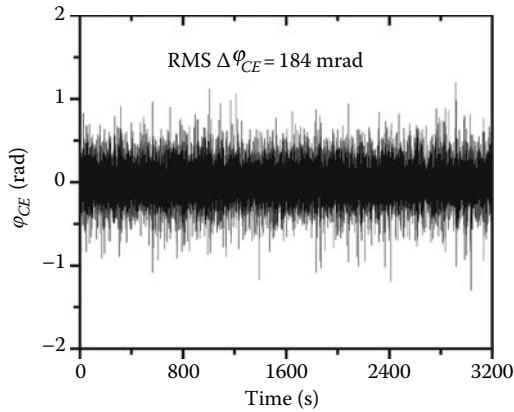


Figure 3.43 Long term stability of CE phase after the adaptive phase modulator. The CE phase was stabilized by feedback controlling the gratings in the stretcher, using the CE phase measured after the phase modulator to determine the error signal. (Reprinted with kind permission from Springer Science+Business Media: *Appl. Phys. B*, Carrier-envelope phase stabilization of 5-fs, 0.5-mJ pulses from adaptive phase modulator, 98, 2010, 291, H. Wang et al.)

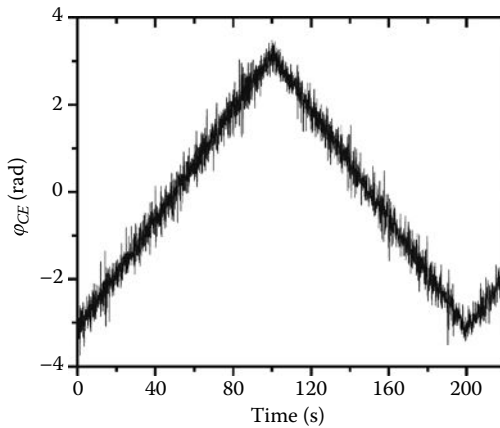


Figure 3.44 CEP sweep from $-\pi$ to π rad. (Reprinted with kind permission from Springer Science+Business Media: *Appl. Phys. B*, Carrier-envelope phase stabilization of 5-fs, 0.5-mJ pulses from adaptive phase modulator, 98, 2010, 291, H. Wang et al.)

modulator was locked with the accuracy of 180 mrad for nearly 1 h. Moreover, by adjusting the grating separation of the stretcher to preset values, the CE phase was swept from $-\pi$ to π , as shown in Figure 3.44. A FROG measurement was done after the adaptive pulse shaper, which shows that pulse duration is 5.1 fs. In conclusion, two-cycle, 0.5 mJ, CE phase-locked laser pulses can be obtained by combining hollow-core fibers with adaptive phase modulators.

3.11 Power Locking for Improving Carrier-Envelope Phase Stability*

As discussed in previous sections, 1% of laser energy fluctuation can cause more than 100 mrad error in CE phase measurements using f -to- $2f$ interferometers, which corresponds to 25 as time jitter between carrier oscillation and pulse envelope for Ti:Sapphire lasers. Under well-controlled environmental conditions, the laser energy fluctuation of typical kilohertz femtosecond laser systems is $>1\%$ RMS due to the pump laser energy fluctuation and other factors, which limits the accuracy of the CE phase control. Power stabilization is also important for studying nonlinear processes where the amount of products depends strongly on the laser intensity, such as high-order harmonic generation. A method has been demonstrated by the author's group to improve the laser power stability of multipass amplifiers by using the Pockels cell located between the oscillator and chirped pulse amplification system that is also used for pulse picking.

3.11.1 Feedback Loop

In the CPA laser system used to demonstrate the power locking, the femtosecond pulse train from a CE phase-stabilized oscillator, with a 77 MHz repetition rate, is sent to a Pockels cell to reduce the repetition rate to 1 kHz. The pulses passing through the Pockels cell pulse picker are stretched, amplified, and compressed, as shown in Figure 3.45. The final output after the grating compressor is 2 mJ, with 30 fs pulse duration. The Pockels cell that served as a pulse picker is also used as a power modulator to stabilize the final output energy.

Two power meters are used in the stabilization scheme. The in-loop power meter uses a Si photodiode as the power probe because of its fast response as compared to power meters based on thermal effects. It is positioned in the beam path of the zero-order diffraction from a compressor grating to take advantage of the otherwise wasted laser energy. There, the measured average power is proportional to the total output laser power. The electric pulse from the photodiode is amplified by the built-in amplifiers of the power meter.

The power fluctuations contain a broad noise spectrum, from DC to 1 kHz. It is difficult to get rid of all of them. The slow power variations are extracted by an external low-pass analog filter applied after the power meter. This signal was sent to a PID controller, which changed the high-voltage pulses applied to the Pockels cell to control its transmittance, i.e., the input power to the amplifier. In this way, the final output power is stabilized by the feedback loop. An output power meter determines the stability.

* This section is adapted from Wang, H., C. Li, J. Tackett, H. Mashiko, C.M. Nakamura, E. Moon, and Z. Chang, Power locking of high-repetition-rate chirped pulse amplifiers, *Appl. Phys. B: Lasers Opt.* 89, 275 (2007).

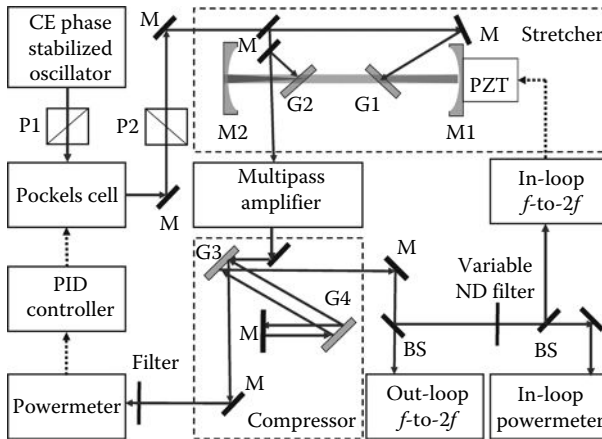


Figure 3.45 Power stabilization system. The in-loop power meter was put in the path of the zero-order diffraction beam and the power signal was sent to the PID controller. By using feedback control, the PID varied the voltage applied on the Pockels cell, which in turn changed the polarization of the output from the oscillator and stabilized the laser intensity. Solid arrows are the laser paths and dashed arrows represent electronic circuits. (Reprinted with kind permission from Springer Science+Business Media: *Appl. Phys. B*, Power locking of high-repetition-rate chirped pulse amplifiers, 89, 2007, 275, H. Wang, C. Li, J. Tackett, H. Mashiko, C.M. Nakamura, E. Moon, and Z. Chang.)

3.11.2 Pockels Cell

A Pockels cell is essentially a voltage-controlled wave plate. Potassium di-deuterium phosphate (KD*P) crystals are commonly used. It is placed between two orthogonally oriented polarizers, i.e., the transmission polarization directions of the polarizers are perpendicular to each other. The laser beam that comes out from the first polarizer cannot pass the second polarizer if there is no voltage applied across the Pockels cell. When a proper voltage called half-wave voltage is applied, it functions as a half-wave plate, which rotates the polarization by 90° . In this case, all laser power can pass the second polarizer. For other voltage values, the beam exiting the Pockels cell is elliptically polarized. Only a portion of the laser power can transmit the second polarizer. Obviously, the throughput depends on the voltage value applied on the Pockels cell.

When the Pockels cell–polarizer combination is used as a pulse picker in CPA systems, a pulse high voltage is applied on the cell. The width of the pulse is typically ~ 10 ns. Without applying power stabilization control, the amplitude of the 1 kHz high-voltage pulses applied to the Pockels cells is constant and is usually set at half-wave voltage, which generated a 1 kHz laser pulse train. In order to choose an appropriate working voltage range for the Pockels cell to compensate the laser power fluctuation, the relation between the voltage applied on the Pockels cell and the laser output power after the compressor is measured, as shown in [Figure 3.46](#).

In the 1–6 kV range, the output power of the CPA system increases with the voltage monotonically. The slope, however, reduces with the

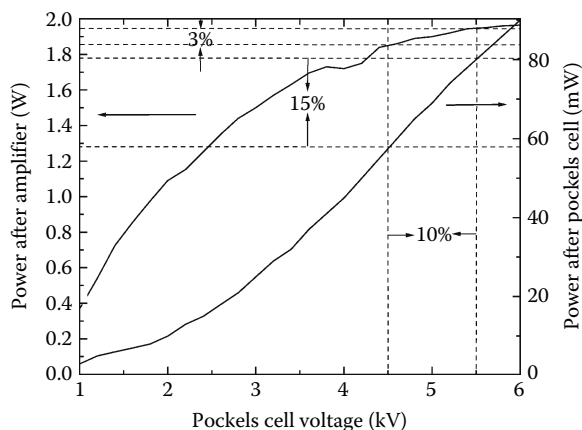


Figure 3.46 The output power versus Pockels cell voltage. The horizontal axis is the voltage applied on the Pockels cell, and the vertical axis is the power measured after Pockels cell and amplifier, respectively. (Reprinted with kind permission from Springer Science+Business Media: *Appl. Phys. B*, Power locking of high-repetition-rate chirped pulse amplifiers, 89, 2007, 275, H. Wang, C. Li, J. Tackett, H. Mashiko, C.M. Nakamura, E. Moon, and Z. Chang.)

voltage due to gain saturation in the Ti:Sapphire crystal. To have enough range for feedback control, the Pockels cell was set to work around 5 kV, which reduced the output power by 10%. Such a small power loss is acceptable. At this setting, 10% voltage adjustments could compensate for 3% laser power fluctuation, which is sufficient to compensate the power fluctuation of many lasers.

3.11.3 Power Stability

The measured laser power noise is shown in Figure 3.47. The vertical axis is the PSD in the $\text{dBV}/\sqrt{\text{Hz}}$ unit. dBV instead of V is used because of the large variation of the noise level in the whole spectral range. By optimizing the PID controller parameters and setting 500 Hz as the cutoff frequency of the low-pass filter, the power noise below 40 Hz is suppressed as evidenced in Figure 3.47. Without power locking, the power fluctuation is 1.33% RMS, as shown in Figure 3.48a. The fluctuation of the out-of-loop power drops to 0.28% RMS when the feedback control is turned on, as shown in Figure 3.48b, which shows the effectiveness of this feedback control scheme.

3.11.4 Carrier-Envelope Phase Stability

In order to investigate the effect of power stabilization on the CE phase locking, besides the in-loop f -to- $2f$ interferometer used to control the grating separation in the stretcher for the slow CE phase drift, an out-of-loop f -to- $2f$ interferometer is used to check the phase stability. Since the white-light generation and second harmonic generation processes in the

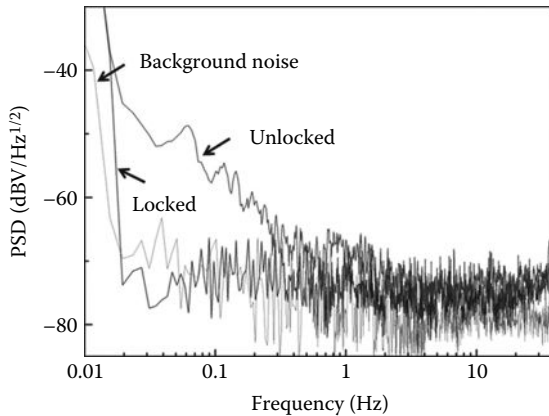


Figure 3.47 The PSD of the analog signal coming out of the power meter. After locking the power, the low-frequency (below 40 Hz) noise was suppressed. (Reprinted with kind permission from Springer Science+Business Media: *Appl. Phys. B*, Power locking of high-repetition-rate chirped pulse amplifiers, 89, 2007, 275, H. Wang, C. Li, J. Tackett, H. Mashiko, C.M. Nakamura, E. Moon, and Z. Chang.)

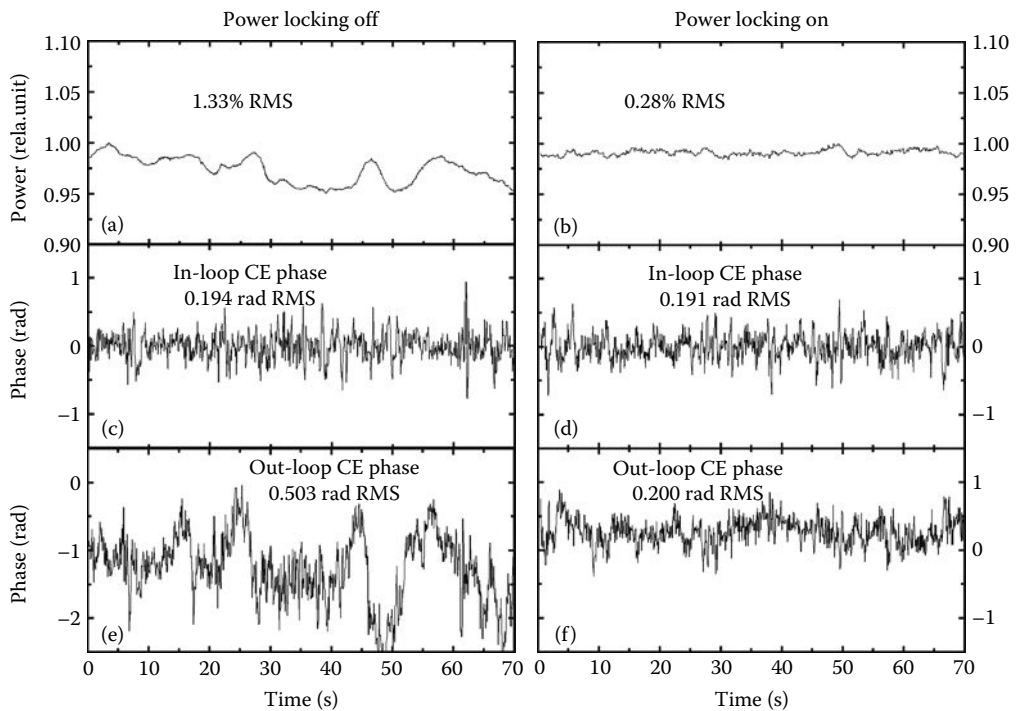


Figure 3.48 The left column and right column are the measurements of the laser power and CE phase stability with and without power locking. (a) and (b) show normalized power. After stabilization the power fluctuation decreases to one fifth of its usual value, (c) and (d) are in-loop CE phase and (e) and (f) are out-of-loop CE phase. (Reprinted with kind permission from Springer Science+Business Media: *Appl. Phys. B*, Power locking of high-repetition-rate chirped pulse amplifiers, 89, 2007, 275, H. Wang, C. Li, J. Tackett, H. Mashiko, C.M. Nakamura, E. Moon, and Z. Chang.)

two f -to- $2f$ interferometers are not identical, the CE phase measured by the two interferometers has different power dependencies. The correlation between power change and the out-of-loop CE phase shift can be seen by comparing Figure 3.48a and e.

The in-loop and the out-of-loop CE phases are measured concurrently with and without power locking. The in-loop f -to- $2f$ interferometer results do not show a significant difference when the power locking is turned on. This is expected. On the contrary, the out-of-loop measurements give very different results for the two cases. As shown in Figure 3.48, without power locking, the difference of the in-loop and out-of-loop standard deviation was 309 mrad. After locking the power, their difference dropped dramatically to 9 mrad, which demonstrated that the reduction of the power fluctuation could significantly improve CE phase stability.

3.12 Carrier-Envelope Phase Measurements with Above-Threshold Ionization

The f -to- $2f$ interferometers are suitable for measuring relative CE phase variations of either few-cycle or multi-cycle lasers. In many cases, the absolute value of the CE phase on the gas target for generating attosecond pulses is still unknown. But the amount of CE phase change on the target should be the same as that measured by the f -to- $2f$ technique. The major advantage of the f -to- $2f$ method is its simplicity. It is also an all-optical measurement.

For few-cycle linearly polarized laser pulses, the ionization probability in each half optical cycle depends on the CE phase. The absolute CE phase value can be determined by measuring the ratio of the ATI electrons emitted in opposite directions along the field of polarization. The device based on this principle is named stereo-ATI phasemeter, invented by Paulus and his colleagues.

Figure 3.49a shows the diagram of the phasemeter. The few-cycle pulse laser beam is focused into a Xe gas filled vacuum chamber. When the linearly polarized laser field ionizes the atoms, the counts of photo-ionized electrons are recorded by two microchannel plate (MCP) detectors. The ratio of the electron yields detected by left and right MCPs is used to represent the measured CE phase. Xe is chosen because its ionization potential is the lowest among all the noble gases and it requires the least amount of laser energy for the ionization to occur. The plateau of the ATI spectrum, which is used to measure the asymmetry ratio, is also flatter than other gases.

The electron ATI spectra are measured with two time-of-flight (TOF) spectrometers located on two sides of the laser-atom interaction region. In a TOF, the electron energy is measured by the time it takes to travel through a certain distance. The principle of electron TOF is discussed in detail in Chapter 7. As an example, the measured spectrum using the CE phase stabilized laser in the author's lab is shown in the intensity map in Figure 3.49c. Here, the ratio of electron yields detected by right (R) and left (L) MCP detectors $(L - R)/(L + R)$ is plotted. During the experiment,

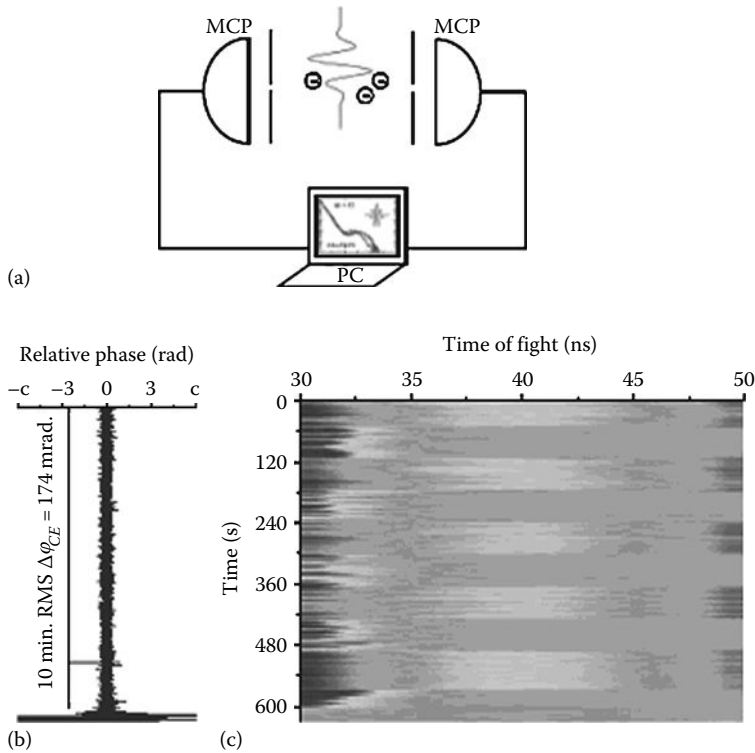


Figure 3.49 (a) The diagram of the stereo ATI phase meter. (b) 10 min locked CE phase results measured by f -to- $2f$ interferometer and (c) Time-of-flight spectrum measured by left and right MCPs in the phase meter. The CE phase was abruptly changed every 60 s by changing the thickness of the wedge plates. More electrons are detected by one of the MCPs or the other. (Reprinted from C. Li, E. Moon, H. Mashiko, C. Nakamura, P. Ranitovic, C.L. Cocke, Z. Chang, and G.G. Paulus, Precision control of carrier-envelope phase in grating based chirped pulse amplifiers, *Opt. Express*, 14, 11468, 2006. With permission of Optical Society of America.)

the relative CE phase of the CPA amplifier was stabilized by feedback controlling the grating separation in the stretcher of the CPA. The RMS jitter of CE phase was 174 mrad for 10 min, as shown in Figure 3.49b.

Every 60 s, the CE phase of the few-cycle laser pulse from a hollow-core and chirped mirror compressor was abruptly changed by π rad by moving a pair of fused silica glass wedge plates in the laser beam. It is evident that the electron spectrum is strongly correlated with the CE phase. Since the yield of high-energy plateau electrons is more sensitive than that of low-energy electrons to the CE phase, the spectrum shows a higher contrast in the range of short TOF (30–34 ns) than in the long time range (slow electrons) when the CE phase was changed by π . Absolute CE phase values can be assigned using the spectra, as demonstrated in other work by Paulus et al.

Problems

- 3.1 Plot the electric field of a cosine pulse with that of a sine pulse for 5 fs pulses centered at 800 nm.
- 3.2 Plot the electric fields of two circularly polarized Gaussian pulses with $\varphi_{CE} = 0$ and $\pi/2$ in the x - y plane as a function of time. Assume that the pulse duration is one laser cycle.
- 3.3 Plot the electric fields of two elliptically polarized Gaussian pulses with $\varphi_{CE} = 0$ and $\pi/2$ in the x - y plane as a function of time. Assume that the pulse duration is one laser cycle and the ellipticity is 0.5.
- 3.4 For the 2.5 fs pulse centered at 750 nm, calculate the ratio between the highest electric field peak and the adjacent peak. Compare it with 25 fs pulses.
- 3.5 Use the Sellmeier Equation of fused silica to calculate the CE phase of a pulse passing through a 1 mm window. The center wavelength is 750 nm.
- 3.6 Use the Sellmeier Equation of fused silica to calculate the CE phase of a pulse passing through a prism pair separated by 1 m. The center wavelength is 750 nm.
- 3.7 The repetition rate of a laser oscillator is 100 MHz. The CE phase changes $\pi/2$ from one pulse to the next in the train. What is the CE offset frequency?
- 3.8 The repetition rate of a laser oscillator is 100 MHz. The CE phase changes 0 rad from one pulse to the next in the train. What is the CE offset frequency?
- 3.9 Draw (a) the electric field of a cosine pulse with 5 fs FWHM centered and 800 nm and (b) a cosine pulse with 5 fs FWHM centered and 1600 nm. Compare the difference between the field strength in percentage at $\omega_0 t = 0$ and $\omega_0 t = \pm\pi$ for the two pulses.
- 3.10 Draw four pulses with 5 fs FWHM centered and 1600 nm making the CE phase change by $-\pi/2$ from one pulse to the next.
- 3.11 Calculate the phase velocity and the group velocity of fused silica at 800 nm.
- 3.12 Calculate the CE phase variation in 100 μm of Ti:Sapphire.
- 3.13 What is the 200 mrad CE phase error in the unit of degrees?
- 3.14 For an 800 nm laser pulse, what is the time jitter between the carrier wave and the pulse envelope for a 200 mrad CE phase error?
- 3.15 Suppose the fifth harmonic generation is a fifth order nonlinear process. When the laser intensity increases by 1%, how much does the harmonic intensity change?

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Semiclassical Model

High-order harmonic generation was discovered in 1987, although the mechanics was not revealed until 1993. In that year, Corkum and Kulan-der independently developed a semiclassical model that solved the mystery of the underlying physics of high harmonic generation. This model, for which, the full quantum treatment is provided in full in Chapter 5, laid the theoretical foundation for attosecond optics.

4.1 Three-Step Model

In the semiclassical model, an attosecond pulse is generated in three steps within one laser cycle. An artistic presentation of the model is shown in [Figure 4.1](#).

For simplicity, we consider the one-dimensional case and assume that the laser is a monochromatic light, linearly polarized in the x direction. Under these conditions, the laser field at a given spatial point can be expressed as

$$\varepsilon_L(t) = E_L \cos(\omega_0 t), \quad (4.1)$$

which is shown in [Figure 4.2](#). The subscript “0” in ω_0 specifies that the field oscillates with the fundamental frequency of the laser. We use the cosine form of the field and the initial phase is set to zero. To make the discussion applicable to lasers with a variety of center frequencies, we use normalized quantities: time is normalized to the laser period, T_0 , and the angular frequency to $\omega_0 = 2\pi/T_0$. For reference, a Ti:Sapphire laser has a period of $T_0 = 2.67$ fs.

Suppose an atom is located at $x = 0$. In the first step, the potential well of the atom where the electron is trapped is turning into a potential barrier by the laser field. The bound electron is freed (ionized) by tunneling through the barrier. This step is a quantum process, which is discussed in detail later in this chapter. We assume that the ionization is instantaneous, which means that the ionization rate at a given time only depends on the laser-field strength at that time. When the electron is freed, its initial position is also at $x = 0$. We further assume that the initial velocity of the electron is $v_0 = 0$.

In the second step, we assume the freed electron moves in the laser field and we neglect the Coulomb field of the atom. The electron is treated classically, which means that the equation of motion of a free electron is



Figure 4.1 The three-step model. (Reprinted from P.B. Corkum and Z. Chang, *Opt. Photon. News*, 19, 24, 2008.)

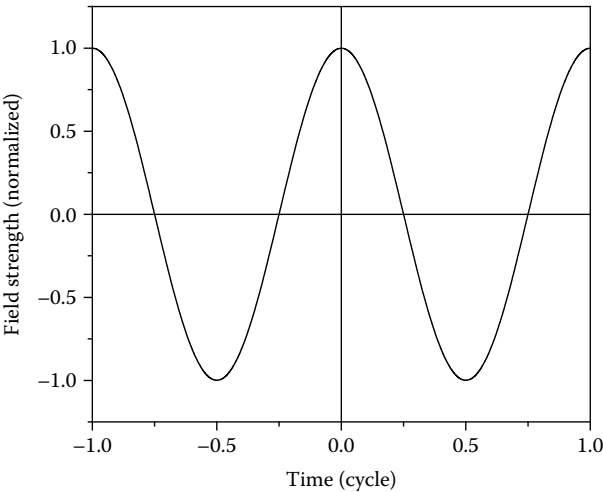


Figure 4.2 The laser field in the three-step model.

$$\frac{d^2x}{dt^2} = -\frac{e}{m_e} \varepsilon_L(t) = -\frac{e}{m_e} E_L \cos(\omega_0 t), \quad (4.2)$$

where $-e$ and m_e are the charge and mass of the electron, respectively.

Assuming an electron is freed at time t' , the solution of the equation is

$$v(t) = -\frac{eE_L}{m_e \omega_0} [\sin(\omega_0 t) - \sin(\omega_0 t')], \quad (4.3)$$

$$x(t) = \frac{eE_L}{m_e \omega_0^2} \{ [\cos(\omega_0 t) - \cos(\omega_0 t')] + \omega_0 \sin(\omega_0 t')(t - t') \}, \quad (4.4)$$

where v is the velocity of the electron.

We define $x_0 = 2eE_L/m_e \omega_0^2$. Equation 4.4 can be normalized to

$$\frac{x(t)}{x_0} = \frac{1}{2} [\cos(\omega_0 t) - \cos(\omega_0 t') + \sin(\omega_0 t') \omega_0 (t - t')], \quad (4.5)$$

The normalized electron trajectory for an electron released at $\omega_0 t' = 0$ is shown in Figure 4.3. The figure shows that x_0 is the maximum displacement of the electron ionized at $t' = 0$. The typical Ti:Sapphire laser intensity for attosecond pulse generation is $E_L = 5 \times 10^{14} \text{ W/cm}^2$, which gives $x_0 = 1.95 \text{ nm}$. This electron returns to the parent ion one cycle later. Electrons ionized at other times take different trajectories; some of them can return, and some drift away.

In the third step, the electron recombines with the parent ion at $x = 0$ and emits a photon. The emitted photon energy is

$$\hbar\omega_X(t) = I_p + \frac{1}{2}mv^2(t) = I_p + 2U_p[\sin(\omega_0 t) - \sin(\omega_0 t')]^2, \quad (4.6)$$

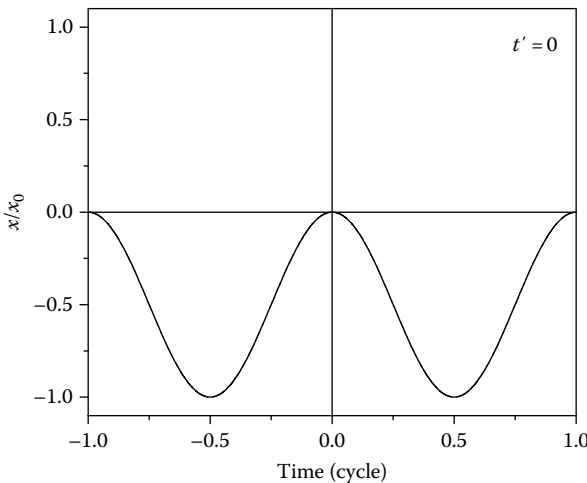


Figure 4.3 The trajectory of an electron in the laser field.

where I_p is the ionization potential of the atom and the ponderomotive energy, U_p , is given by

$$U_p = \frac{(eE_0)^2}{4m\omega_0^2}. \quad (4.7)$$

The electron ionized at $t' = 0$ returns with zero kinetic energy, and the corresponding photon energy is equal to I_p . This step is also a quantum process, which is discussed in the next chapter.

Several important features of high harmonic generation and attosecond pulses can be understood from the classical treatment of the electron in the second step and by using the energy conservation law. A few of these features in particular are the cutoff photon energy of the attosecond/high harmonic spectrum and the chirp of the attosecond pulses. Similar analyses can be applied to understand the energy spectra of the electrons freed by the above-threshold ionization and by the tunneling ionization.

4.1.1 Recombination Time

When the electron returns to the parent ion at time t , its position $x(t) = 0$. This is the time that the recombination occurs. The time t can be found by solving the equation

$$x(t) \propto \cos(\omega_0 t) - \cos(\omega_0 t') + \omega_0 \sin(\omega_0 t')(t - t') = 0. \quad (4.8)$$

4.1.1.1 Graphic Solutions and Kramers–Henneberger Frame

Equation 4.8 has no analytical solutions, but the solutions can be found out graphically, as shown in Figure 4.4. The equation can be rewritten as

$$\cos(\omega_0 t) - \cos(\omega_0 t') = \frac{d}{d(\omega_0 t)} \cos(\omega_0 t) \Big|_{t'} (\omega_0 t - \omega_0 t'). \quad (4.9)$$

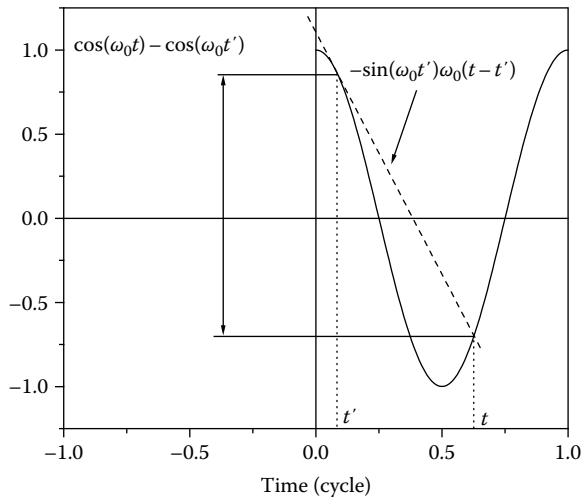


Figure 4.4 Graphic solution of the equation of motion.

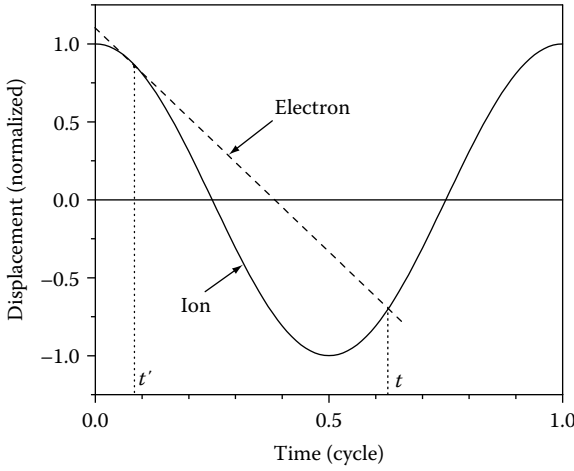


Figure 4.5 The normalized trajectories of the ion and electron in the Kramers–Henneberger frame.

First, we plot $\cos(\omega_0 t)$, then draw a straight line starting at $\cos(\omega_0 t')$ that is tangent to the $\cos(\omega_0 t)$ curve, i.e., the slope is $\frac{d}{d(\omega_0 t)} \cos(\omega_0 t) \Big|_{t'} = -\sin(\omega_0 t')$. If the straight line crosses the $\cos(\omega_0 t)$ curve at a later time t , then t is the recombination time for the electron releasing time t' .

The graphical approach corresponds to a reference frame called Kramers–Henneberger frame, as illustrated in Figure 4.5. In this reference frame, the parent ion is moving periodically such that its displacement follows $\cos(\omega_0 t)$ and the electron motion is a linear displacement. The slope of the line is $-\sin(\omega_0 t')$, just like in the case in Figure 4.4. In other words, the electron and ion motion in this frame provides physical meanings to the graphic method.

It can be seen that electrons emitted at time $\omega_0 t' = 0$ return one cycle (T_0) later $\omega_0 t = 2\pi$. $t - t' = T_0$ is the maximum roundtrip time that electron can have in the laser field. For $\omega_0 t' = \pi/2$, $\omega_0 t = \pi/2$, i.e., electron never leaves the parent ion. All electrons ionized in the time range $\omega_0 t' = 0$ to $\omega_0 t' = \pi/2$ can return to the parent ion. However, if an electron is released during $\omega_0 t' = \pi/2$ to $\omega_0 t' = \pi$, it will never return to the parent ion. Since the laser field is sinusoidal, electrons ionized during $\omega_0 t' = \pi/2$ to $\omega_0 t' = (3/2)\pi$ will return, but from an opposite direction as compared to an electron ionized during $\omega_0 t' = 0$ to $\omega_0 t' = \pi/2$.

4.1.1.2 Numerical Solutions and Fitting Functions

Equation 4.8 can also be solved numerically. The solution is shown in Figure 4.6.

The solution can be well fitted with a simple analytical function

$$\frac{t}{T_0} = \frac{1}{4} - \frac{3}{2\pi} \sin^{-1} \left(4 \frac{t'}{T_0} - 1 \right). \quad (4.10)$$

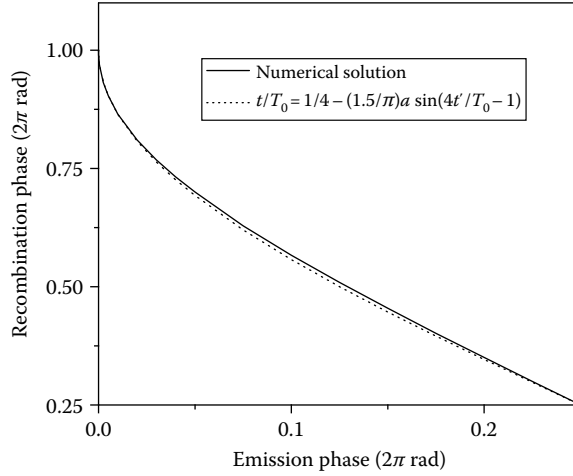


Figure 4.6 Dependence of the recombination time on the releasing time.

Or in terms of phases

$$\omega_0 t = \frac{\pi}{2} - 3 \sin^{-1} \left(\frac{2}{\pi} \omega_0 t' - 1 \right). \quad (4.11)$$

The fitting function is also shown in Figure 4.6 for comparison.

The fact that the returning time spans over 0.75 laser cycle suggests that the emitted electromagnetic pulse may last for $0.75T_0$ time period, which is 2 fs for Ti:Sapphire. The FWHM could be less than $0.75T_0 \times 0.5 = 1$ fs. This is the origin of the attosecond pulse generation.

4.1.2 Return Energy

The kinetic energy of the returning electron normalized by the ponderomotive energy is

$$\frac{K}{U_p} = 2[\sin(\omega_0 t) - \sin(\omega_0 t')]^2. \quad (4.12)$$

Inserting Equation 4.11 into Equation 4.12, we have

$$\frac{K(\omega_0 t')}{U_p} = 2 \left[\cos \left(3 \sin^{-1} \left(\frac{2}{\pi} \omega_0 t' - 1 \right) \right) - \sin(\omega_0 t') \right]^2. \quad (4.13)$$

The dependence of the kinetic energy on the emission time calculated using Equations 4.12 and 4.13 is shown in Figure 4.7, which was obtained by inserting the t and its corresponding t' in Figure 4.6 to Equation 4.10.

The maximum kinetic energy is

$$K_{\max} \approx 3.17 U_p, \quad (4.14)$$

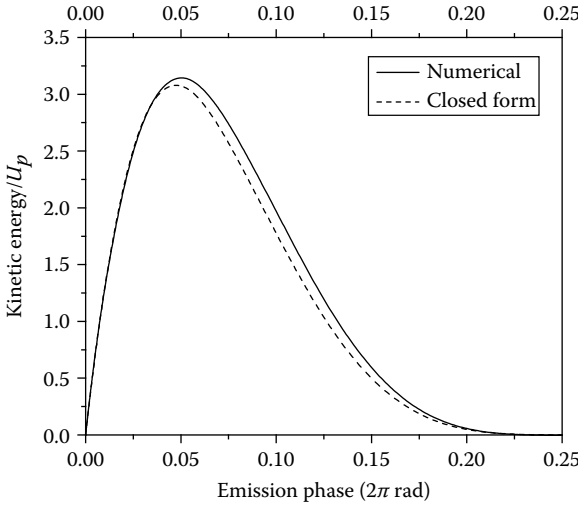


Figure 4.7 The kinetic energy of the returned electron.

which is carried by the electron released at $\omega_0 t' = 0.05 \times 2\pi$ rad and returns at $\omega_0 t = 0.7 \times 2\pi$. Thus, the maximum photon energy is

$$\hbar\omega_{X,\max} = I_p + 3.17U_p. \quad (4.15)$$

With this very important prediction of the semiclassical model, we can calculate the cutoff order of the high harmonic spectrum and the upper limit of the attosecond spectrum.

The ponderomotive energy can also be expressed as

$$U_p[\text{eV}] = 9.33 \times 10^{-14} I_L \lambda_0^2, \quad (4.16)$$

where I_L is the intensity of the laser in W/cm^2 . The unit of the laser wavelength is μm . Equations 4.15 and 4.16 suggest that the cutoff photon energy can be extended by using a long wavelength laser. This prediction has been confirmed by experiments.

For example, for argon atoms, $I_p = 15.78$ eV. The highest intensity of femtosecond lasers they can withstand can reach $3 \times 10^{14} \text{ W}/\text{cm}^2$. The corresponding ponderomotive energy is ~ 18 eV while the maximum photon energy can reach ~ 72 eV! This puts the radiation in the XUV range.

4.1.3 Long and Short Trajectories

Figure 4.7 shows that one electron released at time $t' < 0.05T_0$ can have the same kinetic energy as another one freed at time $t' > 0.05T_0$. An electron released earlier in time will return later in time, as shown in Figure 4.6, thus giving it a longer round trip time. It is for this reason that the trajectory of the electron released before $0.05T_0$ is called the “long-trajectory” electron while those released after $0.05T_0$ are called the “short-trajectory” electrons.

Experimentally, it is possible to suppress the long trajectory by phase matching and spatial filtering, which is discussed in Chapter 5. In that case, only the electrons in the short trajectory can arrive at the detector. The return time of the short trajectory extends from $t = 0.25T_0$ to $0.7T_0$, which is 1.2 fs, or a possible 600 as at FWHM. Therefore, it is possible to generate attosecond pulses by using the short trajectory. To estimate the XUV pulse width more accurately, we need to know the dependence of the ionization rate on the electron recombination time, which is discussed later in the chapter.

4.1.4 Chirp of Attosecond Pulses

Since the kinetic energy of the electron depends on the return time, the photon energy also changes with time, which is the origin of the chirp of attosecond pulses. The degree of the chirp, $d\omega_x(t)/dt \propto dK(t)/dt$, can be obtained from the equation

$$\frac{\hbar\omega_x(t) - I_p}{U_p} = \frac{K(t)}{U_p} = 2[\sin(\omega_0 t) - \sin(\omega_0 t')]^2, \quad (4.17)$$

Equation 4.11 can be rewritten as

$$\sin(\omega_0 t') = \cos\left[\frac{\pi}{2} \sin\left(\frac{1}{3}\omega_0 t - \frac{\pi}{6}\right)\right]. \quad (4.18)$$

Inserting Equation 4.18 into Equation 4.17 gives

$$\frac{K(t)}{U_p} = 2\left\{\sin(\omega_0 t) - \cos\left[\frac{\pi}{2} \sin\left(\frac{1}{3}\omega_0 t - \frac{\pi}{6}\right)\right]\right\}^2. \quad (4.19)$$

which is shown in Figure 4.8.

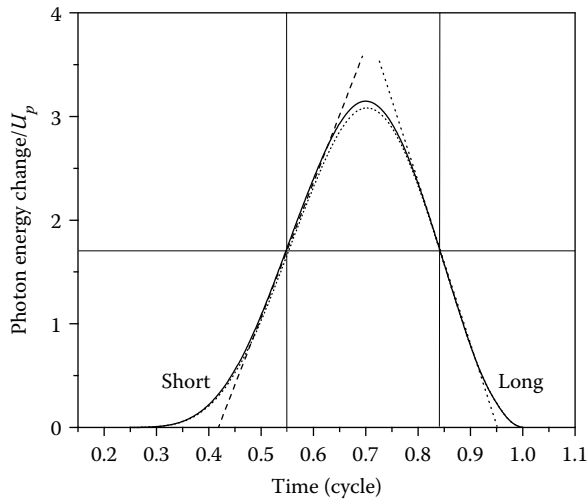


Figure 4.8 Chirp of the attosecond radiation.

Interestingly, the short trajectory is positively chirped, $dK(t)/dt > 0$, whereas the long trajectory is negatively chirped, $dK(t)/dt < 0$. The chirp, i.e., the slope of the curve in Figure 4.8, is almost linear over a broad photon energy range. The values of the chirp can be terminated by the slopes at $\hbar\omega_X(t) - I_p/U_p = 5.5$, which corresponds to $t = 0.55T_0$ for the short trajectory and $t = 0.85T_0$ for the long trajectory.

The slopes can be determined by

$$\frac{1}{U_p} \frac{d}{dt}(\hbar\omega_X) = 4[\sin(\omega_0 t) - \sin(\omega_0 t')] \times \left[\cos(\omega_0 t) - \cos(\omega_0 t') \frac{dt'}{dt} \right] \frac{2\pi}{T_0}. \quad (4.20)$$

Equation 4.20 can be rewritten in a dimensionless format as

$$\frac{T_0}{U_p} \frac{d}{dt}(\hbar\omega_X) = 8\pi[\sin(\omega_0 t) - \sin(\omega_0 t')] \times \left[\cos(\omega_0 t) - \cos(\omega_0 t') \frac{dt'}{dt} \right]. \quad (4.21)$$

Next, we can introduce an auxiliary function that corresponds to electron displacement

$$x(t, t') = \cos(\omega_0 t) - \cos(\omega_0 t') + \sin(\omega_0 t')(\omega_0 t - \omega_0 t'). \quad (4.22)$$

Using the theory of partial derivatives,

$$\frac{dt'}{dt} = \left(\frac{\partial t'}{\partial t} \right)_x = \frac{-\left(\frac{\partial x}{\partial t} \right)_{t'}}{\left(\frac{\partial x}{\partial t'} \right)_t} = \frac{\sin(\omega_0 t) - \sin(\omega_0 t')}{\cos(\omega_0 t')(\omega_0 t - \omega_0 t')}. \quad (4.23)$$

If Equation 4.23 is inserted into Equation 4.21, we have

$$\frac{T_0}{U_p} \frac{d}{dt}(\hbar\omega_X) = 8\pi[\sin(\omega_0 t) - \sin(\omega_0 t')] \times \left[\cos(\omega_0 t) - \frac{\sin(\omega_0 t) - \sin(\omega_0 t')}{\omega_0 t - \omega_0 t'} \right]. \quad (4.24)$$

The chirp is defined as

$$C = \frac{dt}{d(\hbar\omega_X)}, \quad (4.25)$$

Finally,

$$C(t, t') = -\frac{T_0}{U_p} \times \frac{\omega_0(t - t')}{8\pi[\sin(\omega_0 t) - \sin(\omega_0 t')][\sin(\omega_0 t) - \sin(\omega_0 t') - \cos(\omega_0 t)\omega_0(t - t')]}, \quad (4.26)$$

An explicit expression for the chirp as a function of return time can be obtained by using Equation 4.18, which gives

$$C(t) = \frac{T_0}{U_p} \times \frac{1}{4\pi K(t) \left\{ \cos(\omega_0 t) + \frac{\pi}{6} \cos\left(\frac{1}{3}\omega_0 t - \frac{\pi}{6}\right) \sin\left[\frac{\pi}{2} \sin\left(\frac{1}{3}\omega_0 t - \frac{\pi}{6}\right)\right] \right\}}. \quad (4.27)$$

4.1.4.1 Short Trajectory

The releasing time corresponding to $t = 0.55T_0$ is $t' = 0.107T_0$. Inserting them into Equation 4.24, we get

$$\frac{T_0}{U_p} \frac{d}{dt}(\hbar\omega_X) = 14.43. \quad (4.28)$$

Thus, the chirp is

$$C = \frac{dt}{d(\hbar\omega_X)} = 0.069 \frac{T_0}{U_p}. \quad (4.29)$$

To better understand the application of this concept, we can use an example for a Ti:Sapphire laser with a period of $T_0 = 2.67$ fs. When the intensity is 3×10^{14} W/cm², the chirp is $C = 10$ as/eV. The chirp is therefore inversely proportional to the laser intensity, thus $C = 30$ as/eV at 1×10^{14} W/cm².

In laser optics, the unit of the chirp is as², which can be calculated using the following conversion:

$$C[\text{as}^2] = \frac{c[\text{as/eV}]}{1.516} 10^3. \quad (4.30)$$

A typical chirp is 10 as/eV, or 6.6×10^3 as².

Equation 4.30 can also be expressed in terms of laser intensity and the center wavelength. Since $T_0 = \lambda_0/c$, we have

$$C[\text{as/eV}] = 24.7 \times 10^{14} \frac{1}{I_0 \lambda_0}, \quad (4.31)$$

where I_0 is in W/cm² and the unit of the laser wavelength is expressed in μm . It is clear that the chirp can be reduced by either increasing the laser intensity or using a long wavelength laser. The chirp can be compensated by materials that have negative group velocity dispersion, as is discussed in Chapter 1.

4.1.4.2 Long Trajectory

The releasing time corresponding to $t = 0.85T_0$ is $t' = 0.0123T_0$. The chirp then becomes

$$C = -0.059 \frac{T_0}{U_p}. \quad (4.32)$$

The chirp is negative, meaning that the high-energy photons are emitted before the low-energy ones. The magnitude of the chirp is a little less than that of the short trajectory.

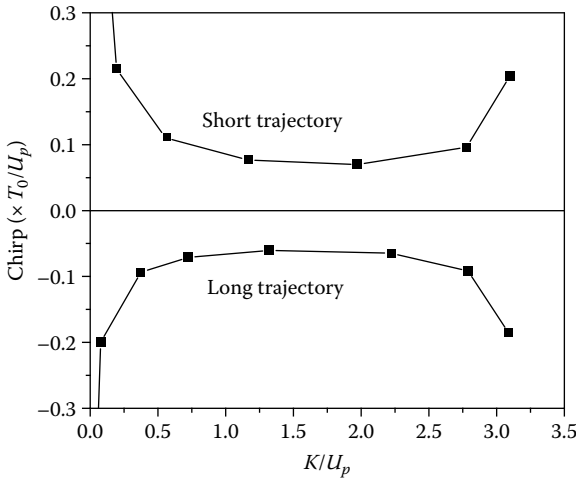


Figure 4.9 The chirp in unit of T_0/U_p as function of K/U_p .

4.1.4.3 The General Case

Equations 4.28 through 4.32 are used in the spectral region where the chirp is close to linear. In general, the attosecond chirp can be obtained using Equation 4.26. The dependence of the chirp on the kinetic energy is shown in Figure 4.9. It is worthwhile to mention that this dependence is not a function of the ionization potential of the atom.

4.1.4.4 High-Order Chirp

The graph in Figure 4.9 shows that the chirp is not a constant near the cutoff of the spectrum. This is partly due to the fact that in the cutoff region the third-order phase and other high-order phases start to show up.

The third-order chirp, TOC, is defined as

$$\text{TOC} = \frac{d}{d\omega_x} \left(\frac{dt}{d\omega_x} \right), \quad (4.33)$$

which is the slope of the curve in Figure 4.9.

4.2 Tunneling Ionization and Multiphoton Ionization

In the three-step model of attosecond pulse generation, the first step is the ionization of an atom by the laser field. The ionization rate is needed to determine the temporal profile of the electron pulse returning to the parent ion, which is related to the attosecond pulse duration. Simple analytic solutions of the ionization rate have been found under various approximations. The rate expressions have been frequently used in order to avoid solving Schrödinger Equation numerically.

4.2.1 The Keldysh Theory

Rare gases are commonly used to generate attosecond pulses because they can withstand high laser intensity. Their ionization potentials are in the range of 12.1–24.5 eV, which is much larger than the photon energy of the NIR laser (<2 eV). In the 1960s, Keldysh developed a very important theory for one-electron atom photoionization by strong lasers when the laser photon energy is much smaller than the ionization potential. Using a first-order perturbation theory, he derived the photoionization formula for the direct transition between the electronic ground state and the Volkov continuum state, which includes oscillatory motion of the free electron in the time-dependent linearly polarized electric field. The intermediate resonance states were not taken into consideration by this theory. Neglecting the resonance bound states as well as the effects Coulomb potential on the continuum states is named strong field approximation.

Subsequently, Faisal considered an S -matrix theory in which the initial bound state is dressed by the laser field and the final ionization state is taken to be noninteracting. Later, Reiss established a rigorous basis for an extended version of the Keldysh theory in which systematic higher-order corrections can be applied to the Keldysh term. Combined, these are well known as the so-called Keldysh–Faisal–Reiss theory.

4.2.1.1 Volkov States

The Schrodinger Equation for an atom in a laser field is given by

$$i\hbar \frac{d}{dt} \Psi(\vec{r}, t) = [H_0 + H_I] \Psi(\vec{r}, t), \quad (4.34)$$

where

H_0 is the contribution to the total Hamiltonian from the atom

$H_I(t)$ is from the laser field

Far from the atom, $H_0 = 0$, the effects of the Coulomb field of the atom can be ignored. The equation is simplified to

$$i\hbar \frac{d}{dt} \psi(\vec{r}, t) = H_I(t) \psi(\vec{r}, t). \quad (4.35)$$

We use $\vec{A}$ to denote the vector potential of the electromagnetic field, and use ϕ to denote the scalar potential. In classic mechanics, the Hamiltonian of an electron in the light field is

$$\begin{aligned} H_c &= \frac{1}{2m_e} \left(\vec{p} + \frac{e}{c} \vec{A} \right)^2 - e\phi \\ &= \frac{1}{2m_e} p^2 + \frac{e}{2m_e c} \left(\vec{A} \cdot \vec{p} + \vec{p} \cdot \vec{A} \right) + \frac{e^2}{2m_e c^2} A^2 - e\phi, \end{aligned} \quad (4.36)$$

where $\vec{p}$ is the canonical momentum. In quantum mechanics, the canonical momentum is replaced by the momentum operator expressed by

$$H_I = \frac{1}{2m_e} \left(\hat{p} + \frac{e}{c} \vec{A} \right)^2 - e\phi, \quad (4.37)$$

where $\hat{p} = -i\hbar\nabla$ is the canonical momentum operator. Apparently

$$H_I = \frac{1}{2m_e} \hat{p}^2 + \frac{e}{2m_e c} (\vec{A} \cdot \hat{p} + \hat{p} \cdot \vec{A}) + \frac{e^2}{2m_e c^2} A^2 - e\phi, \quad (4.38)$$

In general, the $\vec{A}$ and $\hat{p}$ are not commutable, $\hat{p} \cdot \vec{A} - \vec{A} \cdot \hat{p} = -i\hbar \nabla \cdot \vec{A}$. In the Coulomb gauge, $\nabla \cdot \vec{A} = 0$, $\hat{p} \cdot \vec{A} = \vec{A} \cdot \hat{p}$.

Since the Coulomb potential is ignored and there is no other static field, thus $\phi = 0$. Therefore,

$$H_I = \frac{1}{2m_e} \hat{p}^2 + \frac{e}{m_e c} \vec{A} \cdot \hat{p} + \frac{e^2}{2m_e c^2} A^2. \quad (4.39)$$

Assuming the laser is a monochromatic plane wave that is linearly polarized along the direction defined by the unit vector $\vec{\varepsilon}$, the vector potential becomes

$$\vec{A}(t) = -\vec{\varepsilon} A_0 \sin(\omega t), \quad (4.40)$$

and the corresponding electric field is

$$\vec{\mathcal{E}}(t) = \vec{\varepsilon} F \cos(\omega t). \quad (4.41)$$

Here, F is the amplitude maximum of the incident linearly polarized electric field.

The solutions are eigenstates of the canonical momentum with eigenvalue $\vec{p} = \vec{p}_m - \frac{e}{c} \vec{A}(t)$, which is equal to the average drift momentum of the corresponding classical electron. Here $\vec{p}_m$ is the mechanical momentum of the electron. The solution can be expressed as

$$\psi_V(\vec{r}, t) = e^{i\hbar \left[\left[\vec{p}_m - \frac{e}{c} \vec{A}(t) \right] \cdot \vec{r} - \frac{p_m^2}{2m_e} \int_0^t dt' \left[\vec{p}_m - \frac{e}{c} \vec{A}(t') \right]^2 \right]}. \quad (4.42)$$

Using Equation 4.40, we get

$$\begin{aligned} \psi_V(\vec{r}, t) = e^{i\hbar \left[\vec{p}_m \cdot \vec{r} - \frac{p_m^2}{2m_e} t - U_p t \right]} & \sum_{n=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} J_m \left(\frac{e^2 A_0^2}{8\hbar \omega m_e c^2} \right) \\ & \times J_{n-2m} \left(|\vec{\varepsilon} \cdot \vec{p}_m| \frac{e A_0}{\hbar \omega m_e c} \right) e^{in\omega t}. \end{aligned} \quad (4.43)$$

The J_m are m th order Bessel functions of the first kind and the eigenstates are called Volkov states which are plane electron waves, $e^{i(1/\hbar)[\vec{p}_m \cdot \vec{r} - (p_m^2/2m_e)t - U_p t]}$, with many discrete frequency components $e^{in\omega t}$. The spacing between the electron frequency comb is the laser frequency ω while the Bessel functions determine the amplitude of each component. Since $n = -\infty, \dots, \infty$, the negative frequency space is also covered.

4.2.1.2 Fermi's Golden Rule and Photoionization Rate

Fermi's golden rule is a way to calculate the transition rate (probability of transition per unit time) from one energy eigenstate of a quantum system into a continuum of energy eigenstates, due to a perturbation H_I . It is valid when the initial state has not been significantly depleted by scattering into the final states. We consider an atom to begin in an eigenstate Ψ_i of a given Hamiltonian H_0 . Consider the situation where $A^2 \ll e/m_e c \vec{A} \cdot \hat{p}$,

the transition rate (transition probability per unit time) from an initial state Ψ_i to a final state Ψ_f is given, to first order approximation in the perturbation, by

$$w \propto \frac{2\pi}{\hbar} |\langle \Psi_f | H_I | \Psi_i \rangle|^2 \propto \langle \Psi_f | \vec{A} \cdot \vec{p} | \Psi_i \rangle. \quad (4.44)$$

From the Quantum Mechanic relations, we have

$$\begin{aligned} \langle \Psi_f | \vec{A} \cdot \vec{p} | \Psi_i \rangle &\propto \langle \Psi_f | \vec{A} \cdot \nabla V(\vec{r}) | \Psi_i \rangle \propto \langle \Psi_f | \vec{A} \cdot \vec{r} | \Psi_i \rangle \\ &\propto \langle \Psi_f | \vec{\mathcal{E}}(t) \cdot \vec{r} | \Psi_i \rangle. \end{aligned} \quad (4.45)$$

Quantitatively, the rate of photoionization,

$$w = \frac{1}{(2\pi\hbar)^3} \int d^3v \frac{d}{dT} |c_{\vec{v}}(T)|^2 \Big|_{T \rightarrow \infty}, \quad (4.46a)$$

where $\vec{v}$ denotes the momentum of the freely ionizing electron

$$c_{\vec{v}}(T) = \frac{i}{\hbar} \int_0^T dt \langle \psi_{\vec{v}}(\vec{r}, t) | e\vec{r} \cdot \vec{\mathcal{E}} F \cos(\omega t) | \psi_g(\vec{r}) e^{-\frac{i}{\hbar} E_g t} \rangle. \quad (4.46b)$$

The wave function $\psi_g(\vec{r})$ represents the initial electron ground state with binding energy $-E_g$. while $\psi_{\vec{v}}(\vec{r}, t)$ is the final continuum state. In the Keldysh theory, $\psi_{\vec{v}}(\vec{r}, t) = \psi_V(\vec{r}, t)$.

4.2.1.3 Keldysh Parameter

Keldysh introduced a dimensionless parameter to categorize different ionization mechanisms. In atomic units, the Keldysh parameter is expressed as

$$\gamma = \frac{\omega}{F} \sqrt{2I_p} = \sqrt{\frac{I_p}{2U_p}}, \quad (4.47)$$

where

$I_p = -E_g$ is the ionization potential of an atom

ω is the laser frequency

F is the laser field strength

The ponderomotive potential

$$U_p = \frac{F^2}{4\omega^2} \quad (4.48)$$

is the cycle average kinetic energy of an electron in the laser field. The γ parameter measures the ratio between the tunneling time t_{tu} , i.e., the time needed for an electron to cross the Coulomb barrier, and the time during which the same barrier is lowered by the laser field,

$$\gamma = \frac{2\pi}{T_0} \frac{\sqrt{2I_p}}{F} = \frac{t_{tu}}{T_0}. \quad (4.49)$$

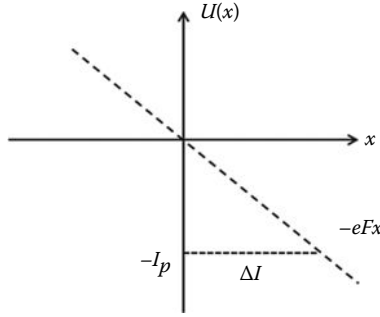


Figure 4.10 The potential barrier formed by the laser field.

In estimating the tunneling time, we assume that the electron velocity inside the barrier is the same as that in the ground state. Under this assumption, the tunneling time becomes

$$t_{tu} = \frac{\Delta I}{v_{gr}}, \quad (4.50)$$

where

ΔI is the width of the barrier

v_{gr} is the velocity of the electron in the ground state

As shown in Figure 4.10, the barrier width can then be given as

$$\Delta I = \frac{I_p}{F}. \quad (4.51)$$

Since

$$\frac{1}{2} m_e v_{gr}^2 = I_p, \quad (4.52)$$

the electron velocity in atomic unit is

$$v_{gr} = \sqrt{2I_p}. \quad (4.53)$$

Thus,

$$t_{tu} = \frac{1}{2} \frac{\sqrt{2I_p}}{F}. \quad (4.54)$$

The Keldysh parameter is defined as

$$\gamma = \frac{t_{tu}}{T_0/2} = \frac{1}{T_0} \frac{\sqrt{2I_p}}{F}. \quad (4.55)$$

In an elliptically polarized field with ellipticity, ξ , the ponderomotive potential is

$$U_p = \frac{e^2 F^2}{4m_e \omega^2} (1 + \xi^2) = \frac{2\pi e^2}{c\omega^2} I. \quad (4.56)$$

Multiphoton ionization of atoms in a laser field occurs when $\gamma \gg 1$. In this case, electrons are pulled out of the atom by absorption of several photons.

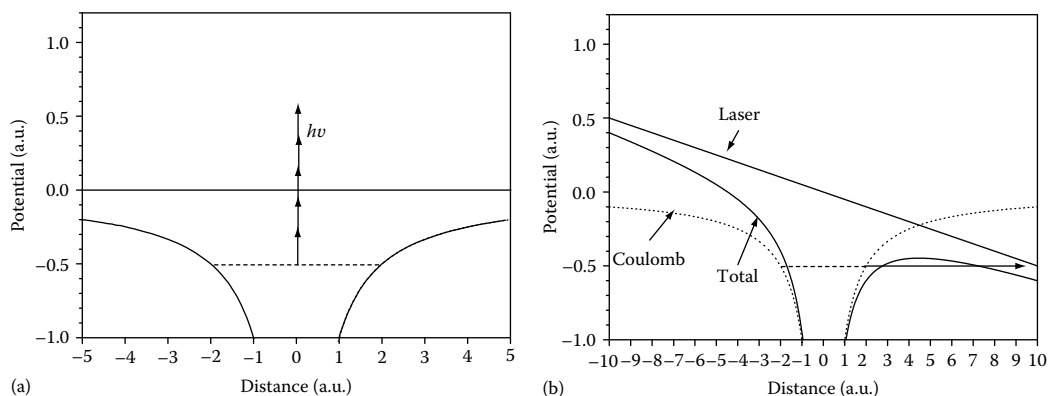


Figure 4.11 (a) Multiphoton ionization. (b) Tunneling ionization.

Tunneling ionization happens when $\gamma \ll 1$, where the electron leaves the atomic core by passing through the Coulomb barrier lowered by the laser field. The ionization rate can be calculated by the Perelomov, Popov, and Terent'ev (PPT) model for both types of ionization. The rate can also be calculated by a simpler Ammosov–Delone–Krainov (ADK) Equation in the tunneling regime, given that the rates from the two models are identical for $\gamma \ll 1$. The multiphoton ionization and the tunneling ionization process are shown in Figure 4.11.

4.2.2 PPT Model

PPT obtained a formula for the actual three-dimensional atoms in the short-range potentials. Their three-dimensional photoionization rate formulas are applicable for hydrogen atoms with arbitrary initial ground states of orbital angular momentum l and magnetic quantum number m . The theory was extended to other types of atoms and ions by the ADK Theory.

A neutral atom A can be ionized by absorbing q photons, leaving an ion with charge Z . The process can be described by $A + q\omega = A^{+Z} + Ze$. For single ionization, $Z=1$. The PPT model was derived for a short-range potential and includes the effect of the long-range Coulomb interaction through the first-order correction in the quasiclassical action, which does not consider any discrete binding states other than the ground state.

In atomic units, the PPT model gives the following total rate of ionization:

$$w_{PPT}(F, \omega) = \sum_{q \geq q_{thr}}^{\infty} w_q(F, \omega), \quad (4.57)$$

where $q_{thr} = \lceil (I_p + U_p)/\omega \rceil$ is the minimum number of photon required to ionize an electron through the multiphoton process, or, the ionization threshold. $I_p + U_p$ is the effective ionization potential considering the AC Stark shift and $\lceil x \rceil$ denotes the ceiling function.

We can assume that the electron is in the state, n, l, m , before the field arrives. Here, n is the principal quantum number, l is the orbital quantum

number, and m is the magnetic quantum number, respectively. In atomic unit, the rate of ionization by absorbing q photons is

$$w_q(F, \omega) = A_q(\omega, \gamma) |C_{n^* l^*}|^2 G_{lm} I_p \left(\frac{2F_0}{F} \right)^{2n^*} \times \left(\frac{2F_0}{F} \frac{1}{\sqrt{1 + \gamma^2}} \right)^{-|m|-1} \frac{4}{\sqrt{3\pi}} \frac{1}{|m|!} \frac{\gamma^2}{1 + \gamma^2} e^{-\frac{2F_0}{3F} g(\gamma)}, \quad (4.58)$$

where

$$A_q(\omega, \gamma) = e^{-\alpha(q-\nu)} w_m \left[\sqrt{\beta(q-\nu)} \right], \quad (4.59)$$

$$\nu = \frac{I_p}{\omega} \left(1 + \frac{1}{2\gamma^2} \right) = \frac{I_p + U_p}{\omega}, \quad (4.60)$$

$$\alpha(\gamma) = 2 \left[\sinh^{-1} \gamma - \frac{\gamma}{\sqrt{1 + \gamma^2}} \right], \quad (4.61)$$

$$\beta(\gamma) = \frac{2\gamma}{\sqrt{1 + \gamma^2}}, \quad (4.62)$$

$$w_m(x) = \frac{x^{2|m|+1}}{2} \int_0^1 \frac{e^{-x^2 t} t^{|m|}}{\sqrt{1-t}} dt. \quad (4.63)$$

We can also write the rate as

$$w_{PPT}(F, \omega) = |C_{n^* l^*}|^2 G_{lm} I_p \left(\frac{2F_0}{F} \right)^{2n^* - |m| - 1} \left(\frac{1}{\sqrt{1 + \gamma^2}} \right)^{-|m| - 1} \times \frac{4}{\sqrt{3\pi}} \frac{1}{|m|!} \frac{\gamma^2}{1 + \gamma^2} e^{-\frac{2F_0}{3F} g(\gamma)} \sum_{q \geq q_{thr}}^{\infty} A_q(\omega, \gamma), \quad (4.64)$$

where $(2F_0/F)^{2n^*}$ is the correction of the long-range Coulomb interaction given that the Coulomb field is expressed as $F_0 = (2I_p)^{3/2}$ and the Keldysh number is $\gamma = \omega (F_0^{1/3}/F)$.

The effective principal quantum number $n^* \equiv Z/\sqrt{2I_p}$, and the effective orbital quantum number is $l^* = n^* - 1$. The three coefficients are

$$|C_{n^* l^*}|^2 = \frac{2^{2n^*}}{n^* \Gamma(n^* + l^* + 1) \Gamma(n^* - l^*)}, \quad (4.65)$$

$$G_{lm} = \frac{(2l+1)(l+|m|)!}{2^{|m|} |m|! (l-|m|)!}, \quad (4.66)$$

$$g(\gamma) = \frac{3}{2\gamma} \left[\left(1 + \frac{1}{2\gamma^2} \right) \sinh^{-1}(\gamma) - \frac{\sqrt{1 + \gamma^2}}{2\gamma} \right]. \quad (4.67)$$

The value of $\sum_{q \geq q_{thr}}^{\infty} A_q(\omega, \gamma)$ can be obtained approximately by setting the upper limit of the sum to $q_{max} - \nu = 10/\alpha(\gamma)$. For even larger values

of q , the contribution to the sum can be neglected because $e^{-\alpha(q-\nu)}$ is much smaller than that of the leading terms.

The unit of the rate w_{PPT} is the number of electrons per atomic unit of time, which corresponds to $41.341w_{PPT}/\text{fs}$. F is also in atomic unit, which can be calculated from the intensity in W/cm^2 by

$$F[\text{a.u.}] = \sqrt{\frac{I[\text{W}/\text{cm}^2]}{3.55 \times 10^{16}}}. \tag{4.68}$$

Noble gases are commonly used for attosecond pulse generation. Except for helium, the outmost orbital of all four other atoms is the p configuration, whereas it is s for He, as shown in Table 4.1. The outmost electrons are ionized first because of their smaller ionization potential. For example, for the neon atom, the ionization potentials of the $2p$ and the $2s$ electron are 21.57 and 48.48 eV, respectively. For the p orbital, the state with $m = 0$ has larger rate than $m = \pm 1$ (see Problem 4.10).

The parameters $|C_{n^*l^*}|^2$ and G_{lm} of the rare gases are listed in Table 4.2. Here, the state that has the highest ionization rate is shown.

The calculated ionization rate of helium by a laser centered at 790 nm is shown in Figure 4.12. In the intensity range of $3 \times 10^{13} - 3.5 \times 10^{14} \text{ W}/\text{cm}^2$, it agrees well with the results from the numerical solution of the Schrödinger Equation, in the limit $I \rightarrow 0$, $w = \sigma I^q$, where σ is the cross section, q is the minimum number of photons required to free an electron from an atom. In this case, at low intensities, $q = 7 \sim 8$. The contributions of different orders to the rate are plotted in Figure 4.13, at three different intensities.

TABLE 4.1 The Electron Configuration of Noble Gases

	Electron Configuration	I_p (eV)	l	m
He	$1s^2$	24.587	0	0
Ne	$1s^2 2s^2 p^6$	21.564	1	0
Ar	$1s^2 2s^2 p^6 3s^2 p^6$	15.759 ($J=3/2$)	1	0
		15.936 ($J=1/2$)		
Kr	$1s^2 2s^2 p^6 3s^2 p^6 d^{10} 4s^2 4p^6$	13.999 ($J=3/2$)	1	0
		14.665 ($J=1/2$)		
Xe	$1s^2 2s^2 p^6 3s^2 p^6 d^{10} 4s^2 p^6 d^{10} 5s^2 p^6$	12.129 ($J=3/2$)	1	0
		13.436 ($J=1/2$)		

TABLE 4.2 The ADK Parameters

	F_0 (a.u.)	n^*	l^*	l	m	$ C_{n^*l^*} ^2$	G_{lm}
He	2.42946	0.74387	−0.25613	0	0	4.25575	1
Ne	1.99547	0.7943	−0.2057	1	0	4.24355	3
Ar	1.24665	0.92915	−0.07085	1	0	4.11564	3
Kr	1.04375	0.98583	−0.01417	1	0	4.02548	3
Xe	0.84187	1.05906	0.05906	1	0	3.88241	3

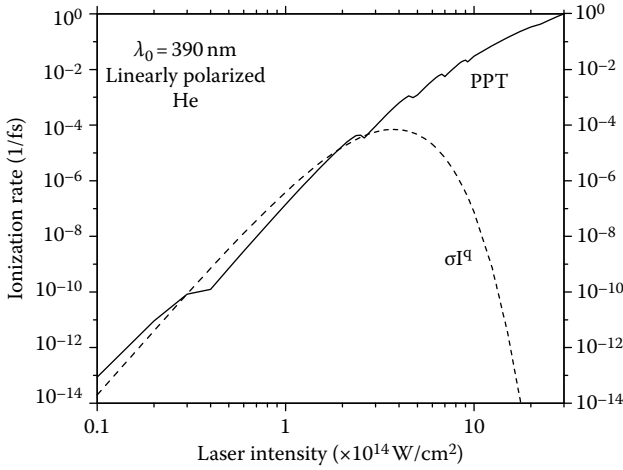


Figure 4.12 PPT rate of helium.

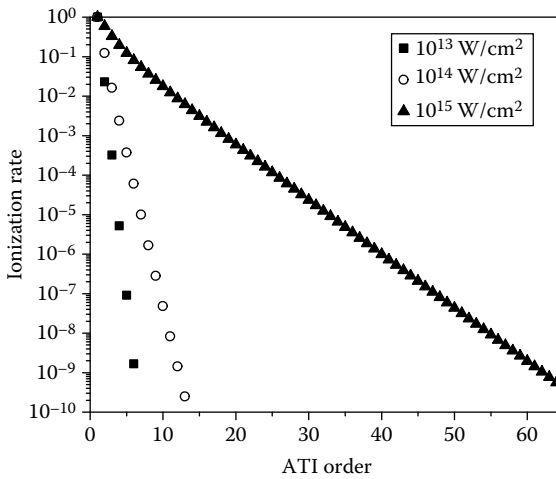


Figure 4.13 Contribution of the first three terms.

4.2.3 ADK Model

Ammosov, Delone, and Krainov derived the expressions for the tunnel ionization probabilities of arbitrary complex atoms and atomic ions. Their theory is essentially an extension of the PPT theory. They took into account that the states of the complex atoms are characterized by the effective principal and orbital quantum numbers. The effective principle quantum number is $n^* \equiv Z/\sqrt{2I_p}$, and the effective orbital quantum number is $l^* = n^* - 1$.

In the tunneling regime, $\gamma \ll 1$, we have $(1/\sqrt{1+\gamma^2})^{-|m|-1} \approx 1$, $\sum_{q \geq q_{thr}}^\infty A_q(\omega, \gamma) \approx 1$, and $g(\gamma) \approx 1$. The ionization can be calculated by the ADK rate by the expression

$$w_{ADK} = |C_{n^*l^*}|^2 G_{lm} I_p \left(\frac{2F_0}{F} \right)^{2n^* - |m| - 1} e^{-\frac{2F_0}{3F}}. \quad (4.69)$$

When $\gamma \gg 1$, the main difference between the PPT and the ADK rate is caused by the $g(\gamma)$ because it is in the exponent of the PPT rate. As concluded by Ilkov et al., use of the ADK theory should be limited to regions of intensity where γ is smaller than 0.5.

4.2.3.1 Cycle-Averaged Rate

For a linearly polarized laser field with amplitude F_a and $2F_0/3F_a \gg 1$, the cycle-averaged rate is

$$\begin{aligned} \bar{w}_{ADK}(F_a) &= \frac{1}{T_0} \int_0^{T_0} w_{ADK}(t) dt \\ &\approx |C_{n^*l^*}|^2 G_{lm} I_p \left(\frac{2F_0}{F_a} \right)^{2n^* - |m| - 1} \frac{2}{\pi} \int_0^{\pi/2} e^{-\frac{2F_0}{3F_a |\cos(x)|}} dx. \end{aligned} \quad (4.70)$$

The integral

$$\begin{aligned} \int_0^{\pi/2} e^{-\frac{2F_0}{3F_a |\cos(x)|}} dx &\approx \int_0^{\infty} e^{-\frac{2F_0}{3F_a} \cosh(x)} dx = K_0 \left(\frac{2F_0}{3F_a} \right) \\ &\approx \sqrt{\frac{\pi}{2}} \frac{1}{\sqrt{\frac{2F_0}{3F_a}}} e^{-\frac{2F_0}{3F_a}}. \end{aligned} \quad (4.71)$$

K_0 in Equation 4.65 is the zero order modified Bessel function of the second kind. The averaged rate therefore becomes

$$\bar{w}_{ADK}(F_a) = \sqrt{\frac{2}{\pi}} \sqrt{\frac{3F_a}{2F_0}} w_{ADK}(F_a). \quad (4.72)$$

Since $\sqrt{2/\pi} \sqrt{3F_a/2F_0} \ll 1$, the cycle averaged rate is much smaller than a field with constant strength. Thus, in one laser cycle, the ionization probability of a linearly polarized laser is much lower than the circularly polarized field.

4.2.3.2 Cycle-Averaged Rate of an Elliptically Polarized Field

An elliptically polarized field can be written as

$$\vec{F}(t) = F_a [\hat{i} \cos(\omega t) + \hat{j} \xi \sin(\omega t)]. \quad (4.73)$$

When the ellipticity $\xi < 1$ and $1 - \xi \gg F_a/F_0$, it was shown by PPT that

$$\bar{w}_{ADK}(F_a) = \sqrt{\frac{1}{1 - \xi^2}} \sqrt{\frac{2}{\pi}} \sqrt{\frac{3F_a}{2F_0}} w_{ADK}(F_a). \quad (4.74)$$

For a linearly polarized field $\xi = 1$, Equation 4.74 becomes Equation 4.72.

4.2.3.3 Saturation Ionization Intensity

The ionization probability of a given atom is determined by the quantum state of the electron, and the laser parameters, which can be calculated from the ionization rate by the integral

$$p = 1 - e^{-\int_{-\infty}^{+\infty} w(t)dt}. \quad (4.75)$$

Figure 4.14 shows the probability as a function of peak intensity of several gases. Attosecond pulse generation has been done at the intensity range of 10^{14} W/cm².

To compare with measured ion yield, the theoretical ion curve is calculated by taking into account the spatial and temporal dependence of the intensity, and consequently the ionization rates in the focal spot region, which is called volume effect.

4.2.4 Attosecond Electron and Photon Pulses

4.2.4.1 Returning Electron Pulse

The shape of the electron pulses that returns the parent ion is related to the ionization rate at the time t' and its corresponding returning time. The relative ADK rate then becomes

$$w(t') = \frac{w_{ADK}(t')}{|C_{n^*l^*}|^2 G_{lm} I_p} = \left[\frac{2F_0}{F|\cos(\omega t')|} \right]^{2n^* - |m| - 1} e^{-\frac{2F_0}{3F|\cos(\omega t')|}}. \quad (4.76)$$

As an example, for $F_0/F = 100$, $n^* = 1$, $m = 0$, the ADK rate is

$$w(t') = \frac{200}{|\cos(\omega t')|} e^{-\frac{200}{3|\cos(\omega t')|}}. \quad (4.77)$$

The relative rate in the 0–1 cycle range is shown in Figure 4.15. It can be seen that the ionization rate for the long trajectory $0 < t' < 0.05T_0$ is much

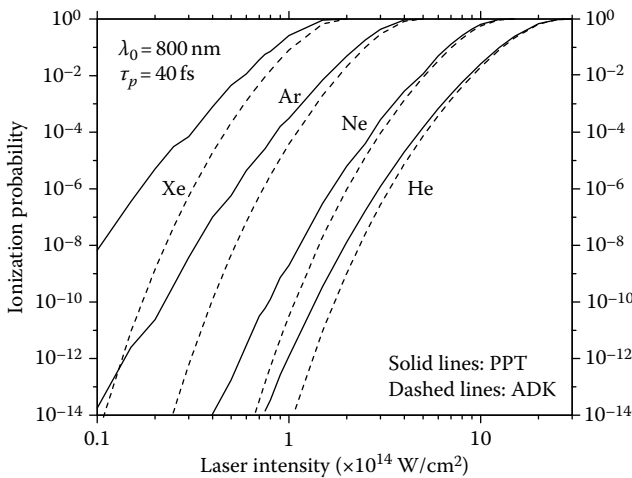


Figure 4.14 Ionization probabilities as a function of peak intensity.

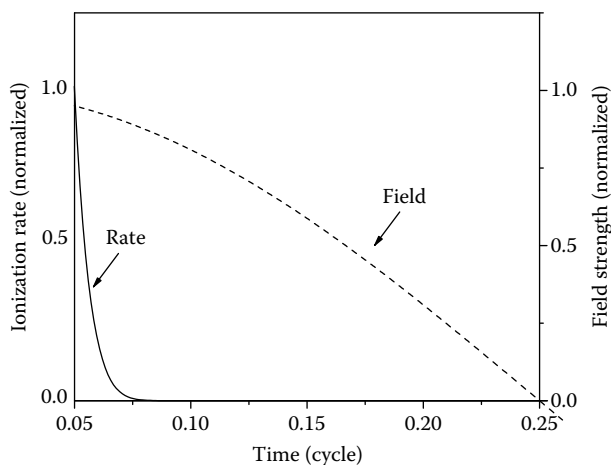


Figure 4.15 The ionization rate for the short trajectory.

larger than that of the short trajectory $0.05T_0 < t' < 0.25T_0$. In attosecond experiments, only the short trajectory contributes to the useful photons. The ionization that produces the long trajectory and also the electrons that do not return consume some of the ground-state population, which is not desirable, but unavoidable.

The ionization rate for the short trajectory is shown in Figure 4.15. The ionization rate reduces by 1000 times when t' changes from $0.05T_0$ to $0.1T_0$. Thus, the electrons generated in the range $0.1T_0 < t' < 0.25T_0$ can be neglected. The returning time range corresponding to $0.05T_0 < t' < 0.1T_0$ is $0.57T_0 < t < 0.7T_0$, which corresponds to 350 as for Ti: Sapphire lasers, which is an indication of the attosecond XUV pulse duration.

4.2.4.2 Attosecond Pulse Train and High-Order Harmonics

Figure 4.16 shows that the ionization rate has two maxima in each laser cycle, which indicates that there a pair of attosecond electron pulses are released per laser cycle. When they return and recombine with the parent ion, two light pulses are emitted. This process repeats from one laser cycle to the next, which results in an attosecond pulse train if the laser pulse contains many optical cycles. The spacing between the two adjacent attosecond pulses is half of a laser cycle. The interference between the pulses in the frequency domain leads to the high-order harmonic peaks.

4.3 Cutoff Photon Energy

Cutoff photon energy is one of the most important parameters in high harmonic and attosecond pulse generations. The highest photon energy achievable when the returning electron recombines with the parent ion is determined by the maximum kinetic energy of the electron as it returns to the nucleus

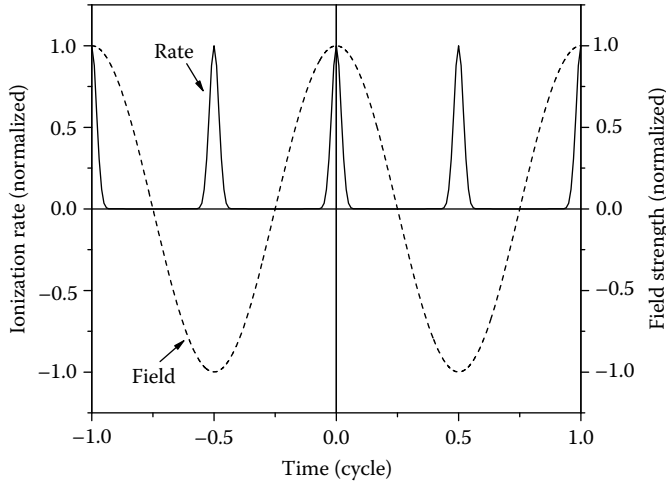


Figure 4.16 The ionization rate in a cosine laser field.

$$\hbar\omega_c = I_p + 3.17U_p, \quad (4.78)$$

where

U_p is the ponderomotive energy

I_p is the ionization potential of the atom

Since the ponderomotive energy is proportional to the laser intensity, this equation seems to indicate that the cutoff photon energy can be increased to any desired value with enough high laser intensity. Unfortunately, this is not true. It is also wrong to think that the cutoff photon energy depends linearly on the ionization potential as the equation indicates.

The light emission from recombination is a quantum process. The coherent XUV radiation is generated by the superposition of the returning electron wave and the ground state electron wave. The ground-state population should not be depleted completely when the freed electron wave returns, which is discussed in detail in Chapter 5. For a pulse laser field, ground-state population can be completely depleted by a portion of the leading edge as depicted in Figure 4.17, leaving no population for the peak portion of the laser pulse. This means that for a given type of atom, there is a maximum laser intensity that ground state can experience. Such intensity is called the saturation intensity, as introduced in Section 4.2.4.

When the depletion of the ground-state population by ionization is taken into account, the above equation becomes

$$\hbar\omega_c = I_p + 3.17U_p(I_S), \quad (4.79)$$

where I_S is the saturation intensity for the given atom and laser parameters.

4.3.1 Saturation Field and Intensity

In the tunneling regime, a simple equation is derived to predict $\hbar\omega_c$ for given experimental conditions, which shows the explicit dependence of

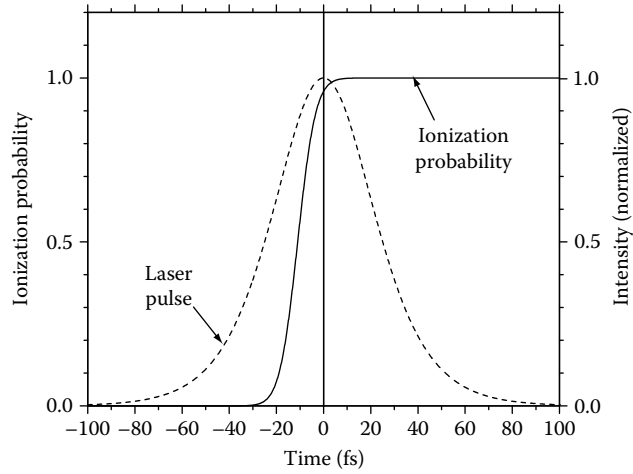


Figure 4.17 Depletion of the ground-state population by the leading edge of the laser pulse.

the cutoff photon energy on the atomic and laser parameters. Assuming that the atom is ionized by the leading edge of a linearly polarized laser pulse, we first derive the analytical expression of the saturation intensity, I_s .

4.3.1.1 Sech Square Pulse

For mathematical simplicity, we assume that the shape of the laser field is a hyperbolic secant function peaked at $t=0$. The field envelope is expressed as

$$F(t) \propto \text{sech}\left(1.76 \frac{t}{\tau_p}\right), \quad (4.80)$$

where τ_p is the FWHM of the intensity envelope. Here, we use F instead of $E(t)$ to be consistent with the literatures. Such a pulse is called a sech square pulse in the laser community because for many applications the primary concern is about the intensity profile, which is the square of the electric field. The field envelope of a sech pulse and a Gaussian pulse is shown in Figure 4.18 for comparison. The fields near the peak are very close to each other. Since the ionization occurs mostly near the peak of the pulse, the results we obtain with the sech pulse can also be applied to Gaussian pulses.

4.3.1.2 Definition of Ionization Saturation

To find out the saturation intensity for the case where the atom is almost fully ionized by the leading edge of the pulse, we calculate the ionization probability by the first half of the laser pulse. The ionization probability is

$$p(t) = 1 - p_0(t), \quad (4.81)$$

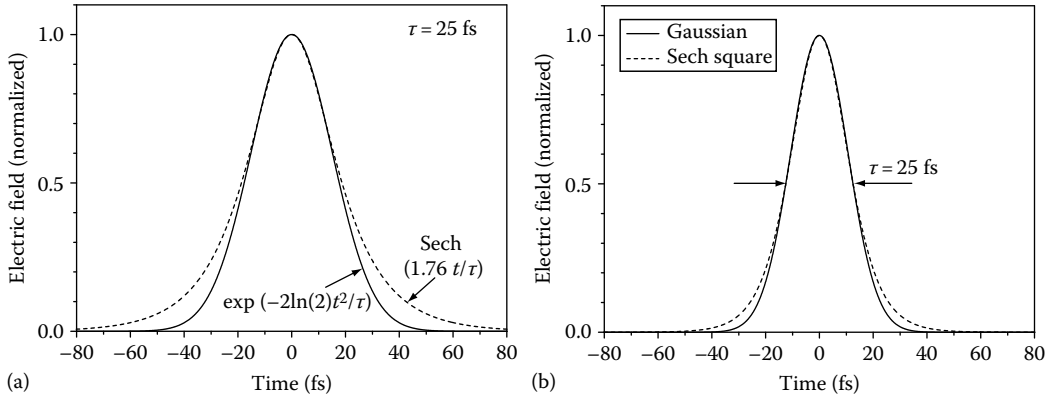


Figure 4.18 Comparison between a sech square pulse and a Gaussian pulse. (a) Field. (b) Intensity.

where p_0 is the ground-state population. The ground-state population and the ionization rate are related by the rate equation by

$$\frac{dp_0}{dt} = -w(t)p_0, \quad (4.82)$$

where $w(t)$ is the ionization rate determined by the instantaneous laser field strength. Consequently, the population at the time of the intensity peak is

$$p_0(t=0) = e^{-\int_{-\infty}^0 w(t)dt}. \quad (4.83)$$

Here, we define the saturation intensity, I_s , at which the ionization probability is $p_s = p(t=0) = 1 - p_0(t=0) = 0.98$. In other words, there are only 2% ground-state populations left for generating attosecond pulses after the peak of the pulse, which can be neglected.

4.3.1.3 ADK Rate

In the tunneling regime, the ADK rate can be used. It can be expressed in the form

$$w_{ADK} = \rho \left(\frac{1}{F_1} \right)^{g+1} e^{-\frac{1}{F_1}}, \quad (4.84)$$

where $\rho = |C_n^*|^2 G_{lm} \frac{2l}{2} 3^{2n^*-|m|-1}$, $g = 2n^* - |m| - 2$, $F_1 = \frac{3F}{2F_0}$.

To perform the integral in Equation 4.83, we case Equation 4.84 into the form

$$w_{ADK} = \rho \left(-\frac{\partial}{\partial \beta} \right)^g \frac{1}{F_1} e^{-\frac{\beta}{F_1}}, \quad (4.85)$$

where β is dummy variable that is set to one at the end of calculation.

4.3.1.4 Circularly Polarized Pulses

Although high harmonics cannot be generated with circularly polarized lasers, it is easy to perform the integral in Equation 4.83 because the field

strength is the same as the field envelope. In other words, there are no fast oscillation terms in the integral. It turns out that the expressions for the saturation intensity of the linearly polarized field is similar to that of the circularly polarized laser.

The quantity, F_1 , related to the field strength can be expressed as

$$F_1(t) = \frac{3F(t)}{2F_0} = F_{10} \operatorname{sech}\left(1.76 \frac{t}{\tau_p}\right). \quad (4.86)$$

The integral is therefore

$$\begin{aligned} & \int_{-\infty}^0 w(t) dt \\ &= \rho \int_{-\infty}^0 \left(-\frac{\partial}{\partial \beta}\right)^g \frac{1}{F_{10} \operatorname{sech}\left(1.76 \frac{t}{\tau_p}\right)} e^{-\frac{\beta}{F_{10} \operatorname{sech}\left(1.76 \frac{t}{\tau_p}\right)}} dt \\ &= \rho \frac{\tau}{1.76} \left(-\frac{\partial}{\partial \beta}\right)^g \frac{1}{F_{10}} \int_{-\infty}^0 \frac{1}{\operatorname{sech}\left(1.76 \frac{t}{\tau_p}\right)} e^{-\frac{\beta}{F_{10} \operatorname{sech}\left(1.76 \frac{t}{\tau_p}\right)}} d\left(1.76 \frac{t}{\tau_p}\right) \\ &= \rho \frac{\tau}{1.76} \left(-\frac{\partial}{\partial \beta}\right)^g \left[\frac{1}{F_{10}} K_1\left(\frac{\beta}{F_{10}}\right) \right]. \end{aligned} \quad (4.87)$$

K_1 in Equation 4.87 is the first order modified Bessel function of the second kind. For the field used in tunneling ionization $1/F_{10} = \beta/F_{10} \gg 1$, we have

$$K_1\left(\frac{\beta}{F_{10}}\right) \approx \sqrt{\frac{\pi}{2}} \frac{1}{\sqrt{\frac{\beta}{F_{10}}}} e^{-\frac{\beta}{F_{10}}}. \quad (4.88)$$

$K_1(x)$ and its asymptotic expression are plotted in Figure 4.19.

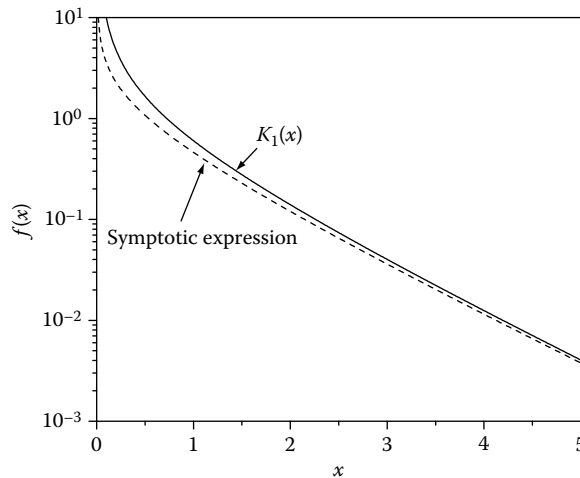


Figure 4.19 Graph of $K_1(x)$ and its asymptotic expression.

Since

$$-\frac{d}{dx} \left(\frac{1}{\sqrt{x}} e^{-x} \right) = \left(\frac{1}{2x} + 1 \right) \frac{1}{\sqrt{x}} e^{-x} \approx \frac{1}{\sqrt{x}} e^{-x}, \quad x \gg 1, \quad (4.89)$$

We have

$$\left(-\frac{\partial}{\partial \beta} \right)^g \left(\frac{1}{\sqrt{\frac{\beta}{F_{10}}}} e^{-\frac{\beta}{F_{10}}} \right) = \frac{1}{F_{10}} \frac{1}{\sqrt{\frac{\beta}{F_{10}}}} e^{-\frac{\beta}{F_{10}}}. \quad (4.90)$$

Set $\beta = 1$, the integral

$$\int_{-\infty}^0 w(t) dt \approx |C_{n^* l^*}|^2 G_{lm} I_p 3^{2n^* - |m| - 1} \frac{\tau_p}{1.76} \sqrt{\frac{\pi}{2}} \frac{1}{F_{10}^{3/2}} e^{-\frac{1}{F_{10}}}. \quad (4.91)$$

Thus,

$$-\ln(1 - p_s) = |C_{n^* l^*}|^2 G_{lm} I_p 3^{2n^* - |m| - 1} \frac{\tau_p}{1.76} \sqrt{\frac{\pi}{2}} \frac{1}{F_{10}^{3/2}} e^{-\frac{1}{F_{10}}}, \quad (4.92)$$

which can be expressed as

$$F_{10} = \frac{1}{\ln \left[\frac{|C_{n^* l^*}|^2 G_{lm} I_p 3^{2n^* - |m| - 1} \frac{\tau_p}{1.76} \sqrt{\frac{\pi}{2}} \frac{1}{F_{10}^{3/2}}}{-\ln(1 - p_s)} \right]}. \quad (4.93)$$

By definition

$$F_{10} = \frac{3F_s}{2F_0} = \frac{3F_s}{2(2I_p)^{\frac{3}{2}}}, \quad (4.94)$$

where F_s is field strength corresponding to the saturation intensity, I_s .

In the denominator, we make an approximation: $F_{10} = 1$. The saturation field peak amplitude can then be expressed as

$$F_s = \frac{\frac{2}{3}(2I_p)^{\frac{3}{2}}}{\ln \frac{|C_{n^* l^*}|^2 G_{lm} I_p 3^{2n^* - |m| - 1} \frac{\tau_p}{1.76} \sqrt{\frac{\pi}{2}}}{-\ln(1 - p_s)}}. \quad (4.95)$$

It is important to keep in mind the quantities in this equation are in atomic units.

4.3.1.5 Linearly Polarized Fields

Attosecond pulse cannot be generated by circularly polarized laser because the returned electron misses the parent ion in such a laser field, which is discussed later in this chapter. For linearly polarized laser, we use

the cycle-averaged ionization rate to calculate the saturation intensity, which is

$$\bar{w}_{ADK}(F_{10}) = \sqrt{\frac{2}{\pi}} \sqrt{F_{10}} w_{ADK}(F_{10}). \quad (4.96)$$

Following the same procedure as the case of circular polarization, we can get the saturation field when the atom is ionized by a linearly polarized laser

$$F_s = \frac{\frac{2}{3}(2I_p)^{\frac{3}{2}}}{\ln \frac{|C_{n^*l^*}|^2 G_{lm} I_p 3^{2n^*-|m|-1} \frac{\tau_p}{1.76} \frac{1}{F_{10}}}{-\ln(1-p_s)}}. \quad (4.97)$$

Again, making the approximation $F_{10} = 1$ in the denominator, we have

$$F_s = \frac{\frac{2}{3}(2I_p)^{\frac{3}{2}}}{\ln \frac{|C_{n^*l^*}|^2 G_{lm} I_p 3^{2n^*-|m|-1} \frac{\tau_p}{1.76}}{-\ln(1-p_s)}}. \quad (4.98)$$

4.3.1.6 Saturation Intensity for Linearly Polarized Field

The atomic unit of intensity is defined as that at which the laser field amplitude is one atomic unit. Thus, the saturation intensity in atomic unit is

$$I_s = F_s^2 = \frac{\frac{32}{9} I_p^3}{\left[\ln \frac{|C_{n^*l^*}|^2 G_{lm} I_p 3^{2n^*-|m|-1} \frac{\tau_p}{1.76}}{-\ln(1-p_s)} \right]^2}. \quad (4.99)$$

For experimentalists, it is more convenient to use W/cm^2 for intensity. One atomic unit of intensity corresponds to $3.55 \times 10^{16} \text{ W}/\text{cm}^2$. One commonly uses eV for ionization potential, and fs for pulse duration. In such units, the saturation intensity becomes

$$I_s = \frac{6.27 I_p^3}{\left[\ln \frac{0.86 |C_{n^*l^*}|^2 G_{lm} I_p 3^{2n^*-|m|-1} \tau_p}{-\ln(1-p_s)} \right]^2} \times 10^{12} \text{ W}/\text{cm}^2. \quad (4.100)$$

This equation is derived by making the approximation $F_{10} = 1$ in the denominator of the saturation field. It can be modified slightly to have better agreement to the saturation intensity calculated by integrating the ADK rate numerically. The modified expression is

$$I_s = \frac{1.7 I_p^{3.5}}{\left[\ln \frac{0.86 |C_{n^*l^*}|^2 G_{lm} I_p 3^{2n^*-|m|-1} \tau_p}{-\ln(1-p_s)} \right]^2} \times 10^{12} \text{ W}/\text{cm}^2. \quad (4.101)$$

4.3.1.7 Ionization Probability

p_s is the ionization probability at the peak of pulse ($t=0$), which is important for attosecond and high harmonic generation. For some other applications, we need to know the ionization probability at the end of pulse ($t=\infty$), which is

$$p = 1 - e^{-\int_{-\infty}^{\infty} w(t) dt}. \quad (4.102)$$

For a linearly polarized laser, the rate integral in atomic units is

$$\begin{aligned} \int_{-\infty}^{\infty} w(t) &= \frac{2}{1.76} \tau_p |C_n^* t^*|^2 G_{lm} I_p 3^{2n^*-|m|-1} \frac{1}{F_{10}} e^{-\frac{1}{F_{10}}} \\ &= 1.14 \tau_p \frac{(3F_{10})^{2n^*-|m|-1}}{F_{10}} W_{ADK}(F_{10}). \end{aligned} \quad (4.103)$$

Thus

$$p_L(F_{10}) = 1 - e^{-1.14 \tau_p \frac{(3F_{10})^{2n^*-|m|-1}}{F_{10}} W_{ADK}(F_{10})}, \quad (4.104)$$

where

$$F_{10} = \frac{3F_a}{2F_0} = \frac{3F_a}{2(2I_p)^{\frac{2}{3}}}. \quad (4.105)$$

Equation 4.104 can be revised to Equation 4.106 by fitting the results from the numerical integration of the ionization rate,

$$p_L(F_{10}) = 1 - e^{-1.14 \tau_p 3^{2n^*-|m|-1} F_{10}^{2.41} W_{ADK}(F_{10})}. \quad (4.106)$$

The comparison of the ionization probability calculated by the PPT, ADK, and Equation 4.106 is shown in Figure 4.20. It can be seen that the

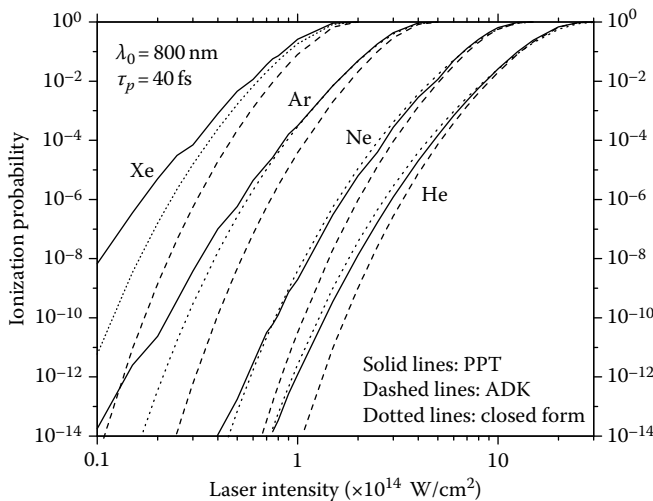


Figure 4.20 Ionization probability calculated with three expressions.

probability calculated by Equation 4.106 is closer to the PPT results, which is more precise than the ADK results. This equation allows us to estimate the ionization probability without performing the integral numerically.

It is also interesting to compare Equation 4.106 with that corresponding to a square pulse with duration τ , which is

$$p_L(F_{10}) = 1 - e^{-\tau_p \bar{W}_{ADK}(F_{10})}. \quad (4.107)$$

For circularly polarized laser pulse, the rate integral in atomic units is

$$\begin{aligned} \int_{-\infty}^{\infty} w(t) dt &= \frac{2}{1.76} \tau_p |C_{n^* l^*}|^2 G_{lm} I_p 3^{2n^* - |m| - 1} \sqrt{\frac{\pi}{2}} \frac{1}{F_{10}^{3/2}} e^{-\frac{1}{F_{10}}} \\ &= 1.42 \tau_p \frac{(3F_{10})^{2n^* - |m| - 1}}{F_{10}^{3/2}} W_{ADK}(F_{10}). \end{aligned} \quad (4.108)$$

By fitting the results with numerical integration of the ionization rate, we obtain the revised expression

$$p(F_{10}) = 1 - e^{-1.42 \tau_p 3^{2n^* - |m| - 1} F_{10}^{1.91} W_{ADK}(F_{10})}. \quad (4.109)$$

4.3.2 Cutoff due to Depletion of the Ground State

To calculate the cutoff photon energy of the high harmonic or attosecond pulse spectrum set by the depletion of the ground state, we substitute

$$I_s = \frac{1.7 I_p^{3.5}}{\left[\ln \frac{0.86 |C_{n^* l^*}|^2 G_{lm} I_p 3^{2n^* - |m| - 1} \tau_p}{-\ln(1 - p_s)} \right]^2} \times 10^{12} \text{ W/cm}^2. \quad (4.110)$$

into

$$\hbar\omega_c = I_p + 3.17 U_p(I_s), \quad (4.111)$$

where $U_p(I_s) = 9.33 \times 10^{14} I_s \lambda^2$. Finally, we obtain an expression for the cutoff photon energy

$$\hbar\omega_c = I_p + \frac{0.5 I_p^{3.5} \lambda^2}{\left[\ln \left(\frac{0.86 I_p 3^{2n^* - 1} G_{lm} C_{n^* l^*}^2 \tau_p}{-\ln(1 - p_s)} \right) \right]^2}, \quad (4.112)$$

where p_s is the ionization probability at the peak of the pulse that defines the saturation of the ionization of the ground-state population, which can be set to 0.98. The unit of $\hbar\omega_c$ and I_p is eV, λ is μm , and τ_p is fs. The values of n^* , G_{lm} , and $C_{n^* l^*}^2$ associated with the quantum parameters of the ground state are found in Table 4.2. Equation 4.112 shows the explicit dependence of the cutoff photon energy on the atomic and laser

parameters. Most importantly, it includes the effects of the ground-state depletion by the leading edge of the driving laser pulse.

To increase the cutoff, we can use atoms or ions that have high ionization potential, or lasers with short pulse duration and long wavelength. The $\hbar\omega_c$ value also determines the shortest attosecond pulses that can be generated.

4.3.2.1 Ionization Potential

Equation 4.112 indicates that the cutoff photon energy should be proportional to approximately the cube of the ionization potential of the atom, i.e., $I_p^{3.5}$. This is because, for a given laser-pulse duration, it is much harder to deplete the ground state of the atoms with a larger ionization potential. As the ADK theory revealed, the ionization rate is an exponential function of the ionization potential.

Experimentally, using 25 fs lasers centered at 800 nm, harmonics up to 29, 41, 61, and 155 have been generated Xe, Kr, Ar, and Ne, with corresponding ionization potentials of 12.13, 13.99, 15.76, and 21.56 eV, respectively, as shown in Figure 4.21. Using Equation 4.112, we can predict that harmonics up to the order of 27, 41, 61, and 163 should be observed from Xe, Kr, Ar, and Ne, respectively. The simple calculations and experimental observations are, therefore, in very good agreement. The measured cutoff order can be lower than the predicted value due to plasma induced defocusing and phase mismatch, which are not included when Equation 4.112 is derived. Phase matching is discussed in Chapter 6. In the experiments, the gas pressure was kept at very low to minimizing the plasma effects.

Ions have even higher ionization potential, and thus it is possible to generate even higher harmonic order from ions. However, preparing pure ion targets (not a plasma) with high density is not as easy as it is for neutral ones.

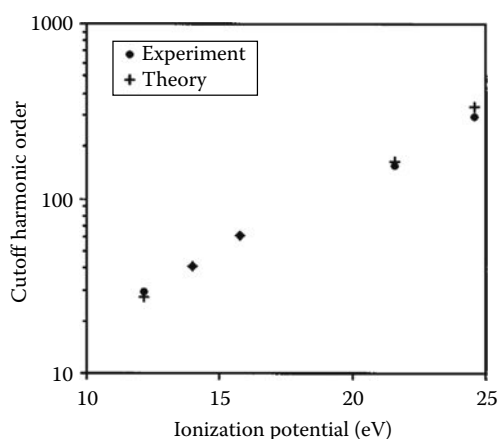


Figure 4.21 Comparison of predicted and observed cutoff photon energies for harmonic generation in the noble gases (on a log scale). (From Z. Chang, A. Rundquist, H. Wang, H. Kapteyn, and M. Murnane, *Phys. Rev. Lett.*, 79, 2967, 1997. Copyright 1997 by the American Physical Society.)

4.3.2.2 Pulse Width

Equation 4.112 reveals that the cutoff photon energy is inversely proportional to the square of the logarithm of the laser-pulse width, so it is clear that using shorter duration laser pulses should result in the generation of higher order harmonics. At the same laser intensity, a pulse with shorter duration causes less ground-state depletion. Thus atoms can experience higher laser intensity when the pulse is shorter, which leads to higher cutoff photon energy.

This is clearly shown in Figure 4.22, which plots the theoretical predictions for an argon atom, for pulse durations in the range between 10 and 100 fs. For even shorter pulse duration, the carrier-envelope phase also affects the cutoff photon energy, which is not taken into account when deriving Equation 4.112.

For example, when Ti:Sapphire laser system generating 26 fs pulses with a center wavelength of 800 nm is used, we expect to observe harmonics up to order 333 from He, which is well within the “water window” region between 4.4 and 2.3 nm, where water is less absorbing than carbon. X-rays in this region are important for imaging live biology samples. The experimentally generated high harmonic spectrum after a 0.4 μm carbon filter is shown in Figure 4.23. The signals below the 155th order are truncated for they are out the measurement range of the spectrometer. The signals above the 183rd order are blocked by the carbon filter. The 155th to 183rd order harmonics are seen as the small intensity modulation on a logarithmic scale. This water window x-ray generation experiment was done in 1997 using a 25 fs Ti:Sapphire CPA laser. Even shorter wavelength XUV light has been generated with 5 fs lasers from hollow-core fiber compressors.

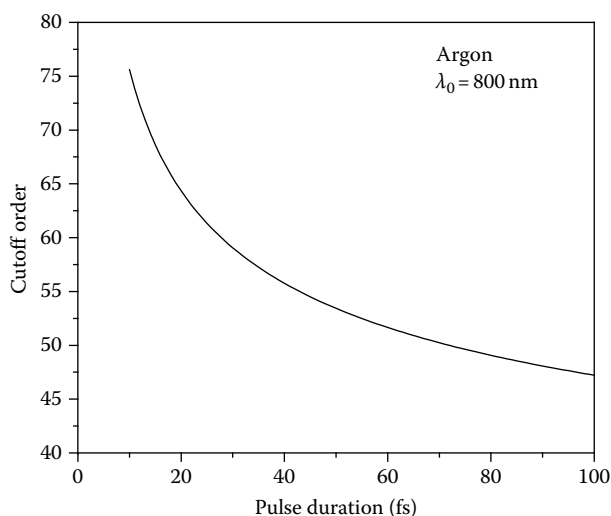


Figure 4.22 Dependence of cutoff order on pulse duration.

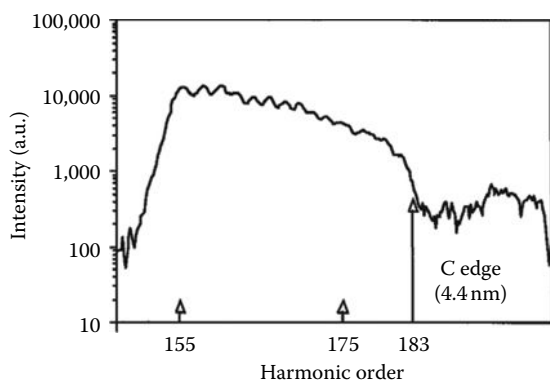


Figure 4.23 Harmonic emission from helium filtered through a $0.4\ \mu\text{m}$ carbon filter. (From Z. Chang, A. Rundquist, H. Wang, H. Kapteyn, and M. Murnane, *Phys. Rev. Lett.*, 79, 2967, 1997. Copyright 1997 by the American Physical Society.)

4.3.2.3 Wavelength of the Driving Laser

It is clear in Equation 4.112 that the cutoff photon energy for a given atomic state is proportional to the square of the wavelength. Figure 4.24 shows the calculation results of the relationship between the cutoff photon energy with the driving field wavelength for fixed pulse duration (25 fs). The figure shows that by changing the driving field wavelength from 0.4 to $4\ \mu\text{m}$, the cutoff of helium is extended from 0.2 to more than $10\ \text{keV}$. A Gaussian spectrum with $2\ \text{keV}$ FWHM bandwidth supports pulses shorter than $1\ \text{as}$! We can thus expect that Zeptosecond pulses be generated using mid-infrared laser in the future.

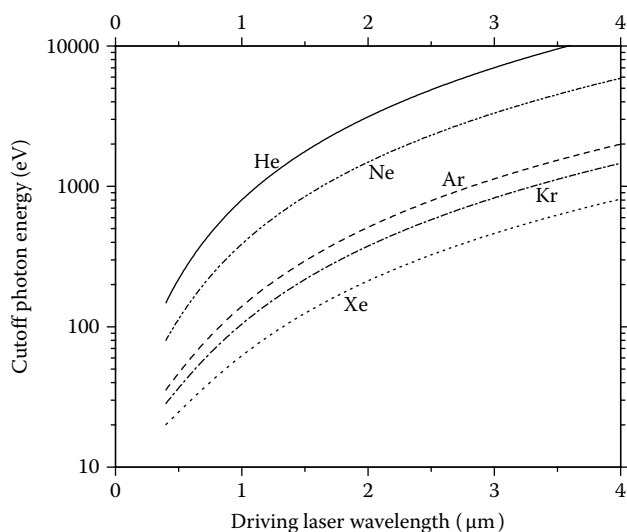


Figure 4.24 Calculated relationship between single-atom HHG cutoff photon energy and the driving wavelength.

The extension of the cutoff photon energy can be understood with the semiclassical three-step model. In the tunneling ionization regime, the ionization rate is independent of the laser wavelength. Therefore, the saturation intensity is the same for pulses with the same duration but different wavelengths. As a result, electrons experience the same field strength at saturation intensities for pulses with different wavelengths. From Newton's laws of mechanics, we know the kinetic energy of an electron acquired in a given electric field is proportional to the square of the travel time in the field. Therefore, the electrons can gain four times more energy when the optical period of the laser is doubled, or the laser wavelength is two times longer.

The first experiments that demonstrate the extension of the cutoff photon energy of the XUV spectrum by using long-wavelength lasers were demonstrated in 2001. The main difficulty of performing such an experiment is the lack of high-power femtosecond lasers with center wavelength above $1\text{ }\mu\text{m}$. The experiment was performed using an optical parametric amplifier (OPA) instead. It is pumped with a Ti:Sapphire CPA laser that delivered 25 fs, 1.2 mJ at sub-kilohertz repetition rate, as depicted in Figure 4.25. The OPA generates up to $100\text{ }\mu\text{J}$ infrared pulses that are tunable from 1.1 to $1.6\text{ }\mu\text{m}$. The OPA pulse duration is also 25 fs.

Because of the low pulse energy, the OPA beam is tightly focused onto the pulsed gas jet to achieve the required intensity. The focal spot size is $20\text{ }\mu\text{m}$ full width at half maximum. The gas density is $1 \times 10^{18}\text{ atoms/cm}^3$ in a $200\text{ }\mu\text{m}$ interaction region. The generated high harmonic signal is measured by a transmission grating based XUV spectrometer. The XUV beam is dispersed by a 2000 line/mm transmission grating on an MCP imaging detector. Another difficulty of the experiment is caused by the low conversion efficiency from the laser energy to the XUV flux. A low noise, 16 bit cooled charge-coupled device (CCD) camera is used to accumulate the spectrum image.

Figure 4.26 shows the results with Xenon gas at four wavelengths. Figure 4.26a is the spectrum produced by the $0.8\text{ }\mu\text{m}$ laser. Figures 4.26b through d are the results produced by the OPA pulses tuned at 1.51 , 1.37 , and $1.22\text{ }\mu\text{m}$, respectively. The results clearly illustrate the cutoff extension by using longer driving-field wavelength. It is worthy to point out the harmonic signal level drops as the wavelength is increased, which can be explained by the quantum theory of the three-step model.

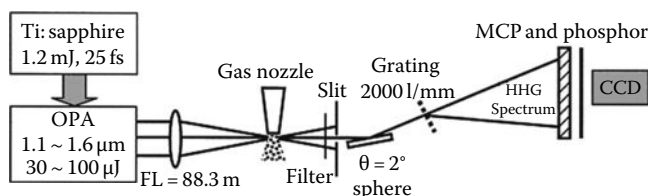


Figure 4.25 Experimental setup for generating high-order harmonics with long wavelength pump. (From B. Shan and Z. Chang, *Phys. Rev. A*, 65, 011804(R), 2001. Copyright 2001 by the American Physical Society.)

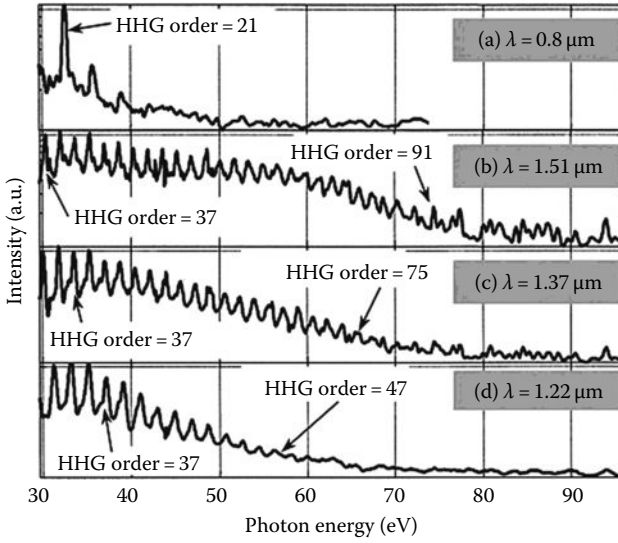


Figure 4.26 HHG by 50 μJ , 25 fs laser pulses of different wavelengths in xenon gas. The HHG wavelength is also tuned by the driving wavelength from OPA. (From B. Shan and Z. Chang, *Phys. Rev. A*, 65, 011804(R), 2001. Copyright 2001 by the American Physical Society.)

It is possible to generate 10 keV x-rays by using a 4 μm wavelength laser. The optical period corresponds to 4 μm is 13.3 fs, thus a 25 fs pulse at this wavelength contains only two cycles! While a 25 fs pulse at 0.8 μm can be delivered directly from a Ti:Sapphire CPA laser, generating 25 fs at 4 μm is yet to be demonstrated.

4.4 Free Electrons in Two-Color Laser Fields

Attosecond pulses can be generated in the combined laser fields at the fundamental and second harmonic frequencies, which is called two-color field. The generation processes can also be analysed using the three-step model. We first examine the cutoff photon energy of the high harmonic spectrum, which is determined by the kinetic energy of the electron gained in the two-color laser field. The method we use here is very similar to that applied to the one-color field.

4.4.1 Equation of Motion

The motion of free electrons in a two-color field can be understood by considering two linearly polarized monochromatic waves with the same polarization direction. The first one is the laser field at the fundamental frequency

$$\varepsilon_1(t) = E_1 \cos(\omega_0 t). \quad (4.113)$$

The second one is the second harmonic (SH) field

$$\varepsilon_2(t) = \alpha E_1 \cos(2\omega_0 t + \phi_{12}), \quad (4.114)$$

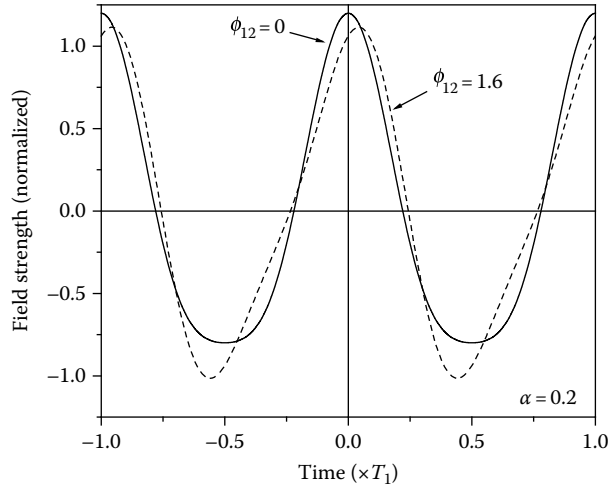


Figure 4.27 The combined two-color field.

where

α is the ratio of the amplitude of the SH field to that of the ω_0 field

ϕ_{12} is the relative phase between the two fields

The total field is

$$\varepsilon(t) = E_1[\cos(\omega_0 t) + \alpha \cos(2\omega_0 t + \phi_{12})]. \quad (4.115)$$

Figure 4.27 shows the total field for $\alpha = 0.20$, $\phi_{12} = 0$ and $\phi_{12} = 1.6\pi$. The intensity of the SH beam is 4%, i.e., α^2 , of the fundamental beam.

The equation of motion of a free electron is

$$\frac{d^2 x}{dt^2} = -\frac{e}{m} E_1[\cos(\omega_0 t) + \alpha \cos(2\omega_0 t + \phi_{12})]. \quad (4.116)$$

Assuming an electron is freed at time t' , and the initial velocity is zero, then the solution of the equation is

$$v(t) = -\frac{eE_1}{m\omega_0} \left[\sin(\omega_0 t) - \sin(\omega_0 t') + \frac{\alpha}{2} [\sin(2\omega_0 t + \phi_{12}) - \sin(2\omega_0 t' + \phi_{12})] \right], \quad (4.117)$$

$$x(t) = \frac{eE_1}{m\omega_0^2} \left\{ [\cos(\omega_0 t) - \cos(\omega_0 t')] + \frac{\alpha}{4} [\cos(2\omega_0 t + \phi_{12}) - \cos(2\omega_0 t' + \phi_{12})] + \omega_0 [\sin(\omega_0 t') + \frac{\alpha}{2} \sin(2\omega_0 t' + \phi_{12})](t - t') \right\}, \quad (4.118)$$

where v is the velocity of the electron. We define $x_1 = 2eE_1/m\omega_0^2$. Equation 4.118 can be normalized to

$$\begin{aligned} \frac{x(t)}{x_1} = \frac{1}{2} \Big\{ & [\cos(\omega_0 t) - \cos(\omega_0 t')] + \frac{\alpha}{4} [\cos(2\omega_0 t + \phi_{12}) \\ & - \cos(2\omega_0 t' + \phi_{12})] + \omega_0 [\sin(\omega_0 t') \\ & + \frac{\alpha}{2} \sin(2\omega_0 t' + \phi_{12})] (t - t') \Big\}. \end{aligned} \quad (4.119)$$

4.4.1.1 Return Time

Return time can be understood by employing a graphic method to examine

$$\begin{aligned} & \left[\cos(\omega_0 t) + \frac{\alpha}{4} \cos(2\omega_0 t + \phi_{12}) \right] - \left[\cos(\omega_0 t') + \frac{\alpha}{4} \cos(2\omega_0 t' + \phi_{12}) \right] \\ & = -\omega_0 \left[\sin(\omega_0 t') + \frac{\alpha}{2} \sin(2\omega_0 t' + \phi_{12}) \right] (t - t'). \end{aligned} \quad (4.120)$$

This is accomplished by plotting the function of the left side as an assistant curve and its tangent at $t = t'$. The solution is the point where the curve and the line cross, as shown in Figure 4.28. Since $\alpha/4$ is a small quantity, the function is similar to the fundamental field. Thus, the return time for a two-color system is not very different from the one-color values. However, unlike the one-color case, the function on the left side is not the same function as the two-color laser field. When the relative phase changes, the laser field changes significantly, but the assistant curve does not change much because of the $\alpha/4$ factor.

By solving the equation $x(t) = 0$ numerically, we find the return time for $\phi_{12} = 0$ and $\phi_{12} = \pi$, as shown in Figure 4.29. These results are slightly different from the one-color case and it is clear the return time depends on the relative phase ϕ_{12} and the relative amplitude α .

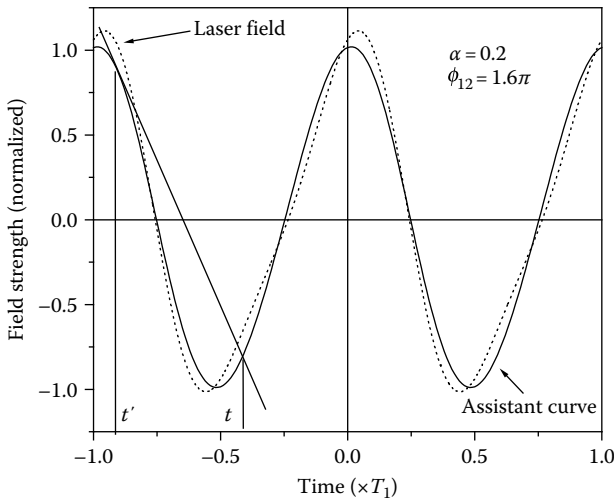


Figure 4.28 Finding the return time in a two-color laser field graphically, $\phi_{12} = 1.6\pi$.

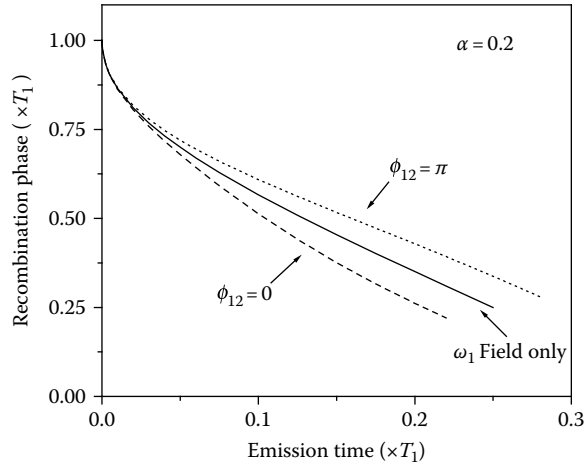


Figure 4.29 Return time for two relative phases, $\phi_{12} = 0$ and $\phi_{12} = \pi$.

4.4.2 Return Energy

The returning velocity can be written as

$$v(t) = v_1(t) + v_2(t), \quad (4.121)$$

where $v_1(t)$ and $v_2(t)$ are the velocities when only the ω_0 or the $2\omega_0$ is present. The velocities can be expressed as

$$v_1(t) = -\frac{eE_1}{m\omega_0} [\sin(\omega_0 t) - \sin(\omega_0 t')], \quad (4.122)$$

$$v_2(t) = -\frac{e\alpha E_1}{m2\omega_0} [\sin(2\omega_0 t + \phi_{12}) - \sin(2\omega_0 t' + \phi_{12})]. \quad (4.123)$$

Consequently, the returning energy can be expressed as

$$K(t) = \frac{1}{2} m [v_1(t) + v_2(t)]^2 = K_1(t) + K_2(t) + 2\sqrt{K_1(t)K_2(t)}, \quad (4.124)$$

where $K_1(t)$ and $K_2(t)$ are the kinetic energy acquired by the electron in the ω_0 or the $2\omega_0$ fields, respectively. $K_2(t)$ depends on the relative phase ϕ_{12} . These two energies normalized to the ponderomotive energy of the ω_0 field U_{p1} are

$$\frac{K_1(t)}{U_{p1}} = 2[\sin(\omega_0 t) - \sin(\omega_0 t')]^2, \quad (4.125)$$

$$\frac{K_2(t)}{\alpha^2 U_{p1}} = [\sin(2\omega_0 t + \phi_{12}) - \sin(2\omega_0 t' + \phi_{12})]^2. \quad (4.126)$$

Notice that Equation 4.125 has the same form as the one-color case. However, for the same emission time, the return time in the two-color field is different from that in the one-color field (Figure 4.30). As a result, the maximum kinetic energy gain from the fundamental field, $K_{1,max}$, is different from the $3.17U_p$, as shown in Figure 4.31. For a numerical reference, $K_{1,max} = 3.47U_p$ for $\phi_{12} = 1.6\pi$, whereas $K_{1,max} = 2.81U_p$ for $\phi_{12} = 0.6\pi$.

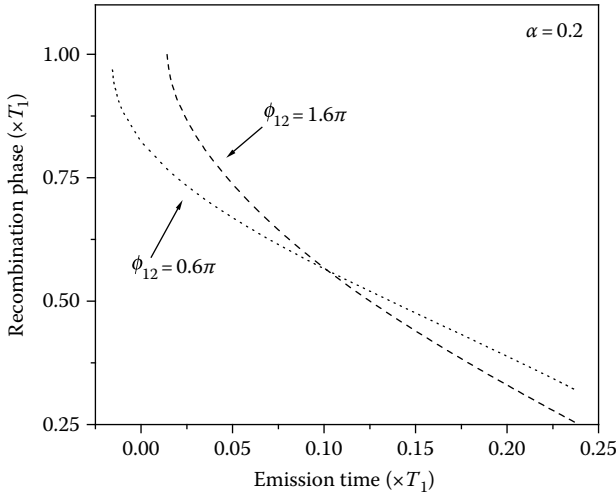


Figure 4.30 Return time for two relative phases, $\phi_{12} = 0.6\pi$ and $\phi_{12} = 1.6\pi$.

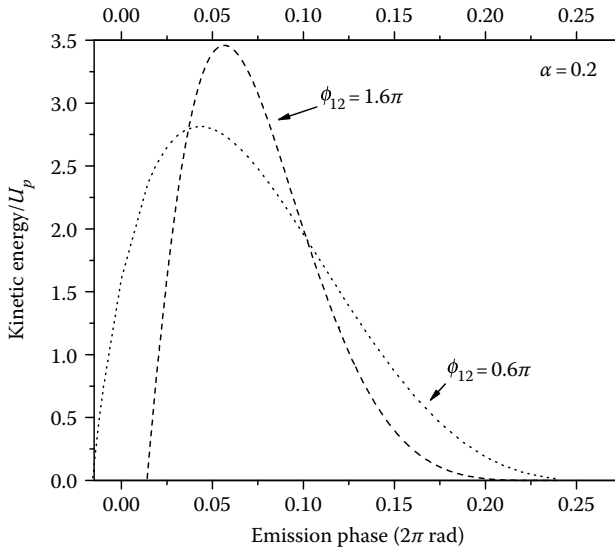


Figure 4.31 Kinetic-energy component $K_1(t)$.

$K_2(t)/\alpha^2 U_{p1}$ is plotted in [Figure 4.32](#) for $\phi_{12} = 0.6\pi$ and $\phi_{12} = 1.6\pi$. It is interesting to see the $K_2(t) \approx 0$ in the cutoff region. The cutoff photon energy is the maximum value of K plus the ionization potential of the atom. We can conclude that adding the SH field at these two phases will not extend the cutoff much.

4.4.3 Two-Color Gating

The ionization rate depends on α and ϕ_{12} . For $\alpha = 0.20$, $\phi_{12} = 0$ and for $F_0/E_1 = 100$, $n^* = 1$, $m = 0$, the rate is shown in [Figure 4.33](#). It is seen that

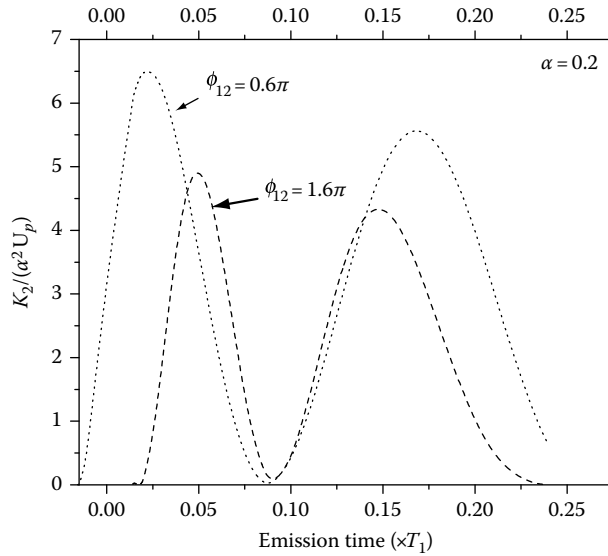


Figure 4.32 Kinetic-energy component $K_2(t)$.

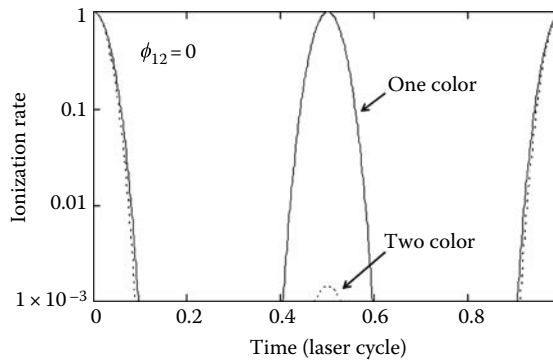


Figure 4.33 Comparison of the ionization rate between the one-color and the two-color lasers, $\phi_{12} = 0$.

the ionization at the half laser cycle is suppressed. Consequently, there will be no attosecond emissions due to the electron emitted there. This increase in the spacing between adjacent attosecond pulses to one laser cycle, which is the foundation of the two-color gating, is discussed in Chapters 7 and 8.

As a comparison, when the relative phase $\phi_{12} = 1.6\pi$, the difference between the ionization rate between two maximas of the field is much less than that of the $\phi_{12} = 0$, as shown in [Figure 4.34](#).

4.5 Polarization Gating

The semiclassical model predicts that when atoms are driven by a linearly polarized femtosecond laser pulse containing multiple optical

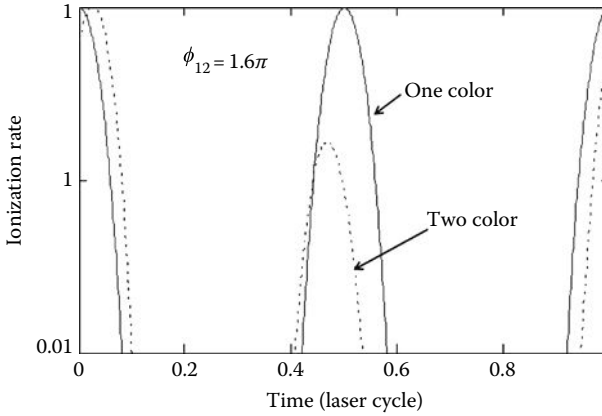


Figure 4.34 Comparison of the ionization rate between the one-color and the two-color lasers, $\phi_{12} = 1.6\pi$.

cycles, a train of attosecond pulses can be generated if the laser intensity is sufficiently high. The three-step process occurs twice a laser cycle, and thus the separation between adjacent pulses is half an optical cycle, which is 1.3 fs for Ti:Sapphire lasers. Attosecond pulse trains are useful light sources for some applications. For cases where the physical processes to be studied last longer than the pulse spacing, it could be hard to interpret the data.

In general, singly isolated attosecond pulses are desirable. When a single attosecond pulse starts the process (the pump) and another attosecond pulse probes it, the time evolution of the system can be mapped out. A scheme for generating single isolated attosecond pulses was first proposed by Corkum in 1994, which is named the polarization gating. It relies on the fact that the attosecond pulse generation efficiency is susceptible to the ellipticity of the driving laser field. It took 12 years for scientists to finally measure the duration of the isolated XUV pulses generated with the polarization gating. In Corkum's the original proposal, two laser pulses with different center frequency are needed. Here, we discuss the principle of polarization gating that requires one center frequency, which is easier to implement experimentally.

4.5.1 Electrons in Elliptically Polarized Laser Fields

4.5.1.1 Laser Field

Consider an electric field with ellipticity ξ , which is defined as the ratio between the amplitude of the minor axis to the major axis of the ellipse that is the trace drawn by the end of the electric vector in a two-dimensional plane as the time increases, as depicted in Figure 4.35. For reference, we define the major axis along the x direction and the minor axis along the y direction. The field can be expressed as

$$\vec{E}(t) = E_{x0} [\hat{i} \cos(\omega_0 t) + \hat{j} \xi \sin(\omega_0 t)], \quad (4.127)$$

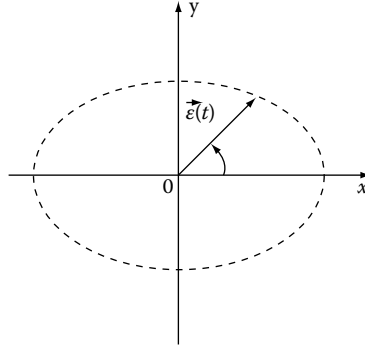


Figure 4.35 Elliptically polarized laser field.

where

E_{x0} is the amplitude of the major axis

$\hat{i}$ and $\hat{j}$ are unit vectors in the x and y directions, respectively

The amplitude of the minor axis is $E_{y0} = \xi E_{x0}$.

4.5.1.2 Equations of Motion

The motion in the x and y directions can be calculated separately as

$$m_e \frac{d^2x}{dt^2} = -eE_{x0} \cos(\omega_0 t) \quad (4.128)$$

and

$$m_e \frac{d^2y}{dt^2} = -e\xi E_{x0} \sin(\omega_0 t). \quad (4.129)$$

The solution of Equation 4.128 is the same as that of the linearly polarized case while the solution of Equation 4.129 is

$$\frac{dy}{dt} = \xi \frac{eE_{x0}}{m\omega_0} [\cos(\omega_0 t) - \cos(\omega_0 t')], \quad (4.130)$$

$$y = \xi \frac{eE_{x0}}{m\omega_0^2} [\sin(\omega_0 t) - \sin(\omega_0 t') - \cos(\omega_0 t')\omega_0(t - t')]. \quad (4.131)$$

Introducing a normalization factor

$$x_{\max} = \frac{2eE_{x0}}{m\omega_0^2}, \quad (4.132)$$

which is the maximum displacement in the x direction in one laser cycle, Equation 4.132 becomes

$$\frac{y}{\xi x_{\max}} = \frac{1}{2} [\sin(\omega_0 t) - \sin(\omega_0 t') - \cos(\omega_0 t')\omega_0(t - t')]. \quad (4.133)$$

An example for $t' = 0$ is shown in [Figure 4.36](#).

The electron released in the range $t' = 0$ to $T_0/4$ will return to $x = 0$, but never returns to $y = 0$. The trajectory in the x - y plane is shown in [Figure 4.37](#).

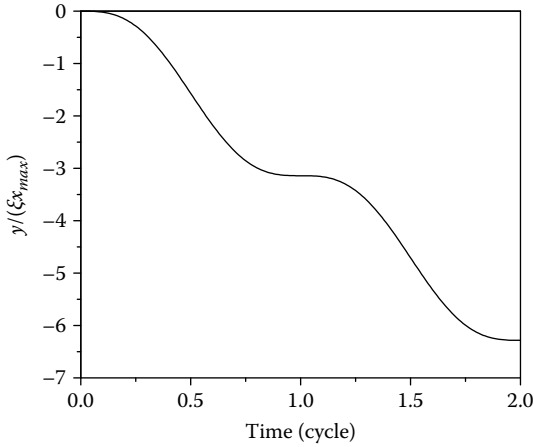


Figure 4.36 The transverse trajectory.

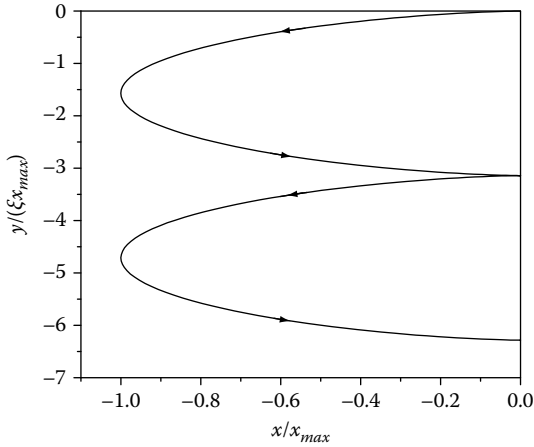


FIGURE 4.37 The transverse trajectory in the x - y plane.

4.5.1.3 Transverse Displacement

For a given electron's birth time, the value of y at the return time in the x direction can be calculated by using Equation 4.133. The numerical solution is shown in Figure 4.38.

The maximum displacement is

$$y_{\max} = \pi \xi x_{\max}, \quad (4.134)$$

which occurs at $t' = 0$ or $t = T$.

The radii of noble gas atoms are listed in Table 4.3. They are on the order of 0.1–0.2 nm. The typical Ti:Sapphire laser intensity for attosecond pulse generation is 5×10^{14} W/cm², $x_{\max} = 2$ nm. For $\xi = 0.1$, $y_{\max} \approx 0.6$ nm, which is comparable to the atomic radius. For even larger ellipticity, the returning electrons released from an atom will miss the parent ion.

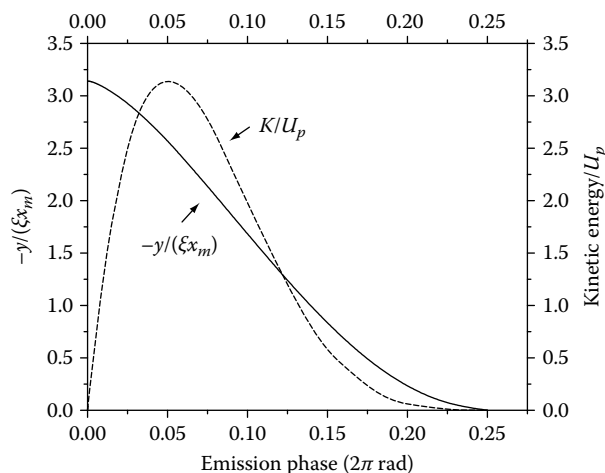


Figure 4.38 The transverse displacement.

TABLE 4.3 Radius of Noble Gas Atoms

	He	Ne	Ar	Kr	Xe
r (nm)	0.130	0.160	0.192	0.198	0.218

4.5.1.4 Quantum Diffusion

Assume the radius of the electron wave packet immediately after tunneling out from an atom is $r_{\perp}$ and the amplitude of the electron wave function is assumed to be $e^{-r/r_{\perp}}$. The transverse velocity can be estimated by the uncertainty principle

$$v_{\perp} = \frac{h}{m_e r_{\perp}}. \quad (4.135)$$

For example, for neon atoms, $r_{\perp} = \sqrt{2} \times 0.16$ nm and $v_{\perp} = 3.2$ nm/fs. The width of the wave function increases with time, i.e., equals $v_{\perp}t + r_{\perp}$. In a linearly polarized field, the amplitude distribution of the ground state and the returning wave packet after one laser cycle (2.67 fs) is shown in Figure 4.39. In a circularly polarized field ($\xi = 1$), the amplitude is shown in Figure 4.40.

The quantum diffusion leads to the reduction of laser attosecond pulse conversion efficiency. However, it allows the recombination to occur even when ellipticity is larger than 0.1. When the ellipticity increases, the amplitude of the part of the returning electron that meets the parent ion decreases. That is the origin of the dependence of high harmonic yield on the ellipticity, which is studied in detail in Chapter 5.

4.5.2 Isolated Attosecond Pulse Generation

4.5.2.1 Principle of the Polarization Gating

The polarization gating is based on the strong dependence of the attosecond pulse generation efficiency on the ellipticity of the near infrared

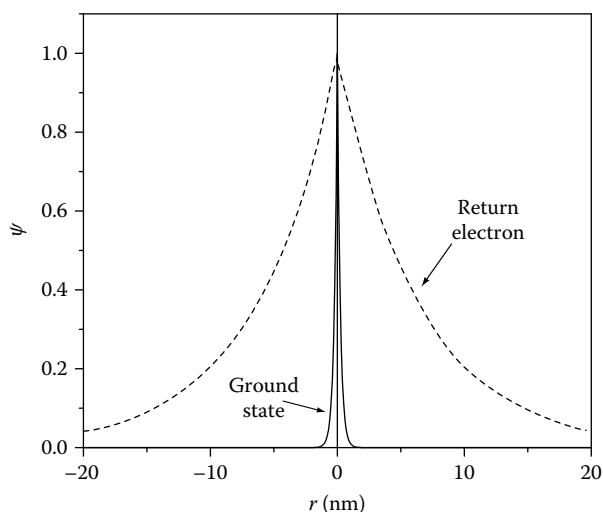


Figure 4.39 Quantum diffusion in linearly polarized field.

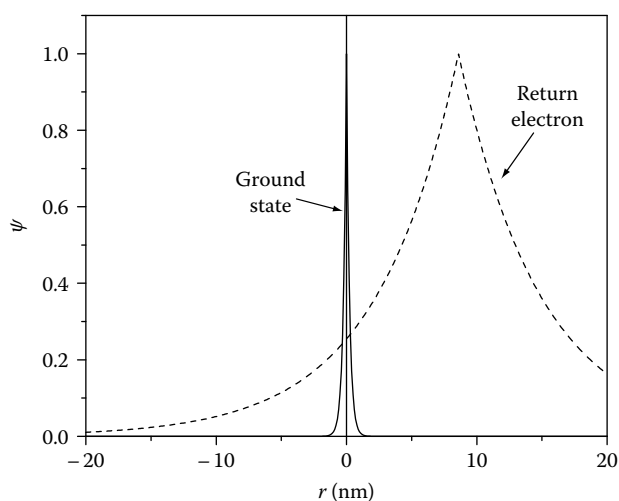


Figure 4.40 Quantum diffusion in circularly polarized field.

laser, ξ . To create a temporal gate, the polarization state of a laser pulse changes from circular to linear and back to circular again. As a result, the electron freed by the laser field will be driven away from the parent ion at both the head and tail parts of the laser pulse by the transverse component of the field. This will eliminate the possibility of recombination of the electron with the parent ion. Thus, attosecond pulses can only be produced by the center portion of the laser pulse that is nearly linearly polarized. This linearly polarized portion is where the gate opens and it could be much shorter than the input pulse duration. The gate width should be narrower than the spacing between adjacent attosecond pulses, 1.3 fs, for Ti:Sapphire lasers to generate singly isolated attosecond pulses.

We can draw an analogy between the polarization gating for generating single isolated attosecond pulses and the Pockels cell pulse picker in femtosecond lasers. In both cases, a single pulse is extracted from a pulse train. Interestingly, the attosecond polarization gating occurs during the generation process. In other words, all the pulses in the train except one are aborted before they are born.

4.5.2.2 Laser Field

The laser field with a time-dependent ellipticity for polarization gating can be generated by the superposition of a left and a right-circularly polarized Gaussian pulse, as illustrated in Figure 4.41, which was suggested by Platonenko and Strelkov in 1999. The advantage of this scheme is that laser pulses with only one center frequency are required, which is easy to implement experimentally.

For a circularly polarized pulse, $E_{x0} = E_{y0} = E_0$, where E_{x0} , E_{y0} , and E_0 are the peak field amplitude components and the total amplitude. We consider the case where E_0 , carrier frequency ω_0 , pulse duration τ_p , and carrier-envelope phase φ_{CE} are the same for the two counter-rotating pulses. The delay between them, T_d , is assumed to be an integral number, n , of optical periods for simplifying the analysis. In principle, T_d can take any value. The FWHM of the spectrum of each circularly polarized is $\Delta\omega = 4\ln 2/\tau_p$.

The electric fields of the left and right circularly polarized pulses propagating in the z direction are

$$\vec{E}_l(t) = E_0 e^{-2\ln(2)\left(\frac{t-T_d/2}{\tau_p}\right)^2} \times [\hat{i} \cos(\omega_0 t + \varphi_{CE}) + \hat{j} \sin(\omega_0 t + \varphi_{CE})](-1)^n, \quad (4.136)$$

and

$$\vec{E}_r(t) = E_0 e^{-2\ln(2)\left(\frac{t+T_d/2}{\tau_p}\right)^2} \times [\hat{i} \cos(\omega_0 t + \varphi_{CE}) - \hat{j} \sin(\omega_0 t + \varphi_{CE})](-1)^n. \quad (4.137)$$

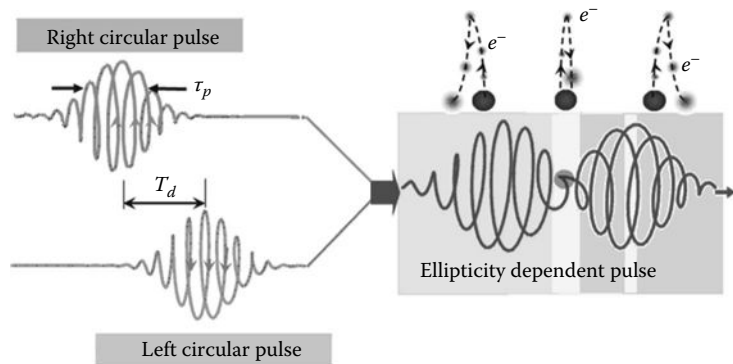


Figure 4.41 Creation of laser pulse for polarization gating. (Reprinted from Shan, B. et al., *J. Mod. Opt.*, 52, 277, 2005.)

The total field can be resolved into two orthogonally polarized components. They are named as driving and gating field, respectively, because of the functions they serve. The driving field polarized in x direction generates the attosecond pulse, whereas the gating field in the y direction suppresses the attosecond emission outside of the polarization gate.

The driving field can be expressed as

$$\varepsilon_{drive}(t) = E_0 \left[e^{-2 \ln 2 \frac{(t+T_d/2)^2}{\tau_p^2}} + e^{-2 \ln 2 \frac{(t-T_d/2)^2}{\tau_p^2}} \right] \cos(\omega_0 t + \varphi_{CE}). \quad (4.138)$$

and the gating field is

$$\varepsilon_{gate}(t) = E_0 \left[e^{-2 \ln 2 \frac{(t+T_d/2)^2}{\tau_p^2}} - e^{-2 \ln 2 \frac{(t-T_d/2)^2}{\tau_p^2}} \right] \sin(\omega_0 t + \varphi_{CE}), \quad (4.139)$$

In the frequency domain, the driving and gating pulses are

$$\begin{aligned} \tilde{E}_{drive}(\omega) &= E_0 e^{-\left(\frac{\omega-\omega_0}{\Delta\omega}\right)^2} \frac{1}{2} \left(e^{-i(\omega-\omega_0)\frac{T_d}{2}} + e^{i(\omega-\omega_0)\frac{T_d}{2}} \right) e^{i(\omega-\omega_0)\frac{T_d}{2}} \\ &= E_0 e^{-\left(\frac{\omega-\omega_0}{\Delta\omega}\right)^2} \cos\left((\omega-\omega_0)\frac{T_d}{2}\right) e^{i(\omega-\omega_0)\frac{T_d}{2}}, \end{aligned} \quad (4.140)$$

and

$$\begin{aligned} \tilde{E}_{gate}(\omega) &= E_0 e^{-\left(\frac{\omega-\omega_0}{\Delta\omega}\right)^2} \frac{1}{2} \left(e^{-i(\omega-\omega_0)\frac{T_d}{2}} - e^{i(\omega-\omega_0)\frac{T_d}{2}} \right) e^{i(\omega-\omega_0)\frac{T_d}{2}} e^{i\frac{\pi}{2}} \\ &= E_0 e^{-\left(\frac{\omega-\omega_0}{\Delta\omega}\right)^2} \sin\left((\omega-\omega_0)\frac{T_d}{2}\right) e^{i\omega\frac{T_d}{2}}. \end{aligned} \quad (4.141)$$

The power spectra of these two fields are $I_{drive}(\omega) = I_0 e^{-2(\omega-\omega_0/\Delta\omega)^2} \cos^2\left[(\omega-\omega_0)\frac{T_d}{2}\right]$ and $I_{gate}(\omega) = I_0 e^{-2(\omega-\omega_0/\Delta\omega)^2} \sin^2\left[(\omega-\omega_0)\frac{T_d}{2}\right]$, respectively. The peak intensity I_0 and the peak field amplitude are related by $I_0 = \epsilon_0 c E_0^2$, which is different from that for the linearly different pulse where $I_0 = 1/2 \epsilon_0 c E_0^2$. The spectra can be measured with a spectrometer. A polarizer can be used to select one of them before sending to the spectrometer. There is a dip at $\omega = \omega_0$ for the gating field spectrum. The delay T_d can be determined from the spectrum modulation.

4.5.2.3 Fields inside the Polarization Gate

A parameter γ can be introduced to describe the delay between the two circular pulses such that

$$T_d = \gamma \tau_p. \quad (4.142)$$

The driving and gating fields can be rewritten as

$$\varepsilon_{drive}(t) = E_0 A_{drive}(t) \cos(\omega_0 t + \varphi_{CE}). \quad (4.143)$$

and

$$\varepsilon_{gate}(t) = E_0 A_{gate} \sin(\omega_0 t + \varphi_{CE}). \quad (4.144)$$

While the envelope of the driving and gating fields are

$$A_{drive}(t) = e^{-2 \ln 2 \left(\frac{t}{\tau_p} + \frac{\gamma}{2} \right)^2} + e^{-2 \ln 2 \left(\frac{t}{\tau_p} - \frac{\gamma}{2} \right)^2}, \quad (4.145)$$

and

$$A_{gate}(t) = e^{-2 \ln 2 \left(\frac{t}{\tau_p} + \frac{\gamma}{2} \right)^2} - e^{-2 \ln 2 \left(\frac{t}{\tau_p} - \frac{\gamma}{2} \right)^2}. \quad (4.146)$$

For mathematical simplicity, we consider the case that the driving field is a constant at the center of the polarization gate. This requires

$$\frac{d^2}{dt^2} A_{drive}(t) \Big|_{t=0} = 0. \quad (4.147)$$

Since

$$\begin{aligned} -\frac{\tau_p}{4 \ln 2} \frac{d}{dt} A_{drive}(t) &= e^{-2 \ln 2 \left(\frac{t}{\tau_p} + \frac{\gamma}{2} \right)^2} \left(\frac{t}{\tau_p} + \frac{\gamma}{2} \right) \\ &\quad + e^{-2 \ln 2 \left(\frac{t}{\tau_p} - \frac{\gamma}{2} \right)^2} \left(\frac{t}{\tau_p} - \frac{\gamma}{2} \right), \end{aligned} \quad (4.148)$$

we can assume

$$\begin{aligned} -\frac{\tau_p^2}{4 \ln 2} \frac{d^2}{dt^2} A_{drive}(t) &= e^{-2 \ln 2 \left(\frac{t}{\tau_p} + \frac{\gamma}{2} \right)^2} \left[1 - 4 \ln 2 \left(\frac{t}{\tau_p} + \frac{\gamma}{2} \right)^2 \right] \\ &\quad + e^{-2 \ln 2 \left(\frac{t}{\tau_p} - \frac{\gamma}{2} \right)^2} \left[1 - 4 \ln 2 \left(\frac{t}{\tau_p} - \frac{\gamma}{2} \right)^2 \right], \end{aligned} \quad (4.149)$$

which leads to

$$\gamma = \frac{1}{\sqrt{\ln 2}} \approx 1.2. \quad (4.150)$$

This corresponds to the case where the delay is not much longer than the pulse duration.

In the range $-1/2\tau_p < t < 1/2\tau_p$, the envelope function of the driving field can be expressed approximately by

$$A_{drive}(t) = \frac{2}{\sqrt{e}} \approx 1.213, \quad (4.151)$$

In the same time range, $A_{gate}(t)$ is close to a linear function. The slope is

$$\begin{aligned} -\frac{\tau_p}{4 \ln 2} \frac{d}{dt} A_{gate}(t) &= e^{-2 \ln 2 \left(\frac{t}{\tau_p} + \frac{\gamma}{2} \right)^2} \left(\frac{t}{\tau_p} + \frac{\gamma}{2} \right) \\ &\quad - e^{-2 \ln 2 \left(\frac{t}{\tau_p} - \frac{\gamma}{2} \right)^2} \left(\frac{t}{\tau_p} - \frac{\gamma}{2} \right). \end{aligned} \quad (4.152)$$

At $t=0$ this becomes

$$\frac{d}{dt} A_{gate}(t) \Big|_{t=0} = 4 \sqrt{\frac{\ln 2}{e}} \frac{1}{\tau_p}. \quad (4.153)$$

Thus,

$$A_{gate}(t) = 4\sqrt{\frac{\ln 2}{e}} \frac{t}{\tau_p} \approx 2 \frac{t}{\tau_p}. \quad (4.154)$$

The two field components are

$$E_{drive}(t) = \frac{2}{\sqrt{e}} E_0 \cos(\omega_0 t + \varphi_{CE}), \quad (4.155)$$

and

$$E_{gate}(t) = 4\sqrt{\frac{\ln 2}{e}} E_0 \frac{t}{\tau_p} \sin(\omega_0 t + \varphi_{CE}). \quad (4.156)$$

4.5.2.4 Electron Trajectories

We consider the case that $\tau_p = 2T_0$. The time range $-1/2\tau_p < t < +1/2\tau_p$ can also be expressed in terms of optical cycles, i.e., $-T_0 < t < +T_0$. The trajectories that correspond to the maximum returning kinetic energy are analyzed.

For $\varphi_{CE} = 0$, there are two short trajectories that electrons are released and returned within the time range. We consider the trajectory in the x and y directions separately. The two field components are

$$\mathcal{E}_{drive}(t) = E_{d0} \cos(\omega_0 t) \quad (4.157)$$

and

$$\mathcal{E}_{gate}(t) = E_{g0} \frac{t}{\tau_p} \sin(\omega_0 t), \quad (4.158)$$

where $E_{d0} = \left(\frac{2}{\sqrt{e}}\right) E_0$, $E_{g0} = 4\sqrt{\ln 2/e} E_0$.

In the x direction, the motion is the same as that in a linearly polarized monochromatic field.

$$\frac{x(t)}{x_d} = \frac{1}{2} [\cos(\omega_0 t) - \cos(\omega_0 t')] + \sin(\omega_0 t') \omega_0 (t - t'). \quad (4.159)$$

Here, $x_d = 2eE_{d0}/m\omega_0^2$, t' is the electron's birth time. The returning energy is the maximum for the electron released at $\omega_0 t' = 0.05 \times 2\pi$ rad and returns at $\omega_0 t = 0.7 \times 2\pi$.

In the y direction, the equation of motion is

$$m \frac{d^2 y}{dt^2} = -eE_{g0} \frac{t}{\tau_p} \sin(\omega_0 t). \quad (4.160)$$

And the velocity is

$$\begin{aligned} & \frac{m\omega_0^2}{eE_{g0}} \frac{dy}{dt} \\ &= \frac{1}{\tau_p} [\omega_0 t \cdot \cos(\omega_0 t) - \sin(\omega_0 t) + \sin(\omega_0 t') - \omega_0 t' \cdot \cos(\omega_0 t')]. \end{aligned} \quad (4.161)$$

The displacement then becomes

$$\frac{m\omega_0^3}{eE_{g0}}\tau_p y = 2[\cos(\omega_0 t) - \cos(\omega_0 t')] + \omega_0 t \cdot \sin(\omega_0 t) - \omega_0 t' \cdot \sin(\omega_0 t') + [\sin(\omega_0 t') - \omega_0 t' \cdot \cos(\omega_0 t')]\omega_0(t - t'). \quad (4.162)$$

For $\omega_0 t' = 0.05 \times 2\pi$ rad and $\omega_0 t = 0.7 \times 2\pi$, the right hand side of equation numerically becomes -6.758 . Thus the transverse displacement is

$$y_c = -\frac{6.758}{2\pi} \frac{T_0}{\tau_p} \frac{eE_{g0}}{m\omega_0^2} \approx -1.08 \frac{T_0}{\tau_p} \frac{eE_{g0}}{m\omega_0^2}. \quad (4.163)$$

The typical Ti:Sapphire laser intensity for attosecond pulse generation is 5×10^{14} W/cm², $y_0 \approx 1$ nm.

4.5.2.5 Polarization Gate Width

The electric field of the NIR laser pulse for polarization gating can be expressed as

$$\vec{\mathcal{E}}(t) = \mathcal{E}_{drive}(t)\hat{i} + \mathcal{E}_{gate}(t)\hat{j}. \quad (4.164)$$

The time-dependent ellipticity is

$$\xi(t) = \frac{A_{gate}(t)}{A_{drive}(t)} = \frac{\left| e^{-2 \ln(2) \left(\frac{t-T_d/2}{\tau_p} \right)^2} - e^{-2 \ln(2) \left(\frac{t+T_d/2}{\tau_p} \right)^2} \right|}{\left| e^{-2 \ln(2) \left(\frac{t-T_d/2}{\tau_p} \right)^2} + e^{-2 \ln(2) \left(\frac{t+T_d/2}{\tau_p} \right)^2} \right|}$$

$$= \frac{\left| 1 - e^{-4 \ln(2) \frac{T_d}{\tau_p} t} \right|}{1 + e^{-4 \ln(2) \frac{T_d}{\tau_p} t}}, \quad (4.165)$$

Figure 4.42 shows the variation of the ellipticity inside the laser pulse using Equation 4.165 when $\tau_p = 8$ fs long and $T_d = 15$ fs.

For harmonic orders higher than the 21st, the harmonic signal drops by more than an order of magnitude when the ellipticity increases from 0 to 0.2. Therefore, the attosecond pulse is generated in the temporal range around $t=0$ and $\xi \leq 0.2$, where the ellipticity decreases almost linearly with $|t|$ in the center portion,

$$\xi(t) \approx \left| 2 \ln(2) \frac{T_d}{\tau_p^2} t \right|. \quad (4.166)$$

The time interval, where the ellipticity is less than a certain threshold value ξ_{th} , is thus

$$\delta t_G = \frac{\xi_{th}}{\ln(2)} \frac{\tau_p^2}{T_d}. \quad (4.167)$$

It is named the polarization gate width. For harmonic order higher than 20, we can set $\xi_{th} = 0.2$. It suggests that there are two ways to reduce the

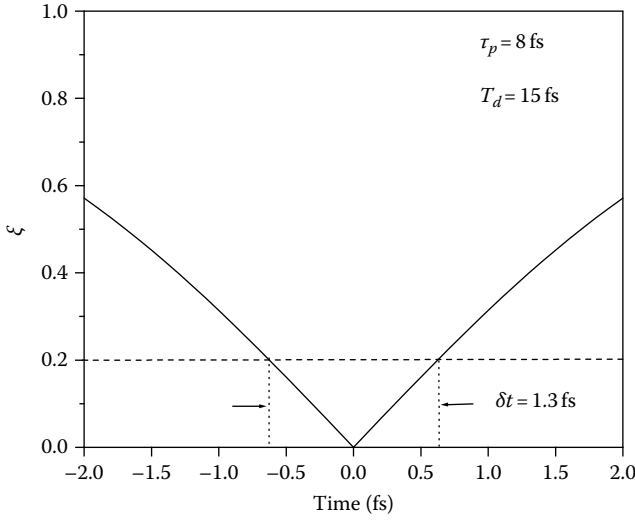


Figure 4.42 Time-dependent ellipticity and polarization gate width.

polarization gate width. The first one is to use shorter pulses; the second one is to increase the delay between the pulses. Reducing pulse width is more effective because of the quadratic dependence.

The second approach is at the cost of losing laser field strength for a given laser-pulse energy. The amplitude of the linear field for a given delay and pulse duration can be calculated from Equation 4.164 at $t = 0$,

$$E(0) = 2E_0 e^{-\frac{\ln(2)}{2} \left(\frac{T_d}{\tau_p}\right)^2}. \quad (4.168)$$

The field is significantly lower than the peak field of each pulse, E_0 , for $T_d \gg \tau_p$, which means that the field outside the gate is much stronger than the inside one. In such case, the conversion efficiency is low because most of the laser energy is outside the gate.

In experiments, one should choose $T_d \approx \tau_p$ and $\delta t_G = T_0/2$. In this case, $\tau_p = \delta t_G/0.3 = T_0/0.6$. For Ti:Sapphire laser, $\tau_p = 2.67 \text{ fs}/0.6 = 4.45 \text{ fs}$. The field amplitude inside the gate, $E(0) = \sqrt{2}E_0$, which is higher than the outside. The gate width

$$\delta t_G = 0.3\tau_p. \quad (4.169)$$

Equation 4.169 indicates that applying polarization gating to high harmonic generation is equivalent to the reduction of the duration of a linearly polarized pulse by a factor of three. When $\delta t_G = T_0/2$, it is expected that only a single attosecond pulse is produced in the plateau region of the XUV spectrum. The calculated required delay time T_d for producing a single isolated pulse as a function of the laser-pulse duration is shown in Figure 4.43.

4.5.2.6 Optics for Creating Laser Pulse for Polarization Gating

Laser field with the required time-dependent ellipticity can be constructed by a simple method illustrated in Figure 4.44. The first quartz plate is a

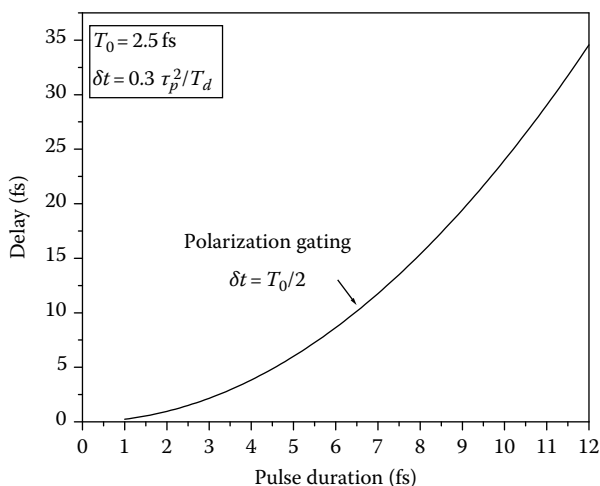


Figure 4.43 The required delay between two counter-rotating circularly polarized pulses for extracting a single isolated attosecond pulse from a pulse train. The gate width equals to one half of an optical cycle.

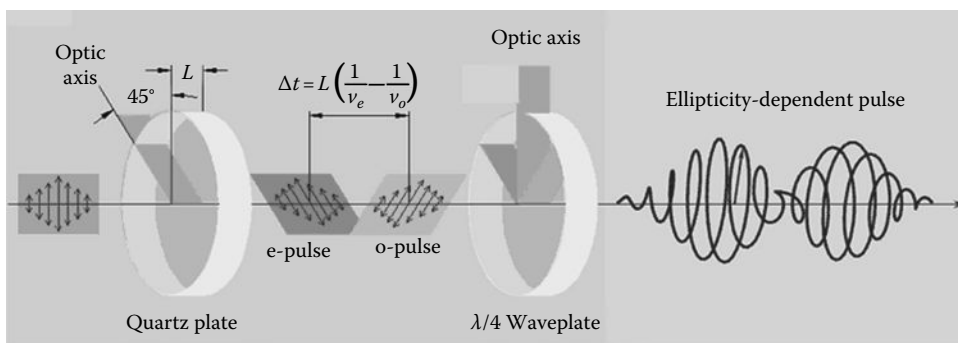


Figure 4.44 Time-dependent ellipticity and polarization gate width. (Reprinted from Shan, B. et al., *J. Mod. Opt.*, 52, 277, 2005.)

multiple order whole-wave plate where the linearly polarized input pulse is evenly divided into an o-ray and an e-ray by setting its optic axis 45° with respect to the input polarization. The o-pulse and the e-pulse are separated in time because the o-pulse and e-pulse travelled at different group velocities, v_o and v_e . The delay is proportional to the plate thickness, L . The durations of the two pulses are almost the same because the difference of the group velocity dispersions is small for the two polarization orientations. An achromatic quarter-wave plate is placed with its optic axis along the initial input polarization (45° to o-pulse and e-pulse). It can convert broadband o-pulse and e-pulse into left- and right-circularly polarized light. The superposition of these two pulses produced a pulse whose ellipticity changes rapidly with time. The dispersion introduced by these two components can be compensated by chirped mirrors.

4.5.2.7 Upper Limit of Laser-Pulse Duration

Although the gate width of polarization gating can be set to half a laser cycle with very long laser pulse as long as the delay between the two circularly polarized pulses is large enough, it is just one of the necessary conditions for generating isolated attosecond pulses. Another necessary condition is that the ground-state population of the atom responsible for the attosecond light emission inside the polarization gate cannot be zero. Otherwise, there will be no atom emits XUV light. In other words, the ionization of the target atom by the laser field before the polarization gate should not completely deplete the ground-state population. The ionization probability of argon atom as a function of the laser-pulse duration is shown in Figure 4.45. The calculations are done with the ADK rate and the gate width is kept as half of a laser cycle. The pulse duration at which the ground state is almost completely depleted is ~ 7 fs, which is upper limit for argon atom.

Figure 4.46 shows the ionization probabilities of the Helium atom within the NIR laser pulse for gating. The initial pulse durations are 5 and 10 fs, respectively. The laser intensities are the same for both pulses at time $t = 0$ fs. The attosecond pulses are generated by electrons freed at the time interval between the two dashed lines. The ionization probability at $t = 0$ is 93.4% for the 10 fs pulses. We can conclude that pulses less than 10 fs should be used for generating isolated attosecond pulse with Helium atoms.

4.6 Summary

The semiclassical model is intuitive and easy to use. However, the amplitude information of the attosecond pulses is lost in the calculations. The quantum theory discussed in Chapter 5 allows the calculation of both the amplitude and phase of the attosecond radiation, and is more powerful.

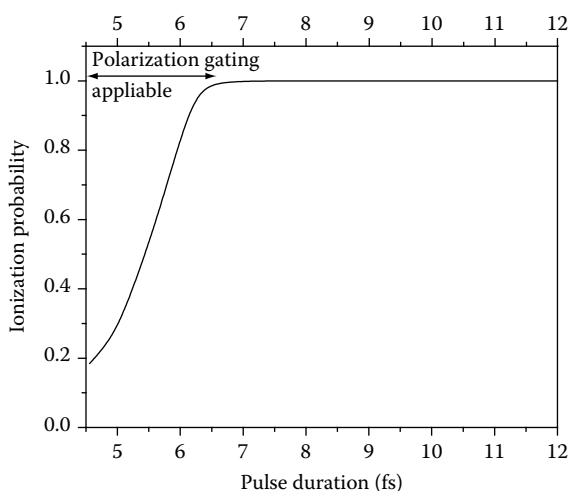


Figure 4.45 Ionization probability of the atom in the ground state.

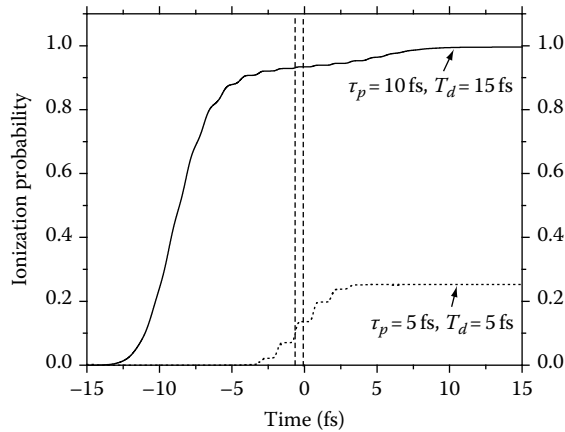


Figure 4.46 The ionization probabilities of a helium atom in laser fields with a time dependent ellipticity. The laser pulse is formed by the combination of a left-hand circularly polarized pulse and a right-hand circularly polarized pulse. Solid line: both circular pulses are 10 fs and the delay between them is 15 fs. The dotted line is obtained when the pulse duration is 5 fs and the delay is 5 fs. The intensity at $t=0$ is 1.4×10^{15} W/cm². The carrier-envelope phase of the laser pulse is $\pi/2$ rad. The high harmonics are generated within the time interval between the two dashed lines. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

Problems

- 4.1 Plot the maximum electron displacement x_0 in the intensity range of 1×10^{14} to 1×10^{15} W/cm² for lasers centered at 0.8 and 1.6 μm .
- 4.2 Plot the ponderomotive energy in the intensity range of 1×10^{14} to 1×10^{15} W/cm² for lasers centered at 0.8 and 1.6 μm .
- 4.3 Plot trajectories for two electrons, one ionized at $\omega_0 t' = \pi/4$ and another at $\omega_0 t' = \pi/2$. The laser period is 2.67 fs and the intensity is 1×10^{15} W/cm². Find the time that the electrons return to the parent ion for the first time.
- 4.4 Find the return time for the two electrons in Problem 4.3. Compare the results with the findings in Problem 4.3.
- 4.5 Plot the return time span for the laser wavelength range of 400–2000 nm.
- 4.6 Plot the return time spans of the long and short trajectory for the laser wavelength range of 400–2000 nm.
- 4.7 Calculate the chirp of the short trajectory at two intensities, 1×10^{14} and 1×10^{15} W/cm² for lasers centered at 0.8 and 1.6 μm . Use the as/eV unit.
- 4.8 Calculate the Keldysh parameter when Xe atoms are placed in a Ti:Sapphire laser field with 1×10^{14} W/cm². Compare that with He atoms in the same laser field.
- 4.9 Calculate the electron tunneling times and the electron velocities in the barrier in Problem 4.8.
- 4.10 For $l=1$, prove that the PPT rate for $m=0$ is larger than those for $m=\pm 1$. (Hint: compare G_{lm} .)

- 4.11 Calculate and plot the ionization rate of an argon atom by laser centered at 790 nm in the intensity range of 3×10^{13} to 3.5×10^{14} W/cm². Compare the magnitude of the rate with that of helium shown in Figure 4.10.
- 4.12 Plot the electron trajectory in a two-color laser field.
- 4.13 Plot the return time in a two-color laser field.
- 4.14 Plot the returning energy in a two-color laser field.
- 4.15 Plot the return time for $\alpha = 0, 0.1$, and 0.2 . The relative phases $\phi_{12} = 0$.
- 4.16 For $F_a/F_0 = 0.01$, calculate the ratio between the cycle averaged ionization rate and the ADK rate.
- 4.17 Calculate the ionization probability of Ar atom with linearly polarized 20 fs lasers at 1×10^{14} W/cm². Compare that with 200 fs lasers at the same intensity. Explain the reason the probability depends on the pulse duration.
- 4.18 Calculate the saturation intensity of argon atoms with linearly polarized 20 fs lasers. Compare that with 200 fs lasers. Explain the reason the saturation intensity depends on the pulse duration.
- 4.19 Calculate the saturation intensity of argon and neon atoms with linearly polarized 20 fs lasers. Explain the reason the saturation intensity depends on the atomic species.
- 4.20 For a 7 fs laser centered at 750 nm, calculate the delay between the two counter-rotating circular pulse so that the polarization gate width is half of optical cycle.
- 4.21 When a quartz plate is used to obtain the delay in Problem 4.20 in Figure 4.44, calculate the thickness of the quartz plate.

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Strong Field Approximation

The semiclassical model provides an intuitive picture of high-harmonic and attosecond pulse generation. One of the successes of the model is the prediction of the cutoff photon energy; however, it does not give the strength of the radiation. The phase information of the harmonic field is also missing to a certain degree.

Although the high-intensity laser–atom interaction problems can be tackled by solving the time-dependent Schrödinger equation (TDSE) numerically, the time it takes to calculate one high harmonic spectrum from a single atom is rather long. It becomes a problem when TDSE is combined with optical wave equations to investigate the phase-matching issues where the spectra of many atoms must be calculated, as discussed in Chapter 6.

It was demonstrated by Lewenstein and his collaborators that the equation can be solved analytically under the strong field approximation (SFA). The analytical solution provides a convenient tool for studying attosecond radiation. It is not only fast but also provides clear explanations of the physical mechanisms of attosecond pulse generation and characterization.

5.1 Analytical Solution of the Schrödinger Equation

5.1.1 Approximations

5.1.1.1 Dipole Radiation and Dipole Moment

Lewenstein et al. have developed an analytical quantum theory to describe high-order harmonic generation from atoms interacting with an arbitrarily polarized laser field. The theory assumes that the harmonic field is radiated by electric dipoles, i.e., the moving electrons in the laser field around the stationary nucleus of an atom, as shown in [Figure 5.1](#).

Suppose that the electron in a dipole oscillates in a sinusoidal motion along the x direction such that

$$x(t) = x_0 \cos(\omega t). \quad (5.1)$$

Then, the dipole moment

$$d(t) = d_0 \cos(\omega t), \quad (5.2)$$

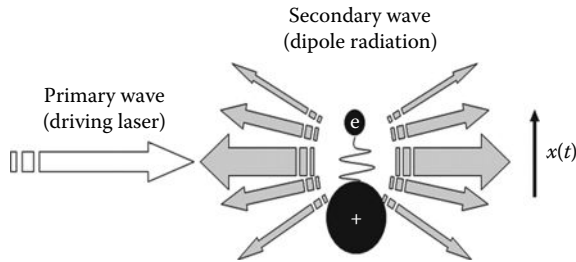


Figure 5.1 Dipole radiation from an atom excited by an external field.

where $d_0 = -ex_0$. In general, dipole moment is a vector in the direction from the negative charge to the positive charge.

According to the classical electrodynamics theory, in the spatial region far from the dipole, the electric field radiated from the dipole is

$$\varepsilon(t, R, \theta) = \frac{d_0 \omega^2}{4\pi\epsilon_0 c^2} \frac{\sin\theta}{R} e^{i\omega(t - \frac{R}{c})}, \quad (5.3)$$

where

ϵ_0 is the electric permittivity of vacuum

c is the speed of light in vacuum R is the distance from the dipole

θ is angle between the R vector and the axis of the dipole

It is interesting to see that the field amplitude is proportional to the square of the frequency; thus, the radiation is more efficient for the high frequencies.

The dipole moment of an atom in a strong laser field is a complicated function of time. Its Fourier transform contains a large component at the driving laser frequency, and many other components at the harmonics of the laser frequency. It is difficult to calculate the absolute amplitude of the dipole moment at each frequency for multielectron atoms such as Argon even by solving TDSE numerically. Here, we try to find a close-form expression for the dipole moment of hydrogen-like atoms.

For simplicity, we sometimes use atomic units in this chapter, i.e., $e = m_e = \hbar = a_0 = 1$, where a_0 is the Bohr radius. The dipole moment can be expressed as $-e\vec{r} = -\vec{r}$, where $\vec{r}$ is the position of the electron when the nucleus is at the origin. In quantum theory, the expectation value of the dipole moment of an atom in the time domain is

$$\vec{r}(t) = \langle \Psi(\vec{r}, t) | \vec{r} | \Psi(\vec{r}, t) \rangle, \quad (5.4)$$

where $\Psi(\vec{r}, t)$ is the time dependent wave function of the system. Once $\vec{r}(t)$ is obtained from the quantum theory, the high harmonic field can be calculated using the classical Equations 5.3.

5.1.1.2 Single Active Electron Approximation

The Schrödinger equation for atoms containing many electrons in a strong laser field is very difficult (if possible) to solve analytically, so we assume that there is only one electron in the atom/ion/molecule that responds to the external laser field. For the sake of argument, we use atom to refer any of these particles. Without the laser field, the Schrödinger equation in a.u. for the system is

$$i\frac{\partial}{\partial t}|\Psi(\vec{r},t)\rangle = \left[-\frac{1}{2}\nabla^2 + V(\vec{r})\right]|\Psi(\vec{r},t)\rangle, \quad (5.5)$$

where

∇^2 is the Laplacian operator

$V(\vec{r})$ is the Coulomb potential of the atom

For the hydrogen atom, the potential is

$$V(r) = -\frac{e^2}{4\pi\epsilon_0 r^2} \text{ (in SI units)} = -\frac{1}{r^2} \text{ (in a.u.)}. \quad (5.6)$$

The wave function for the ground state is then expressed as

$$|\Psi(\vec{r},t)\rangle = |0\rangle e^{\frac{E_g}{\hbar}t} \text{ (in SI units)} = |0\rangle e^{-iI_p t} \text{ (in a.u.)}, \quad (5.7)$$

where the ionization potential $I_p = -E_g = m_e e^4 / (4\pi\epsilon_0)^2 2\hbar^2 = 13.6 \text{ eV} = \frac{1}{2} \text{ a.u.}$ The space-dependent part of the wave function is

$$|0\rangle = \frac{1}{\sqrt{\pi}} \left(\frac{1}{a_0}\right)^{\frac{3}{2}} e^{-\frac{r}{a_0}} \text{ (SI)} = \frac{1}{\sqrt{\pi}} e^{-r} \text{ a.u.}, \quad (5.8)$$

where the Bohr radius is

$$a_0 = \frac{4\pi\epsilon_0\hbar^2}{m_e e^2} \text{ (SI)} = 1 \text{ a.u.} \quad (5.9)$$

As another example, the time independent wave function of the ground state of a hydrogen-like s-wave is

$$|0\rangle = \frac{\alpha^{3/4}}{\sqrt{\pi}} e^{-\sqrt{\alpha}r} \text{ a.u.}, \quad (5.10)$$

where $\alpha = 2I_p$.

5.1.1.3 Electric Dipole Approximation

When the atom is placed in a spatial point where the laser field is $\vec{\mathcal{E}}_L(t)$, the Schrödinger equation under the electric dipole approximation is

$$i\frac{\partial}{\partial t}|\Psi(\vec{r},t)\rangle = \left[-\frac{1}{2}\nabla^2 + V(\vec{r}) - \vec{\mathcal{E}}_L(t) \cdot \vec{r}\right]|\Psi(\vec{r},t)\rangle, \quad (5.11)$$

The term $-e\vec{\mathcal{E}}_L(t) \cdot \vec{r} = -\vec{\mathcal{E}}_L(t) \cdot \vec{r}$ is the potential energy of the electron in the light field.

5.1.1.4 Strong Field Approximation

In the SFA, it is assumed that the intense laser field promotes the ground-state electron directly to the continuum states, bypassing all the excited states. Thus, there are no resonance processes. Only the ground state and the continuum states need to be included in the theory.

The laser intensity for attosecond generation is on the order of 10^{14} W/cm^2 . As revealed in Chapter 4, the freed electron in such a strong field can move 2 nm away before returning to the parent ion. In the spatial region, where the electron motion is accelerated by the laser field, the Coulomb field of the atom is much weaker than the laser field, and, thus, is ignored.

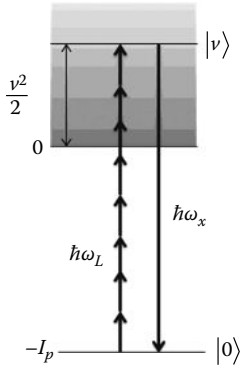


Figure 5.2 Energy levels involved in the SFA.

The SFA is shown in Figure 5.2. This model is valid when the Keldysh parameter is much less than one and the photon energy of the dipole radiation is much larger than the ionization potential of the atom.

5.1.1.5 Continuum-State Wave Function

In atomic units, the electron mass $m_e = 1$, therefore the mechanical momentum is $m_e \vec{v} = \vec{v}$, where $\vec{v}$ is the velocity of the electron. In quantum mechanics, the wave function for a free electron travelling with a velocity $\vec{v}$ is

$$|\vec{v}\rangle e^{i\omega_e t} = e^{i(\omega_e t - \vec{k}_e \cdot \vec{r})}. \quad (5.12)$$

The wave vector is expressed as

$$\vec{k}_e = \frac{m_e \vec{v}}{\hbar} = \vec{v} \text{ a.u.}, \quad (5.13)$$

and the frequency as

$$\omega_e = \frac{\frac{1}{2} m_e v^2}{\hbar} = \frac{1}{2} v^2 \text{ a.u.} \quad (5.14)$$

Thus, $|\vec{v}\rangle = e^{-i\vec{v} \cdot \vec{r}}$.

5.1.1.6 Total Wave Function

After making the strong field assumptions, the Schrödinger Equation 5.11 has an analytical solution

$$|\Psi(\vec{r}, t)\rangle = e^{-iI_p t} \left[a(t) |0\rangle + \int d^3 \vec{v} b(\vec{v}, t) |\vec{v}\rangle \right], \quad (5.15)$$

where $a(t)$ is the amplitude of wave function of the ground state, $|0\rangle$. $b(\vec{v}, t)$ is related to the population of the continuum state $|\vec{v}\rangle$. Notice that a time-dependent term $e^{-iI_p t}$ is factored out from the continuum wave function.

Another natural way to expand the wave function would be

$$|\Psi(\vec{r}, t)\rangle = a(t) |0\rangle e^{-iI_p t} + \int d^3 \vec{v} b(\vec{v}, t) |\vec{v}\rangle e^{i\omega_e t}. \quad (5.16)$$

Thus, $b(\vec{v}, t)$ is the wave function of the freed electron in momentum space. We will use the expression 5.15 in this chapter to be consistent with what has been used in the literature.

5.1.1.7 Dipole Moment

Inserting Equation 5.15 into Equation 5.4, we get

$$\begin{aligned} \vec{r}(t) &= a^2(t) \langle 0 | \vec{r} | 0 \rangle + \int d^3 \vec{v} a^*(t) b(\vec{v}, t) \langle 0 | \vec{r} | \vec{v} \rangle \\ &\quad + \int d^3 \vec{v} a(t) b(\vec{v}, t) \langle \vec{v} | \vec{r} | 0 \rangle + \int d^3 \vec{v} b^2(\vec{v}, t) \langle \vec{v} | \vec{r} | \vec{v}' \rangle. \end{aligned} \quad (5.17)$$

Using the notation of a dipole transition-matrix element from the ground state to the continuum state,

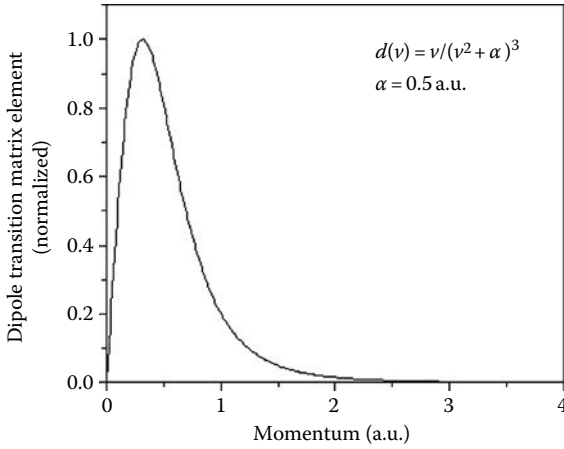


Figure 5.3 Dipole transition matrix element for 1s state.

$$\vec{d}(\vec{v}) = \langle \vec{v} | \vec{r} | 0 \rangle, \quad (5.18)$$

and ignoring the continuum–continuum contribution, $\langle \vec{v} | \vec{r} | \vec{v}' \rangle$, as well as the slow variation term, $a^2(t) \langle 0 | \vec{r} | 0 \rangle$, we have

$$\vec{r}(t) = \int d^3\vec{v} a^*(t) b(\vec{v}, t) \vec{d}^*(\vec{v}) + c.c. \quad (5.19)$$

The physical meaning of this equation is that the dipole radiation for high harmonic generation originated from the recombination of the returning electron wave $b(\vec{v}, t)$ with the ground state with wave function amplitude $a(t)$. This corresponds to the third step of the semiclassical model. Apparently, if the ground state is completely depleted by laser-field ionization when the electron returns, the dipole radiation is seized.

The dipole transition-matrix elements have been studied extensively for atom–synchrotron interaction in the past and can therefore be considered as known quantities. For example, for the 1s state of a hydrogen-like atom,

$$\vec{d}_{1s}(\vec{v}) = i \frac{2^{7/2}}{\pi} \alpha^{5/4} \frac{\vec{v}}{(v^2 + \alpha)^3}, \quad (5.20)$$

which is plotted in Figure 5.3.

The ground-state population, $a(t)$, can be calculated using the PPT or ADK ionization rate discussed in Chapter 4. The only unknown quantity is $b(\vec{v}, t)$.

5.1.2 Continuum Wave Packet

Since many continuum states are populated when the atom is ionized by the strong laser field, a wave packet is formed. The wave packet also evolves in the laser field, which is described by $b(\vec{v}, t)$. Only the portion of the wave packet that returns to the parent ion contributes the high harmonic-generation process. It can be shown that

$$b(\vec{v}, t) = i \int_{-\infty}^t dt' a(t') \vec{\varepsilon}_L(t') \cdot \vec{d} \left[\vec{v} + \vec{A}(t) - \vec{A}(t') \right] e^{iS[\vec{v} + \vec{A}(t) - \vec{A}(t'), t, t']}, \quad (5.21)$$

which is obtained by inserting Equation 5.15 into Equation 5.11. The product $\vec{\varepsilon}_L(t') \cdot \vec{d} \left[\vec{v} + \vec{A}(t) - \vec{A}(t') \right]$ describes the population of the state $|\vec{v}\rangle$ due to the ionization by the laser field at time t' .

The quantity

$$S[\vec{v} + \vec{A}(t) - \vec{A}(t'), t, t'] = \int_{t'}^t dt'' \left[\frac{1}{2} \left(\vec{v} + \vec{A}(t) - \vec{A}(t'') \right)^2 + I_p \right] \quad (5.22)$$

corresponds to the classical action of the electron in the laser field, $\vec{\varepsilon}_L(t) = -\frac{\partial}{\partial t} \vec{A}(t)$, which is the phase that the electron wave acquires from the electron birth time t' to the time t . Equations 5.21 and 5.22 are the close-form solutions of the TDSE. It still contains two integrals, which can be further simplified for some special cases.

5.1.2.1 Analytical Approach to Solve the Schrödinger Equation

The derivations of the expression 5.21 are tedious, but it worthwhile to go through. The left side of Equation 5.11 is

$$\begin{aligned} i \frac{\partial}{\partial t} |\Psi\rangle &= i \frac{\partial}{\partial t} \left[e^{-iI_p t} \left[|0\rangle + \int d^3 \vec{v} b(t) e^{-i\vec{v} \cdot \vec{r}} \right] \right] \\ &= i \left[-iI_p |0\rangle e^{-iI_p t} - iI_p e^{-iI_p t} \int d^3 \vec{v} \dot{b}(t) e^{-i\vec{v} \cdot \vec{r}} + e^{-iI_p t} \int d^3 \vec{v} \dot{b} e^{-i\vec{v} \cdot \vec{r}} \right] \\ &= e^{-iI_p t} \left[I_p |0\rangle + I_p \int d^3 \vec{v} b e^{-i\vec{v} \cdot \vec{r}} + i \int d^3 \vec{v} \dot{b} e^{-i\vec{v} \cdot \vec{r}} \right], \end{aligned} \quad (5.23)$$

where $\dot{b} = \frac{\partial b}{\partial t}$.

The right side of Equation 5.11 is expressed as

$$\begin{aligned} &\left(-\frac{\nabla^2}{2} + V - \varepsilon_L x \right) |\Psi\rangle \\ &= e^{-iI_p t} \left(-\frac{\nabla^2}{2} + V - \varepsilon_L x \right) \left[|0\rangle + \int d^3 \vec{v} b e^{-i\vec{v} \cdot \vec{r}} \right] \\ &= e^{-iI_p t} \left\{ \left(-\frac{\nabla^2}{2} + V \right) |0\rangle + \left(-\frac{\nabla^2}{2} + V \right) \int d^3 \vec{v} b e^{-i\vec{v} \cdot \vec{r}} \right. \\ &\quad \left. \times \int d^3 \vec{v} b e^{-i\vec{v} \cdot \vec{r}} - \varepsilon_L x |0\rangle - \varepsilon_L x \int d^3 \vec{v} b e^{-i\vec{v} \cdot \vec{r}} \right\} \\ &= e^{-iI_p t} \left[I_p |0\rangle + \left(\frac{v^2}{2} + V \right) \int d^3 \vec{v} b e^{-i\vec{v} \cdot \vec{r}} - \varepsilon_L x |0\rangle \right. \\ &\quad \left. - \varepsilon_L \left(i \frac{\partial}{\partial v_x} \right) \int d^3 \vec{v} b e^{-i\vec{v} \cdot \vec{r}} \right]. \end{aligned} \quad (5.24)$$

In the last step, we use the results $\left(-\frac{\nabla^2}{2} + V\right)|0\rangle = I_p|0\rangle$, which is the time-independent Schrödinger equation. We also use the equations $\nabla^2 e^{-i\vec{v}\cdot\vec{r}} = -v^2$ and $x = i\frac{\partial}{\partial v_x}$.

Let the last expression of Equation 5.23 equal the last expression of Equation 5.24, we have

$$\begin{aligned}\int d^3\vec{v}\dot{b}|\vec{v}\rangle &= -i\left(\frac{v^2}{2} - I_p + V\right) \int d^3\vec{v}be^{-i\vec{v}\cdot\vec{r}} - \varepsilon_L\left(\frac{\partial}{\partial v_x}\right) \int d^3\vec{v}be^{-i\vec{v}\cdot\vec{r}} + i\varepsilon_L x|0\rangle \\ &= -i\left(\frac{v^2}{2} - I_p + V\right) \int d^3\vec{v}b|\vec{v}\rangle - \varepsilon_L \int d^3\vec{v}\frac{\partial b}{\partial v_x}|\vec{v}\rangle + i\varepsilon_L x|0\rangle. \quad (5.25)\end{aligned}$$

Finally

$$\begin{aligned}\left\langle\vec{v}'\left|\int d^3\vec{v}\dot{b}\right|\vec{v}\right\rangle &= -i\left\langle\vec{v}'\left|\left(\frac{v^2}{2} - I_p + V\right) \int d^3\vec{v}b\right|\vec{v}\right\rangle - \varepsilon_L \\ &\quad \times \left\langle\vec{v}'\left|\int d^3\vec{v}\frac{\partial b}{\partial v_x}\right|\vec{v}\right\rangle + i\varepsilon_L\langle\vec{v}'|x|0\rangle. \quad (5.26)\end{aligned}$$

The normalization of the wave function means $\int d^3\vec{v}\langle\vec{v}'|\vec{v}\rangle = 1$, thus

$$\begin{aligned}\frac{db}{dt} &= -i\left(\frac{v^2}{2} - I_p\right)b - i\left\langle\vec{v}'\left|V(r) \int d^3\vec{v}b\right|\vec{v}\right\rangle \\ &\quad - \varepsilon_L\frac{\partial b}{\partial v_x} + i\varepsilon_L\langle\vec{v}'|x|0\rangle \quad (5.27)\end{aligned}$$

For the continuum state, $r \gg 1$ a.u., $V(r) = \frac{1}{r^2} \approx 0$

$$\frac{db}{dt} = -i\left(\frac{v^2}{2} - I_p\right)b - \varepsilon_L\frac{\partial b}{\partial v_x} + i\varepsilon_L \cdot d_x(\vec{v}). \quad (5.28)$$

5.1.2.2 Solution of the Differential Equation

The solution of the equation is

$$\begin{aligned}b(\vec{v}, t) &= i \int_{-\infty}^t dt' \varepsilon_L(t') d_x \\ &\quad \times \left[\vec{v} + \vec{A}(t) - \vec{A}(t')\right] e^{-i \int_{t'}^t dt'' \left[(\vec{v} + \vec{A}(t) - \vec{A}(t''))^2 / 2 + I_p\right]}, \quad (5.29)\end{aligned}$$

which can be obtained in the following way.

First, we rewrite Equation 5.28 as

$$\frac{db}{dt} + i\left(\frac{v^2}{2} - I_p - i\varepsilon_L(t)\frac{\partial}{\partial v_x}\right)b = i\varepsilon_L(t) \cdot d_x(\vec{v}). \quad (5.30)$$

This can then be written as

$$\frac{db}{dt} + g(t)b = h(t), \quad (5.31)$$

where $h(t) = i\varepsilon_L(t)d_x[\vec{v}(t)]$, and $g(t) = i\left[\frac{v^2(t)}{2} + I_p\right] + \varepsilon_L(t)\frac{\partial}{\partial v_x} \approx i\left[\frac{v^2(t)}{2} + I_p\right]$. We dropped the real part of $g(t)$ because its effects are much smaller than the fast phase introduced by the imaginary part.

Multiply both sides by $\mu(t) = e^{\int g(t)dt}$ and we have

$$\mu(t) \left[\frac{db}{dt} + g(t)b \right] = \mu(t)h(t), \quad (5.32)$$

which is equivalent to

$$\frac{d}{dt} [\mu(t)b(t)] = \mu(t)h(t). \quad (5.33)$$

The solution to Equation 5.33 is

$$b(t) = \frac{1}{\mu(t)} \int \mu(t)h(t)dt + C. \quad (5.34)$$

The initial condition is $b(-\infty) = 0$, i.e., the continuum state is unpopulated before the laser pulse arrive, therefore $C = 0$, and

$$b(t) = \frac{1}{\mu(t)} \int_{-\infty}^t h(t')\mu(t')dt'. \quad (5.35)$$

The physical meaning of this equation is that the population is the accumulation of electrons ionized by the laser field at time t' .

Using this result, we know the solution of Equation 5.30 is

$$b(\vec{v}, t) = i \frac{1}{e^{\int_{-\infty}^t g(t')dt'}} \int_{-\infty}^t dt' \vec{\varepsilon}_L(t') \cdot \vec{d}[\vec{v}(t')] e^{\int_{t'}^t g(t'')dt''}. \quad (5.36)$$

Or

$$b(\vec{v}, t) = i \frac{1}{e^{i \int_{-\infty}^t [\frac{1}{2}v^2(t') + I_p] dt'}} \int_{-\infty}^t dt' \vec{\varepsilon}_L(t') \cdot \vec{d}[\vec{v}(t')] e^{i \int_{t'}^t [\frac{1}{2}v^2(t'') + I_p] dt''}. \quad (5.37)$$

5.1.2.3 Conservation of Canonical Momentum

In the laser field, the canonical momentum is conserved. The canonical momentum of a particle with charge q is defined as

$$\vec{p} = \vec{v}(t) + q\vec{A}(t). \quad (5.38)$$

For an electron, $q = -e$, thus

$$\vec{p} = \vec{v}(t) - e\vec{A}(t). \quad (5.39)$$

But, in atomic units, $e = 1$, so

$$\vec{p} = \vec{v}(t) - \vec{A}(t). \quad (5.40)$$

The canonical momentum at the time, t' , when an electron is freed, is equal to that at a later time t , which leads to

$$\vec{p} = \vec{v}(t) - \vec{A}(t) = \vec{v}(t') - \vec{A}(t'). \quad (5.41)$$

Or

$$\vec{v}(t') = \vec{v}(t) - \vec{A}(t) + \vec{A}(t'). \quad (5.42)$$

For laser field with arbitrary polarization state, we replace $\vec{d}_x$ by $\vec{d}$. Finally, we have

$$b(\vec{v}, t) = i \frac{1}{e^{i \int_{-\infty}^t \left(\frac{1}{2} [\vec{v} - \vec{A}(t) + \vec{A}(t')]^2 + I_p \right) dt'}} \int_{-\infty}^t dt' \vec{\varepsilon}_L(t') \cdot \vec{d} \left[\vec{v} - \vec{A}(t) + \vec{A}(t') \right] e^{i \int_{t'}^t \left\{ \frac{1}{2} [\vec{v} - \vec{A}(t) + \vec{A}(t'')]^2 + I_p \right\} dt''}. \quad (5.43)$$

Or

$$b(\vec{v}, t) = i e^{-i \int_{-\infty}^t dt' \frac{1}{2} [\vec{v} + \vec{A}(t) - \vec{A}(t')]^2} \int_{-\infty}^t dt' \vec{\varepsilon}_L(t') \cdot \vec{d} \left[\vec{v} + \vec{A}(t) - \vec{A}(t') \right] e^{iS}, \quad (5.44)$$

where

$$S = \int_{t'}^t \left\{ \frac{1}{2} \left[\vec{v} - \vec{A}(t) + \vec{A}(t'') \right]^2 + I_p \right\} dt'' \quad (5.45)$$

corresponds to the action the electron acquires in the laser field.

Equation 5.45 can be approximated by

$$b(\vec{v}, t) \approx i e^{-i \frac{1}{2} v^2 t} \int_{-\infty}^t dt' \vec{\varepsilon}_L(t') \cdot \vec{d} \left[\vec{v} + \vec{A}(t) - \vec{A}(t') \right] e^{iS}. \quad (5.46)$$

The total plane-wave function of the continuum state component with momentum $\vec{v}$ in the laser field is

$$\left\{ i \int_{-\infty}^t dt' \vec{\varepsilon}_L(t') \cdot \vec{d} \left[\vec{v} + \vec{A}(t) - \vec{A}(t') \right] e^{iS} \right\} e^{i(\vec{v} \cdot \vec{r} - \frac{1}{2} v^2 t)}. \quad (5.47)$$

The term $i \int_{-\infty}^t dt' \vec{\varepsilon}_L(t') \cdot \vec{d} \left[\vec{v} + \vec{A}(t) - \vec{A}(t') \right] e^{iS}$ is the complex amplitude of the electron wave component. It means that electrons ionized in the time range t' to t may all contribute to the momentum component $\vec{v}$ at time t . The product $\vec{\varepsilon}_L(t') \cdot \vec{d} \left[\vec{v} + \vec{A}(t) - \vec{A}(t') \right]$ is essentially the ionization rate at the time t' , which can be replaced by the more accurate PPT or ADK rare introduced in Chapter 4.

5.1.3 Saddle-Point Approach

By replacing the classical mechanical momentum, $\vec{v}$, with the canonical moment, $\vec{p}(t) = \vec{v} + \vec{A}(t)$, the dipole moment can be expressed as

$$\vec{r}(t) = \int d^3 \vec{p} a^*(t) b(\vec{p}, t) \vec{d}^* [\vec{p} - \vec{A}(t)] + c.c., \quad (5.48)$$

where

$$b(\vec{p}, t) \approx i \int_{-\infty}^t dt' \vec{\varepsilon}_L(t') \cdot \vec{d} [\vec{p} - \vec{A}(t')] a(t') e^{-iS(\vec{p}, t, t')}. \quad (5.49)$$

Therefore, the dipole moment of an atom in the time domain is

$$\begin{aligned} \vec{r}(t) = i \int_{-\infty}^t dt' \int d^3\vec{p} a^*(t) \vec{d}^* [\vec{p} - \vec{A}_L(t)] e^{-iS(\vec{p}, t, t')} a(t') \vec{\varepsilon}_L(t') \\ \cdot \vec{d} [\vec{p} - \vec{A}_L(t')] + c.c. \end{aligned} \quad (5.50)$$

The Fourier transform of $\vec{r}(t)$ gives the high harmonic spectrum.

Equation 5.50 can be explained with the three step semiclassical model. The term $\vec{\varepsilon}_L(t') \cdot \vec{d} [\vec{p} - \vec{A}_L(t')]$ describes the transition from the ground state to the continuum at time t' through field ionization in the laser field. The electron wave in the continuum propagates until time t and acquires a phase factor equal to $e^{iS(\vec{p}, t, t')}$. The kinetic energy gained by the electrons is expressed by the quasi-classical action S . The electron recombines at time t with an amplitude equal to $\vec{d}^* [\vec{p} - \vec{A}_L(t)]$, which generates the XUV light. The ground-state amplitude $a(t')$ at the time of electron release is added to take into account the depletion of the ground-state population.

5.1.3.1 One-Dimensional Saddle Point Approximation

The integral over the momentum can be performed approximately by using the saddle point method. We introduce the one-dimensional (1D) saddle point method first, which is then extended to the three-dimensional (3D) case.

The value of a function $f(x)$ near the point x_0 can be estimated by the Taylor Expansion

$$f(x) = f(x_0) + f'(x_0)(x - x_0) + \frac{1}{2}f''(x_0)(x - x_0)^2 + \dots \quad (5.51)$$

When $f'(x_0) = \frac{d}{dx}f(x)|_{x_0} = 0$, and the high order can be neglected, i.e.,

$$f(x) = f(x_0) + \frac{1}{2}f''(x_0)(x - x_0)^2. \quad (5.52)$$

The integral

$$\begin{aligned} \int_a^b e^{f(x)} dx &= \int_a^b e^{f(x_0) + \frac{1}{2}f''(x_0)(x-x_0)^2} dx \\ &= e^{f(x_0)} \int_{-\infty}^{+\infty} e^{\frac{1}{2}f''(x_0)(x-x_0)^2} dx. \end{aligned} \quad (5.53)$$

Using the Gaussian Integral formula

$$\int_{-\infty}^{+\infty} e^{-x^2} dx = \sqrt{\pi}, \quad (5.54)$$

we have

$$\int_a^b e^{f(x)} dx = \sqrt{\frac{2\pi}{f''(x_0)}} e^{f(x_0)}. \quad (5.55)$$

This approach to perform the integral is called the saddle point method.

5.1.3.2 3D Saddle-Point Method

In the 3D case, the Taylor expansion is

$$\begin{aligned} f(x, y, z) &\approx f(x_0, y_0, z_0) + f_x(x_0, y_0, z_0)(x - x_0) + f_y(x_0, y_0, z_0)(y - y_0) \\ &\quad + f_z(x_0, y_0, z_0)(z - z_0) \\ &\quad + \frac{1}{2} \begin{bmatrix} f_{xx}(x_0, y_0, z_0)(x - x_0)^2 + f_{yy}(x_0, y_0, z_0)(y - y_0)^2 + f_{zz}(x_0, y_0, z_0)(z - z_0)^2 \\ + 2f_{xy}(x_0, y_0, z_0)(x - x_0)(y - y_0) \\ + 2f_{xz}(x_0, y_0, z_0)(x - x_0)(z - z_0) \\ + 2f_{yz}(x_0, y_0, z_0)(y - y_0)(z - z_0) \end{bmatrix}. \end{aligned} \quad (5.56)$$

Consider the case when $f_x(x_0, y_0, z_0) = f_y(x_0, y_0, z_0) = f_z(x_0, y_0, z_0) = 0$, but the second-order derivatives are not zero. The point x_0, y_0, z_0 is called the saddle point, which means the gradient at x_0, y_0, z_0 is zero.

At this point,

$$\begin{aligned} f(x, y, z) &= f(x_0, y_0, z_0) \\ &\quad + \frac{1}{2} \begin{bmatrix} f_{xx}(x_0, y_0, z_0)(x - x_0)^2 + f_{yy}(x_0, y_0, z_0)(y - y_0)^2 + f_{zz}(x_0, y_0, z_0)(z - z_0)^2 \\ + 2f_{xy}(x_0, y_0, z_0)(x - x_0)(y - y_0) \\ + 2f_{xz}(x_0, y_0, z_0)(x - x_0)(z - z_0) \\ + 2f_{yz}(x_0, y_0, z_0)(y - y_0)(z - z_0) \end{bmatrix}. \end{aligned} \quad (5.57)$$

The integral

$$\int_a^b \int \int e^{f(x, y, z)} dx dy dz \approx e^{f(x_0, y_0, z_0)} \int \int \int_{-\infty}^{+\infty} e^{\frac{1}{2}[g(x, y, z)]} dx dy dz, \quad (5.58)$$

where $g(x, y, z)$ is the second-order term in Equation 5.54.

Suppose A is a symmetric positive-definite invertible covariant tensor of rank two. Then,

$$\int \int \int_{-\infty}^{+\infty} e^{\frac{1}{2}[g(x, y, z)]} dx dy dz = \int_{-\infty}^{+\infty} e^{-\frac{1}{2}A_{ij}x^i x^j} d^3x = \sqrt{\frac{(2\pi)^3}{\det A}}. \quad (5.59)$$

The elements of the tensor are

$$\begin{aligned} A_{11} &= f_{xx}(x_0, y_0, z_0), A_{12} = f_{xy}(x_0, y_0, z_0), A_{13} = f_{xz}(x_0, y_0, z_0), \\ A_{21} &= f_{yx}(x_0, y_0, z_0), A_{22} = f_{yy}(x_0, y_0, z_0), A_{23} = f_{yz}(x_0, y_0, z_0), \\ A_{31} &= f_{zx}(x_0, y_0, z_0), A_{32} = f_{zy}(x_0, y_0, z_0), A_{33} = f_{zz}(x_0, y_0, z_0). \end{aligned} \quad (5.60)$$

The tensor is symmetric, because

$$\begin{aligned} A_{12} &= A_{21} = f_{xy}(x_0, y_0, z_0), \quad A_{13} = A_{31} = f_{xz}(x_0, y_0, z_0), \quad A_{23} \\ &= A_{32} = f_{yz}(x_0, y_0, z_0). \end{aligned} \quad (5.61)$$

The tensor is positive-definite, since the point (x_0, y_0, z_0) is the saddle point. The determinant

$$\begin{aligned} \text{Det}(A) &= \begin{vmatrix} A_{11} & A_{12} & A_{13} \\ A_{21} & A_{22} & A_{23} \\ A_{31} & A_{32} & A_{33} \end{vmatrix} = \begin{vmatrix} A_{11} & A_{12} & A_{13} \\ A_{12} & A_{22} & A_{23} \\ A_{13} & A_{23} & A_{33} \end{vmatrix} \\ &= A_{11}A_{22}A_{33} - A_{13}^2A_{22} + A_{12}A_{23}A_{13} - A_{12}^2A_{33} \\ &\quad + A_{13}A_{23}A_{12} - A_{23}^2A_{11}. \end{aligned} \quad (5.62)$$

The saddle point condition of the quasi-classical action

$$S(\vec{p}, t, t') = \int_{t'}^t dt'' \left\{ \frac{[\vec{p} - \vec{A}(t'')]^2}{2} + I_p \right\} \quad (5.63)$$

is defined as

$$\nabla_{\vec{p}} S = \int_{t'}^t dt'' \nabla_{\vec{p}} \left\{ \frac{[\vec{p} - \vec{A}(t'')]^2}{2} + I_p \right\} = 0, \quad (5.64)$$

which leads to

$$\int_{t'}^t [\vec{p}_s - \vec{A}(t'')] dt'' = 0, \quad (5.65)$$

where $\vec{p}_s$ is the saddle point. Thus

$$\vec{p}_s \cdot (t - t') - \int_{t'}^t \vec{A}(t'') dt'' = 0. \quad (5.66)$$

Finally, the momentum is

$$\vec{p}_s(t - t') = \int_{t'}^t \frac{\vec{A}(t'') dt''}{(t - t')}. \quad (5.67)$$

The action can also be expressed as

$$\begin{aligned} S &= \frac{1}{2} \int_{t'}^t \left[(p_x^2 + p_y^2 + p_z^2) - 2(p_x \hat{i} + p_y \hat{j} + p_z \hat{k}) \vec{A}(t'') \right. \\ &\quad \left. + A(t'')^2 + 2I_p \right] dt''. \end{aligned} \quad (5.68)$$

Thus

$$\begin{aligned} A_{11} &= \frac{\partial}{\partial p_x} \left(\frac{\partial}{\partial p_x} S \right) = \frac{\partial}{\partial p_x} \left\{ \frac{1}{2} \int_{t'}^t [2p_x - 2i\vec{A}(t'')] dt'' \right\} \\ &= \int_{t'}^t 1 dt'' = t - t', \end{aligned} \quad (5.69)$$

$$\begin{aligned} A_{22} &= \frac{\partial}{\partial p_y} \left(\frac{\partial}{\partial p_y} S \right) = \frac{\partial}{\partial p_y} \left\{ \frac{1}{2} \int_{t'}^t [2p_y - 2\hat{J}\vec{A}(t'')] dt'' \right\} \\ &= \int_{t'}^t 1 dt'' = t - t', \end{aligned} \quad (5.70)$$

and

$$\begin{aligned} A_{33} &= \frac{\partial}{\partial p_z} \left(\frac{\partial}{\partial p_z} S \right) = \frac{\partial}{\partial p_z} \left\{ \frac{1}{2} \int_{t'}^t [2p_z - 2\hat{K}\vec{A}(t'')] dt'' \right\} \\ &= \int_{t'}^t 1 dt'' = t - t'. \end{aligned} \quad (5.71)$$

Since

$$\frac{\partial}{\partial p_x} \left(\frac{\partial}{\partial p_y} S \right) = \frac{\partial}{\partial p_x} \left\{ \frac{1}{2} \int_{t'}^t [2p_y - 2\hat{J}\vec{A}(t'')] dt'' \right\} = 0, \quad (5.72)$$

i.e.,

$$A_{12} = A_{13} = A_{23} = 0. \quad (5.73)$$

$$\begin{aligned} Det(A) &= \left[\frac{\partial}{\partial p_x} \left(\frac{\partial}{\partial p_x} iS \right) \right] \left[\frac{\partial}{\partial p_y} \left(\frac{\partial}{\partial p_y} iS \right) \right] \left[\frac{\partial}{\partial p_z} \left(\frac{\partial}{\partial p_z} iS \right) \right] \\ &= -i(t - t')^3, \end{aligned} \quad (5.74)$$

$$\int_{-\infty}^{+\infty} e^{-\frac{i}{2} A_{ij} x^i x^j} d^3 x = \sqrt{\frac{(2\pi)^3}{-i(t - t')^3}} = i \left(\frac{\pi}{i(t - t')/2} \right)^{3/2}. \quad (5.75)$$

Finally, the dipole moment contains a time integral only

$$\begin{aligned} \vec{r}(t) &= i \int_{-\infty}^t dt' \left(\frac{\pi}{i(t - t')/2} \right)^{3/2} \vec{d}^* [\vec{p}_s(t, t') - \vec{A}_L(t)] e^{iS(\vec{p}_s, t, t')} \vec{e}_L(t') \\ &\quad \cdot \vec{d} [\vec{p}_s(t, t') - \vec{A}_L(t')] + c.c. \end{aligned} \quad (5.76)$$

If we let $\tau = t - t'$, then we have

$$\vec{r}(t) = i \int_0^\infty d\tau \left(\frac{\pi}{\epsilon + i\tau/2} \right)^{3/2} \vec{d}^* [\vec{p}_s(t, \tau) - \vec{A}_L(t)] e^{iS(\vec{p}_s, t, \tau)} \vec{e}_L(t - \tau) \cdot \vec{d}(\vec{p}_s(t, \tau) - \vec{A}_L(t - \tau) + c.c. \quad (5.77)$$

where $\vec{p}_s(t, \tau) = \frac{\int_{t-\tau}^t \vec{A}_L(t'') dt''}{\tau}$. The infinitesimal ϵ comes from the regularized Gaussian integration around the saddle point.

Since

$$\vec{v}(t'') = \vec{p}_s - \vec{A}_L(t''), \quad (5.78)$$

and

$$x(t) - x(t') = \int_{t'}^t \vec{v}(t'') dt'', \quad (5.79)$$

we have

$$x(t) - x(t') = \int_{t'}^t [\vec{p}_s - \vec{A}_L(t'')] dt'' = \nabla_{\vec{p}} S = 0. \quad (5.80)$$

i.e., the electron born at t' returns to the same position at time t , which is another physical meaning of the saddle point. The saddle point method is also called stationary phase method because the action S is related to the phase of the electron in the continuum state. It is stationary around the saddle point $\vec{p}_s$ where $\nabla_{\vec{p}} S = 0$.

5.1.4 Dipole Moment for Linearly Polarized Driving Laser

5.1.4.1 Laser Field

Equation 5.77 is valid for driving lasers with arbitrary time profile and polarization state, such as the one for the polarization gating. For simplicity, in this section, we consider the case while the laser field is linearly polarized along the x direction and is monochromatic, i.e.,

$$\mathcal{E}(t) = E_0 \cos(\omega_0 t). \quad (5.81)$$

For studying high-order harmonic generation, we can choose the unit of the frequency to be the fundamental frequency of the laser. Using such a unit, the laser field can be simplified as

$$\mathcal{E}(t) = E_0 \cos(t), \quad (5.82)$$

$$A_x(t) = -E_0 \sin(t), \quad (5.83)$$

$$A_y(t) = A_z(t) = 0. \quad (5.84)$$

Then the unit of the time is $1/(2\pi)$ rad. In other words, $t = 2\pi$ corresponds to one laser cycle.

The x component of the dipole moment is

$$x(t) = i \int_0^\infty d\tau \left(\frac{\pi}{\epsilon + i\tau/2} \right)^{3/2} d_x^* [p_{sx}(t, \tau) - A_x(t)] \times e^{iS(p_{sx}, t, \tau)} E_0 \cos(t - \tau) d_x [p_{sx}(t, \tau) - A_x(t - \tau)] + c.c. \quad (5.85)$$

5.1.4.2 Momentum and Action

The stationary value of the canonical momentum component along the x direction is

$$p_{sx}(t, \tau) = \int_{t-\tau}^t A_x(t') dt' / \tau = E_0 [\cos(t) - \cos(t - \tau)] / \tau, \quad (5.86)$$

which is shown in Figure 5.4. It reaches its maximum when the releasing time is within one laser cycle.

The y and z components of the momentum are zero, because

$$p_{sy}(t, \tau) = \int_{t-\tau}^t A_y(t') dt' / \tau = 0, \quad (5.87)$$

$$p_{sz}(t, \tau) = \int_{t-\tau}^t A_z(t') dt' / \tau = 0. \quad (5.88)$$

The action corresponding to the stationary phase is

$$S_s(p_{sx}, t, \tau) = \int_{t-\tau}^t dt' \left\{ \frac{1}{2} [p_{sx} - A_x(t')]^2 + I_p \right\} = (I_p + U_p) \tau - \frac{2U_p [1 - \cos(\tau)]}{\tau} - U_p C(\tau) \cos(2t - \tau), \quad (5.89)$$

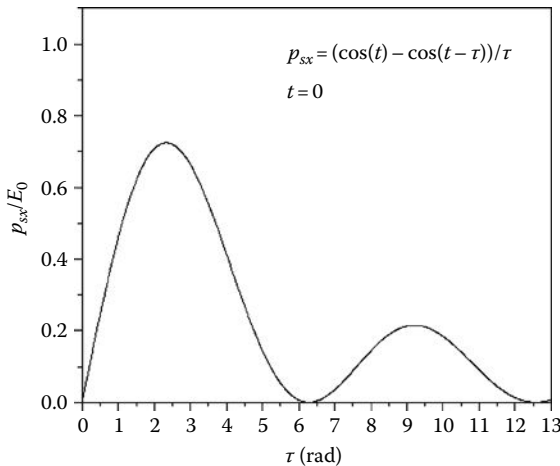


Figure 5.4 Canonical momentum as a function of the releasing time.

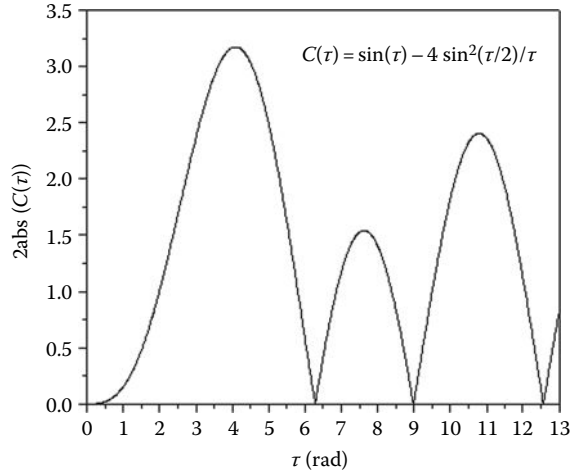


Figure 5.5 Function $2|C(\tau)|$.

where

$$C(\tau) = \sin(\tau) - \frac{4 \sin^2(\tau/2)}{\tau}. \quad (5.90)$$

The range of action changing with t is $2U_p|C(\tau)|$, which is a function of return time τ , as shown in Figure 5.5. The action can be defined as the integral of the kinetic energy (plus I_p), over t' , which is the reason that $2U_p|C(\tau)|$ is the kinetic energy gain of the electron at t ,

$$K(t) - K(t - \tau) = \frac{1}{2} [p_{sx}(t, \tau) - A_x(t)]^2 - \frac{1}{2} [p_{sx}(t, \tau) - A_x(t - \tau)]^2. \quad (5.91)$$

As is shown in Chapter 4, the kinetic energy of the return electron normalized by the ponderomotive energy calculated by solving Newton's equation is

$$\frac{K(t, \tau)}{U_p} = 2[\sin(t) - \sin(t - \tau)]^2. \quad (5.92)$$

It has been shown that the results calculated by this equation are very close to the quantum results

$$2|C(\tau)| = 2 \left| \sin(\tau) - \frac{4 \sin^2(\tau/2)}{\tau} \right|. \quad (5.93)$$

5.1.5 Dipole Transition Matrix Element

The field-free dipole matrix is given by $\vec{d}(\vec{v}) = \langle \vec{v} | \vec{r} | 0 \rangle$. Suppose that $u(\vec{r})$ is the wave function of electron in the continuum state. In the SFA model, the wave function is approximated by a plane wave:

$$|\vec{v}\rangle = u(\vec{r}) = \frac{1}{(2\pi)^{3/2}} e^{-i\vec{k} \cdot \vec{r}}. \quad (5.94)$$

$|0\rangle = u_b(\vec{r})$ is the field-free ground-state wave function of the atom.

For hydrogen-like atoms:

$$u_b(r, \theta, \phi) = R_{nl}(r) Y_{lm}(\theta, \phi), \quad (5.95)$$

where $Y_{lm}(\theta, \phi)$ is the spherical harmonic and the radial eigenfunction is

$$R_{nl}(r) = \frac{2^{l+1}}{(n+l)!} \left(\frac{(n-l-1)!}{n(n+1)!} \right)^{1/2} \gamma^{l+3/2} e^{-\gamma r} r^l L_{n+l}^{2l+1}(2\gamma r) \quad (5.96)$$

and $\gamma = \frac{z}{n} = (2I_p)^{1/2}$. We introduce a parameter $\alpha = 2I_p$, hence $\gamma = \sqrt{\alpha}$.

The $1s$ state is chosen as the eigenstate although for most inert gasses, the p state is the ground state. For the $1s$ state, $n = 1$, $l = 0$, $m = 0$, $Y_{00}(\theta, \phi) = \frac{1}{\sqrt{4\pi}}$, so

$$u_b(r, \theta, \phi) = \frac{\alpha^{3/4}}{\pi^{1/2}} e^{-\sqrt{\alpha} r}. \quad (5.97)$$

The dipole matrix $\vec{d}(\vec{p}) = \langle u | \vec{r} | u_b \rangle$ can be calculated as follows:

$$\vec{d}(\vec{p}) = \frac{i}{\omega} \int d^3 r u^*(\vec{r}) \hat{p} u_b(\vec{r}), \quad (5.98)$$

where

$\omega = \frac{k^2}{2} + I_p$ is the emitted photon energy

$\hat{p}$ is the momentum operator

We thus have

$$\vec{d}(\vec{p}) = i \frac{1}{(2\pi)^{3/2}} \int d^3 r e^{-i\vec{k} \cdot \vec{r}} \hat{p} u_b(\vec{r}). \quad (5.99)$$

Since $\hat{p}$ is a Hermitian operator, it commutes with $e^{-i\vec{k} \cdot \vec{r}}$. The function $e^{-i\vec{k} \cdot \vec{r}}$ is an eigenfunction of $\hat{p}$ with eigenvalue $-\vec{k}$ such that $\hat{p} e^{-i\vec{k} \cdot \vec{r}} = -\vec{k} e^{-i\vec{k} \cdot \vec{r}}$. Therefore, we have

$$\vec{d}(\vec{p}) = i\vec{k} \frac{1}{\sqrt{(2\pi)^3}} \int d^3 r e^{-i\vec{k} \cdot \vec{r}} u_b(\vec{r}) = i\vec{k} \psi_b(\vec{k}), \quad (5.100)$$

where $\psi_b(\vec{k})$ is the Fourier-transform of $u_b(\vec{r})$, i.e., the normalized momentum space wave function for the ground state of b , which is

$$\psi_b(p, \vartheta, \phi) = F_{nl}(p) Y_{lm}(\vartheta, \phi). \quad (5.101)$$

The radial eigenfunction in momentum space is

$$F_{nl}(p) = \frac{2^{2l+5/2} l!}{\sqrt{\pi}} \left(\frac{n(n-l-1)!}{(n+l)!} \right)^{1/2} \gamma^{-3/2} \frac{s^l}{(s^2+1)^{l+2}} C_{n-l-1}^{l+1} \times \left(\frac{s^2-1}{s^2+1} \right). \quad (5.102)$$

where $s = p/\gamma = p/\sqrt{2I_p}$. For the s state, $Y_{00}(\vartheta, \phi) = \frac{1}{\sqrt{4\pi}}$. $C_m^\lambda(x)$ is the Gegenbauer polynomials. The first few of them are

$$\begin{aligned} C_0^\lambda(x) &= 1, \\ C_1^\lambda(x) &= 2\lambda x, \\ C_2^\lambda(x) &= -\lambda + 2\lambda(1+\lambda)x^2, \\ C_3^\lambda(x) &= -2\lambda(1+\lambda)x + \frac{4}{3}\lambda(1+\lambda)(1+\lambda)x^3. \end{aligned} \quad (5.103)$$

If we consider the noble gases as hydrogen-like atoms with different $\gamma = \sqrt{2I_p}$ and n, l values, then the wave functions for them can be obtained. For helium, $n=1, l=0$. For Ne, Ar, Kr, and Xe, $l=1, n=2,3,4,5$ respectively. Of course, for these two or multielectro atoms, this approach is just an approximation. It explains qualitatively the dependence of the dipole transition-matrix element as a function of momentum.

5.1.6 Coulomb Corrections

5.1.6.1 Correction to the Recombination Term

The high harmonic and attosecond spectrum can be more precisely calculated through the dipole acceleration, instead of the dipole moment. According to the Ehrenfest theorem, the acceleration can be expressed as

$$\frac{d^2\vec{r}}{dt^2} = \frac{d}{dt} \left\langle \frac{d\vec{r}}{dt} \right\rangle = -\langle \Psi | \nabla V(\vec{r}) | \Psi \rangle + \vec{\varepsilon}_L(t) \cdot \vec{r}(t), \quad (5.104)$$

which is essentially Newton's second law. The effects the Coulomb potential is included by the term $\nabla V(\vec{r})$.

When the laser field $\varepsilon_L(t)$ is pulsed, the wave function that takes into account the depletion of ground-state population is

$$|\Psi(r, t)\rangle = a(t)|0\rangle e^{iI_p t} + \int d^3\vec{v} b(\vec{v}, t) |\vec{v}\rangle e^{i\omega_e t}. \quad (5.105)$$

Inserting the wave function into Equation 5.104 and keeping the dominating terms, we get

$$\frac{d^2\vec{r}}{dt^2} \approx \int d^3\vec{v} a^*(t) b(\vec{v}, t) \langle 0 | \nabla V(\vec{r}) | \vec{v} \rangle + c.c. \quad (5.106)$$

We already know that

$$b(\vec{p}, t) \approx i \int_0^t dt' \vec{\varepsilon}_L(t') \cdot \vec{d} [\vec{p} - \vec{A}(t')] a(t') e^{iS(\vec{p}, t, t')}. \quad (5.107)$$

Thus,

$$\begin{aligned} \frac{d^2\vec{r}}{dt^2} = i \int_0^t dt' \int d^3\vec{p} a^*(t) \langle 0 | \nabla V(\vec{r}) | \vec{p} - \vec{A}_L(t) \rangle e^{iS(\vec{p}, t, t')} \vec{\varepsilon}_L(t') \\ \cdot \vec{d} [\vec{p} - \vec{A}_L(t')] + c.c. \end{aligned} \quad (5.108)$$

Finally applying saddle point approximation, we have

$$\begin{aligned} \frac{d^2\vec{r}}{dt^2} = i \int_0^\tau d\tau \left(\frac{\pi}{\epsilon + \frac{i\tau}{2}} \right)^{\frac{3}{2}} a^*(t) \langle 0 | \nabla V(\vec{r}) | \vec{p}_s(t, \tau) - \vec{A}_L(t) \rangle \\ \times e^{iS(\vec{p}_s, t, \tau)} \vec{\varepsilon}_L(t - \tau) \cdot \vec{d} [\vec{p}_s - \vec{A}_L(t - \tau)] a(t - \tau) + c.c. \end{aligned} \quad (5.109)$$

It can be seen that the correction is for the recombination step.

5.1.6.2 Correction to the Ionization Step

When the laser field is linearly polarized along the x direction, the acceleration

$$\frac{d^2x}{dt^2} = i \int_0^\infty d\tau \left(\frac{\pi}{\epsilon + \frac{i\tau}{2}} \right)^{\frac{3}{2}} a^*(t) \left\langle 0 \left| \frac{\partial}{\partial x} V(\vec{r}) \right| p_{sx}(t, \tau) - A_x(t) \right\rangle \\ \times e^{iS(\vec{p}_s, t, \tau)} \mathcal{E}_L(t - \tau) \cdot d_x [p_{sx} - A_x(t - \tau)] a(t - \tau) + c.c. \quad (5.110)$$

If the ground state is $1s$, it can be shown that term $\mathcal{E}_L(t - \tau) \cdot \vec{d}_x [p_{sx}(t, \tau) - A_x(t - \tau)]$ in Equation 5.110 for calculating the rate that electron is freed to the continuum state can be replaced by

$$-\frac{(2I_p)^{1/4}}{|\mathcal{E}_L(t - \tau)|} \sqrt{\frac{w(t - \tau)}{\pi}}$$

where $w(t - \tau)$ is the ADK or PPT ionization rate. Consequently Gordon and Kärtner obtained the expression

$$\frac{d^2x}{dt^2} = -i\pi(2I_p)^{\frac{1}{4}} \int_0^\infty d\tau \left(\frac{1}{\epsilon + \frac{i\tau}{2}} \right)^{\frac{3}{2}} \\ \times a^*(t) \left\langle 0 \left| \frac{\partial}{\partial x} V(\vec{r}) \right| p_{sx}(t, \tau) - A_x(t) \right\rangle \\ \times e^{iS(p_{sx}, t, \tau)} a(t - \tau) \frac{\sqrt{w(t - \tau)}}{|\mathcal{E}_L(t - \tau)|} + c.c. \quad (5.111)$$

The results calculated using expression 5.111 closer to that from solving the Schrödinger equation than Equation 5.109.

5.1.6.3 Matrix Element

The values of $\left\langle 0 \left| \frac{\partial}{\partial x} V(\vec{r}) \right| \vec{v} \right\rangle$ for noble gas atoms have been provided by Gordon et al., which was obtained by solving the Schrödinger equation for multielectron atoms. The values are shown in [Figure 5.6](#).

It is possible to find analytical fitting functions for $\left\langle 0 \left| \frac{\partial}{\partial x} V(\vec{r}) \right| \vec{v} \right\rangle$. As an example, for helium atom, the function

$$f(v) = 0.26\alpha^{\frac{5}{4}} \frac{v}{\left(v^2 + \frac{3}{2}\alpha\right)^{\frac{3}{4}}} \quad (5.112)$$

is a good fitting function for the result shown in [Figure 5.7](#). $\alpha = 1.807$ atomic unit for helium, which decreases slower with v than that from the dipole matrix element neglecting the Coulomb field, which is $\frac{2^{3/2}}{\pi} \alpha^{\frac{5}{4}} \frac{v^5}{(v^2 + \alpha)^3}$ for $v^2 \gg \alpha$.

Lin's group calculated the dipole transition-matrix elements for noble gases using the scattering wave instead of plane wave. The comparison between the photoabsorption cross section which is related to the dipole transition-matrix elements is shown in [Figure 5.8](#).

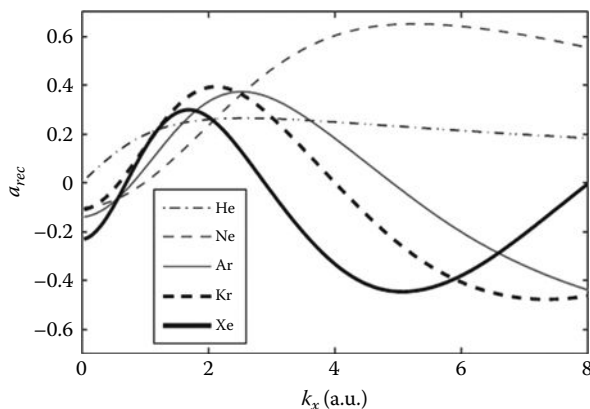


Figure 5.6 Calculated $a_{rec} = \left\langle 0 \left| \frac{\partial}{\partial x} V(\vec{r}) \right| \vec{v} \right\rangle$. (Reprinted with permission from A. Gordon, F. X. Kärtner, N. Rohringer, and R. Santra, *Phys. Rev. Lett.*, 96, 223902, 2006. Copyright 2006 by the American Physical Society.)

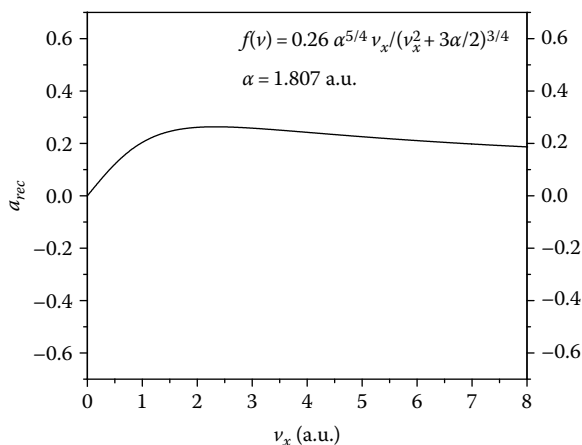


Figure 5.7 Fitting function for the calculated $a_{rec} = \left\langle 0 \left| \frac{\partial}{\partial x} V(\vec{r}) \right| \vec{v} \right\rangle$ of helium.

5.2 Temporal Phase of Harmonic Pulses*

The SFA has been used to explain the experiment's observation that the high harmonic spectrum is susceptible to the temporal phase of the driving laser pulses, which can be understood by examining the temporal phase of the high harmonic field.

The amplitude and phase of a high harmonic spectrum corresponding to a given set of laser parameters can be obtained by performing a Fourier transform of the expression 5.77. By doing so, it is found that the

* This section is adapted from Chang, Z., A. Rundquist, H. Wang, I. Christov, H.C. Kapteyn, and M.M. Murnane, Temporal phase control of soft-x-ray harmonics emission, *Phys. Rev. A* 58, R30 (1998).

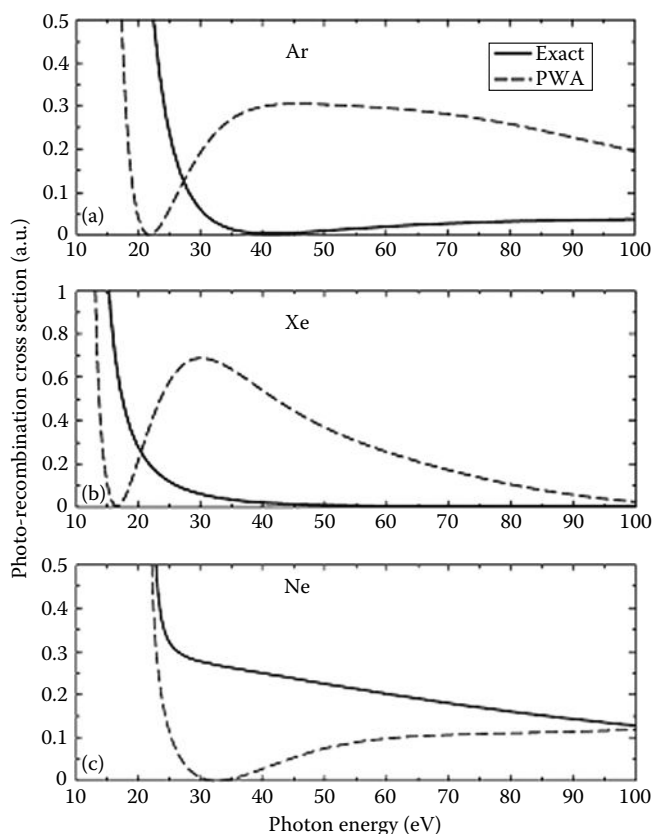


Figure 5.8 Photorecombination cross sections of Ar, Xe, and Ne, obtained by using exact scattering wave functions (solid curves) and within the plane-wave approximation (dashed curves) for the continuum electrons. (Reprinted with permission from A.-T. Le et al., *Phys. Rev. A*, 78, 623814, 2008. Copyright 2008 by the American Physical Society.)

spectrum of a given order q has certain width $\Delta\omega_q$. If that order is filtered out, it would be a femtosecond XUV pulse. Furthermore, SFA predicted that the temporal phase of the XUV pulse depends strongly on the temporal phase of the driving laser.

5.2.1 Intrinsic Dipole Phase

For simplicity, we assume that the driving laser is monochromatic. We also neglect the depletion of the ground state population. Under these two assumptions, each high harmonic is a monochromatic XUV light.

In one laser cycle, the phase of the dipole moment responsible for generating the radiation contributing to the single frequency q th harmonic is determined by the value of the action acquired along the most relevant saddle point trajectory, i.e.,

$$\Phi_{dipole,q} = S(p_{sx}, t, \tau), \quad (5.113)$$

where

$$S(p_{sx}, t, \tau) = I_p \tau + U_p \left\{ \tau - \frac{2[1 - \cos(\tau)]}{\tau} - \left[\sin(\tau) - \frac{4 \sin^2(\tau/2)}{\tau} \right] \cos(2t - \tau) \right\}. \quad (5.114)$$

For the discussion in this section, we are only interested in harmonic orders close to the cutoff, which are generated, from the classical point of view, by an electron that is released at a time of $t - \tau = 0.3$ rad, and that returns at a time of $t = 4.37$ rad, i.e., $\tau = 4.07$ rad. For the cutoff harmonics, expression 5.114 gives

$$S = 4.07 I_p + 3.2 U_p. \quad (5.115)$$

Since U_p is proportional to the laser intensity, it is clear that the phase of dipole is intensity dependent. Therefore, for short pulse excitation where the intensity changes rapidly with time, the dipole phase is also time dependent. The first term in Equation 5.115 does not depend on laser intensity, which is ignored in the discussion here.

In SI unit

$$\Phi_{dipole,q}(U_p) = 3.2 \frac{U_p}{\hbar \omega_0}. \quad (5.116)$$

The discovery of the intensity-dependent dipole phase is one of the triumphs of the SFA. Such a phase plays a similar role as the nonlinear phase caused by the nonlinear index of refraction in conventional nonlinear optics. It is worthy pointing out that the dipole phase does not depend on laser intensity in perturbative low order harmonic generation, such as second harmonic generation in a BBO crystal.

5.2.2 Gaussian Analysis of the Temporal Phase

Now, we consider the case where the driving laser is pulses. For simplicity, we use the Gaussian theory introduced in Chapters 1 and 2 to discuss the temporal phase of the XUV pulse centered at ω_q .

5.2.2.1 Laser Pulses

The driving laser field at given time t is expressed as

$$\varepsilon_L(t) = E_1 e^{-\Gamma_1 t^2} e^{i\omega_0 t}, \quad (5.117)$$

where

E_1 is the peak field

ω_0 is the carrier frequency

The chirp of the pulse b_1 is included in the complex Gaussian parameter

$$\Gamma_1 = a_1 - ib_1, \quad (5.118)$$

where

$$a_1 = 2 \ln(2) \frac{1}{\tau_1^2}, \quad (5.119)$$

and τ_1 is the full width at half maximum of the pulse.

For a given spectrum bandwidth, $b_1 = 0$ corresponds to a transform-limited pulse, which has the minimum pulse duration. The chirp of the laser can be tuned by using a phase modulator, or by changing the spacing of the grating in the pulse stretcher or compressor in a chirped pulse amplifier system. Unfortunately, the laser pulse duration is changed at the same time. When the grating separation in the stretcher is increased, it introduces a net positive chirp ($b_1 > 0$) on the pulse, and increases the pulse duration. Decreasing the grating separation also broadens the pulse, but introduces a net negative chirp ($b_1 < 0$). The observed effects on the high harmonic spectrum come from both the laser chirp and pulse width.

5.2.2.2 High Harmonic Pulses

The electric field of the q th harmonic is given by

$$\varepsilon_q(t) = E_q e^{-a_q t^2} e^{i[q\omega_0 t + \Phi_{total}(t)]}, \quad (5.120)$$

where E_q is the peak field, and

$$a_q = 2 \ln(2) \frac{1}{\tau_q^2}. \quad (5.121)$$

τ_q is the full width at half maximum of the harmonic pulse. $\Phi_{total}(t)$ is the total temporal phase, which can be written as the sum of two contributions

$$\Phi_{total,q}(t) = \Phi_{dipole,q}[U_p(t)] + \Phi_{laser,q}(t), \quad (5.122)$$

where $\Phi_{dipole,q}[U_p(t)]$ is the phase of the induced dipole moment, which is laser intensity dependent, and

$$\Phi_{laser,q}(t) = qb_1 t^2 \quad (5.123)$$

is the phase introduced by the chirp of the laser pulses.

At the peak of the driving laser pulse where the harmonics near cutoff are generated, the NIR pulse shape can be approximated by a parabola,

$$U_p(t) = U_{p0} e^{-2a_1 t^2} \approx U_{p0} (1 - 2a_1 t^2), \quad (5.124)$$

where U_{p0} is the ponderomotive potential at the peak of the pulse. Using Equations 5.116 and 5.124, we have

$$\Phi_{dipole,q}(t) = 3.2 \frac{U_{p0}}{\hbar\omega_0} (1 - 2a_1 t^2). \quad (5.125)$$

Such a phase corresponds to a negative chirp because of the negative sign in front of the parabolic term. This induced negative chirp can be explained by the harmonic generation process near the peak of a short laser pulse, which causes the emission from subsequent cycles on the pulse leading edge to be earlier in phase from cycle to cycle. As we know, for intense femtosecond laser propagating through a glass slab, the change of the index refraction by the laser field leads to self-phase modulation. The dipole phase in high harmonic generation is another type of self-phase modulation. Such a self-phase modulation can affect the chirp and spectrum of the high harmonic pulse, like in the femtosecond laser case.

It is interesting to note that since the cutoff harmonic order q is given by $(I_p + 3.2U_{p0})/\hbar\omega_0$, for very high orders we can approximate $3.2 \frac{U_{p0}}{\hbar\omega_0}$ by q . After omitting the DC phase shift, we then obtain

$$\Phi_{total,q}(t) = q(-2a_1 + b_1)t^2, \quad (5.126)$$

By combining Equation 5.126 with Equation 5.120, we can thus describe the harmonic field by

$$\varepsilon_q(t) = E_q e^{-\Gamma_1 t^2} q e^{iq\omega_0 t}, \quad (5.127)$$

where

$$\Gamma_q = a_q - ib_q \quad (5.128)$$

is the complex Gaussian parameter, and

$$b_q = q(-2a_1 + b_1) \quad (5.129)$$

is the chirp parameter of the q th harmonic pulse. One should keep in mind that the chirp discussed here is different from the attosecond pulse chirp discussed in Chapter 4.

The effect of the laser chirp on the chirp of the harmonic pulse can be seen in Figure 5.9, which plots the predictions of Equation 5.129. When the driving laser pulse is transform-limited ($b_1 = 0$), the harmonics in the cutoff region are negatively chirped ($b_q < 0$), since $a_1 > 0$. This is a very interesting discovery.

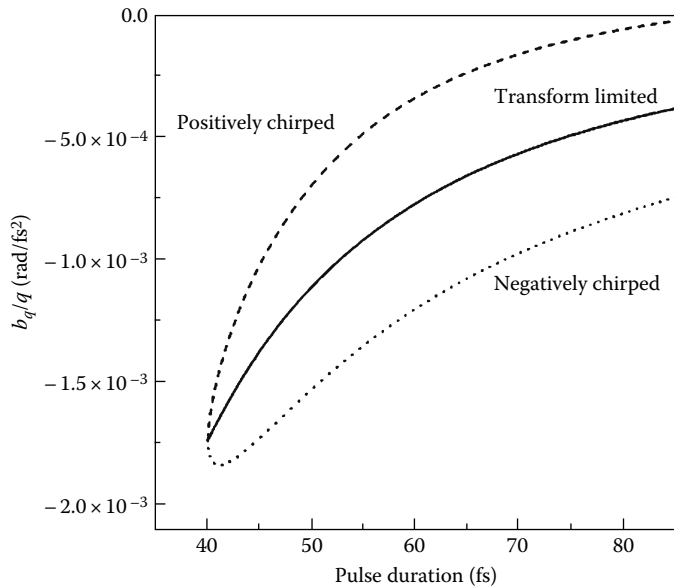


Figure 5.9 Normalized chirp parameter (b_q/q) of the emitted harmonic pulse as a function of laser-pulse duration. The sign of the chirp of the laser pulse is indicated from each curve. (From Z. Chang, A. Rundquist, H. Wang, I. Christov, H.C. Kapteyn, and M.H. Murnane, *Phys. Rev. A*, 58, R30, 1998. Copyright 1998 by the American Physical Society.)

When the laser pulse is negatively chirped ($b_1 < 0$), then the negative chirp of the harmonic pulse is enhanced. On the other hand, if the laser pulse is positively chirped ($b_1 > 0$), it will compensate for the chirp induced on the harmonic field by the dipole phase to some extent, depending on the relative values of b_1 and a_1 . Thus, the chirp of the harmonic pulse can be controlled by the driving laser pulse.

5.2.2.3 High Harmonic Spectrum

The FWHM spectral width of the q th order harmonic is given by

$$\Delta\omega_q = \sqrt{8 \ln(2) a_q \left[1 + \left(\frac{b_q}{a_q} \right)^2 \right]}. \quad (5.130)$$

The induced chirp, b_q , on the harmonic pulse will broaden the bandwidth by a factor of $\left[1 + \left(\frac{b_q}{a_q} \right)^2 \right]$ compared to the transform-limited harmonic case for an identical XUV pulse width.

For two driving lasers pulses with the same pulse duration but opposite chirp, the XUV pulse duration should be the same. In this case, the harmonic chirp, $|b_q|$, induced by a negatively chirped laser pulse is much larger than for a positively chirped pulse, as shown in Figure 5.9. If the harmonic pulse duration parameter a_q is the same, then corresponding spectral width $\Delta\omega_q$ is much larger for negatively chirped NIR pulses than the positively chirped pulses. When $\Delta\omega_q$ approaches $2\hbar\omega_0$, the harmonic peaks will not be well resolved. However, such a continuous spectrum should not be confused with the one corresponding to an isolated attosecond pulse.

5.2.3 Experimental Results

5.2.3.1 Using 40 fs Lasers

The chirp of the harmonic pulse is much more difficult to measure than the spectrum. The effects of driving laser chirp on the high harmonic-spectrum shape were examined experimentally. The effects are easier to observe for the higher orders before the contribution for the driving laser to the total temporal phase is proportional to the harmonic order q . For this reason, harmonics in the 5–6 nm range ($q = 800 \text{ nm}/6 \text{ nm} \approx 130$) generated from helium atom were studied.

To generate such high-order harmonics, a 10 mJ Ti:Sapphire chirped pulse amplifier system that generates near-transform-limited 40 fs laser pulses was used. The laser beam was focused on a helium gas jet with a $100 \mu\text{m}$ diameter spot. The peak intensity of laser at the focus is $1.8 \times 10^{15} \text{ W/cm}^2$. The gas nozzle diameter was 1 mm, while the gas pressure was approximately 8 torr. The x-rays were dispersed using a flat-field soft x-ray spectrometer.

The chirp of the driving laser pulse was varied by adjusting the separation of the gratings in the pulse stretcher of the chirped pulse amplifier. The resultant high harmonic spectra near the cutoff were

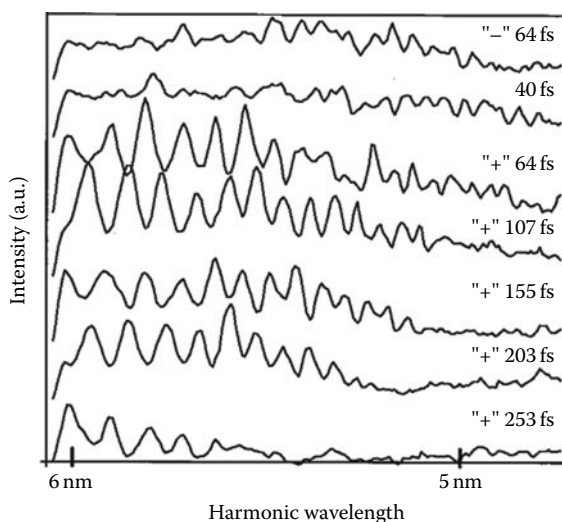


Figure 5.10 Experimentally measured harmonic spectra for positively (+ sign) and negatively (– sign) chirped laser-excitation pulses. (From Z. Chang, A. Rundquist, H. Wang, I. Christov, H.C. Kapteyn, and M.H. Murnane, *Phys. Rev. A*, 58, R30, 1998. Copyright 1998 by the American Physical Society.)

observed to change dramatically as a function of laser chirp, as shown in Figure 5.10. The harmonic peaks are observed to broaden dramatically as the pump pulse is negatively chirped, eventually merging into a continuum. However, for positive laser chirp, the peaks become narrower and well resolved, which is consistent with the theoretical predictions.

5.2.3.2 Numerical Simulation Results

In order to understand the experimentally observed spectra, harmonic generation was simulated using the SFA. Hydrogen-like dipole matrix elements were used to simulate the helium atom. Only the single atom spectra were calculated, whereas the effects of the phase-matching are not included. The results of the simulations for helium using the experimental laser parameters are shown in Figure 5.11, which clearly show that for positively chirped driving laser pulses, the high harmonic peaks are narrow, while for negatively chirped excitation pulses, the peaks are smeared, which qualitatively is in agreement with the experimental data. The results suggest that the experimental observed phenomena are mainly from the single atom response instead of from propagation effects. It also demonstrates the importance of the intrinsic dipole phase in determining the harmonic emission spectra.

5.2.3.3 Few-Cycle Driving Laser

In experiments where the driving laser chirp is changed by tuning the grating separation, the high-order phase also changes, which makes it

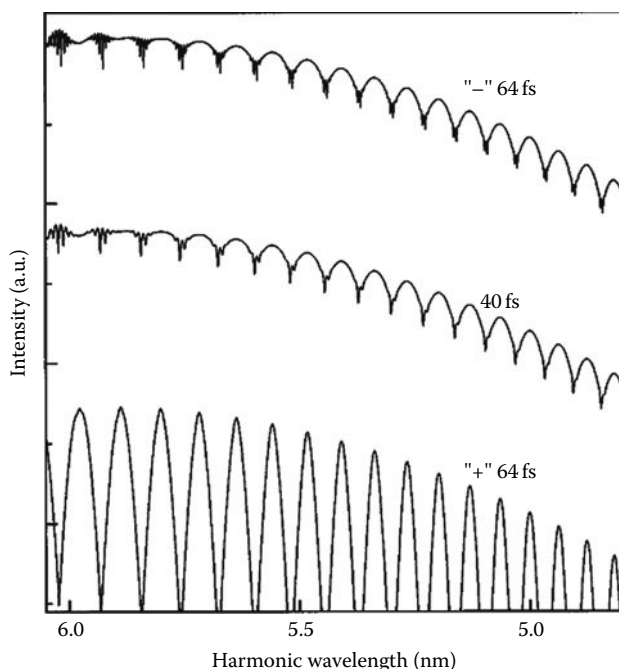


Figure 5.11 Numerical simulation of high harmonic emission spectra near cutoff for different values of chirp of the excitation pulses: positively chirped pulses (+ sign) and negatively chirped pulses (– sign). (From Z. Chang, A. Rundquist, H. Wang, I. Christov, H.C. Kapteyn, and M.H. Murnane, *Phys. Rev. A*, 58, R30, 1998. Copyright 1998 by the American Physical Society.)

difficult to identify the contribution of each phase order. The problem can be solved by using a phase modulator as the one introduced in Chapter 2. Controlling of the high harmonic spectra has also been demonstrated with the 0.55 mJ, 5.2 fs pulses by independently changing the GDD and high-order spectral phases using a phase modulator.

In the experiments, the laser beam centered at 780 nm from the hollow-core fiber followed by the phase modulator was focused to an Argon gas cell with a length of 1.4 mm and a backing pressure of 30 torr. The gas target was placed approximately 2 mm after the focus to optimize the phase-matching for the short trajectory. The spectrum in the 20–37 nm range was measured. The harmonics are in the plateau region.

As seen in Figure 5.12a, when the GDD was increased, the harmonic peaks shift to longer wavelengths, like what was discovered with the 40 fs laser pulses. This is due to the ionization of the target atom in the leading edge of the driving laser. The most interesting feature is the asymmetric dependence on the positive chirp as compared to the negative chirp. The asymmetry is even stronger for third order to fifth order phases as shown in Figure 5.12b through d. The mechanism for the disappearance of the harmonic signal for relatively large negative chirp is yet to be clarified.

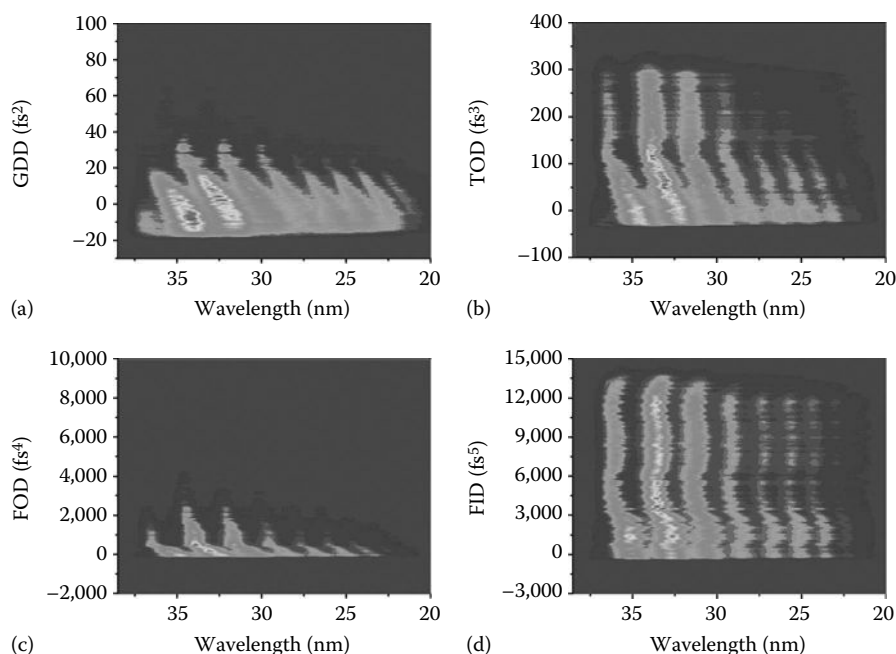


Figure 5.12 Dependence of high-order harmonic spectra on the high-order phases of the driving laser pulses. (Reprinted H. Wang, Y. Wu, C. Li, H. Mashiko, S. Gilbertson, and Z. Chang, Generation of 0.5 mJ, few-cycle laser pulses by an adaptive phase modulator, *Opt. Express*, 16, 14448, 2008. With permission of Optical Society of America.)

5.3 Effects of Molecular Orbital Symmetry*

As discussed in Chapter 4, the yield of high harmonic generation from atoms depends strongly on the ellipticity of the driving lasers. Under an elliptically polarized laser field, the re-collision electron is driven away by the transverse field component from its parent ion so that the XUV flux drops quickly with the driving field ellipticity. This property is used in polarization gating to produce single isolated attosecond pulses. For the polarization gating to work effectively, one prefers to use target that shows stronger ellipticity dependence. Therefore, it is interesting to know whether high harmonic generation from some molecules is more susceptible to ellipticity than atoms. Motivated by such applications, the dependence of high-order harmonic generation yield on the ellipticity of the driving laser field for O_2 and N_2 molecules was compared with Ar atom experimentally in the author's lab. With the help of the SFA for molecules, the different behaviors of these targets have been explained by the orbital symmetries of the valence electrons.

* This section is adapted from Shan, B., S. Ghimire, and Z. Chang, Effect of orbital symmetry on high-order harmonic generation from molecules, *Phys. Rev. A* 69, 021404(R) (2004).

5.3.1 Experimental Results

5.3.1.1 Ellipticity Control

The experiments were carried out using a Ti:Sapphire chirped pulse amplification laser running at a repetition rate of 1 kHz. It produces 25 fs pulses with 4 mJ energy at a center wavelength of 790 nm. The linearly polarized laser field is converted to the elliptically polarized with controllable ellipticity before focusing on the gas target. The gas nozzle has an outlet diameter of 75 μm and the gas density in the interaction region was estimated to be $\sim 5 \times 10^{17}/\text{cm}^3$. High harmonic generation in four types of gases, Ar, Xe, O₂, and N₂, was studied, the molecules were randomly oriented.

The XUV beam from the gas target is sent to a transmission grating XUV spectrometer described in Chapter 4, where the spectral image was enhanced by an 80 mm diameter MCP intensifier and recorded on a CCD camera. The large MCP allows the measurement of the XUV spectrum over a broad range. The signal level is rather low when the ellipticity is large. A cooled CCD is used to reduce the background noise of the detector, which allowed accumulation of the imager over a long period of time.

It is well known that the diffraction efficiency of transmission gratings strongly depends on whether the XUV polarization direction is parallel or perpendicular to the lines of the grating. The efficiency is typically higher when the two are orthogonal. For high harmonic generation with elliptically polarized laser, we expect that the electric field of the XUV is also elliptically polarized. The major axis of the XUV ellipse is in the same direction as the NIR ellipse. Thus, to minimize the grating efficiency variation, the orientation of the major axis of the XUV or NIR should not change when the ellipticity is varied.

The ellipticity of the NIR laser was varied by a combination of a rotatable half-wave plate and fixed quarter-wave plate, as illustrated in Figure 5.13. The optic axis of the quarter-wave plate was set along the dispersion direction of the transmission grating. By rotating the half-wave plate, the ratio of the two field components, one is parallel and the other is perpendicular, to the optic axis of the quarter-wave plate is changed.

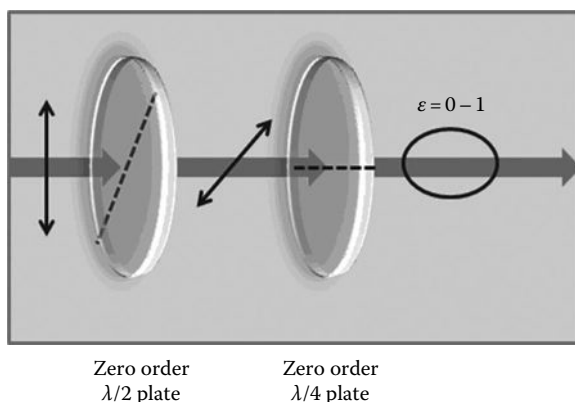


Figure 5.13 Optics setup to vary the ellipticity of the laser without changing the orientation of the polarization ellipse.

In this way, the ellipticity of the laser beam after the two wave-plates was varied from 0 (linearly polarized along the gating dispersion direction) to 1 (circularly polarized) whereas the major axis of the polarization ellipse does not change. With such a configuration, the effect of the polarization dependent diffraction efficiency of the grating on the measured XUV results is minimized.

5.3.1.2 High Harmonic Cutoff

For generating the shorter attosecond pulse, we would like to know whether XUV spectrum can be extended to a shorter wavelength when molecules are used as the generation gas. The dependence of the harmonic cutoff photon energy with the driving laser intensity for molecules was compared with that for atoms using linearly polarized laser. The laser power is changed by rotating a half-wave plate in front of a polarizer before the grating compressor of the CPA laser so that the pulse duration is kept at a constant value. The cutoff of the harmonic spectrum is defined as the highest detectable harmonic order.

The measured relationships between cutoff energy and laser intensity are shown in Figure 5.14. When the laser intensity reaches the ionization saturation intensity, all the ground-state population is depleted. Above the saturation intensity, the cutoff order does not increase any more. The highest cutoff harmonic order of the N_2 is almost the same as that of Ar. However, the maximum cutoff from O_2 is much higher than that from xenon even though the two have comparable ionization potentials. This can be explained by the ionization suppression of O_2 because its highest occupied molecular orbital (HOMO) is an antibonding type, as pointed out by Guo and Gibson.

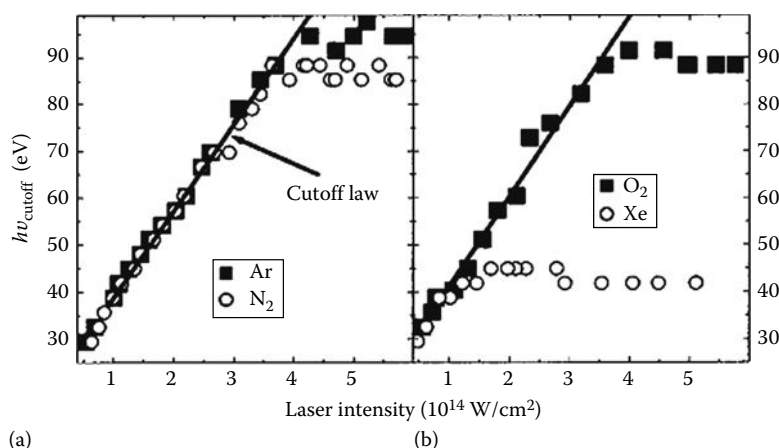


Figure 5.14 Measured high harmonic cutoff with respect to the laser intensity for two pairs of atomic and molecular gases with similar ionization potential and different orbital symmetry. (a) Ar vs. N_2 ; (b) Xe vs. O_2 . (From B. Shan, S. Ghimire, and Z. Chang, *Phys. Rev. A*, 69, 021404(R), 2004. Copyright 2004 by the American Physical Society.)

5.3.1.3 Ellipticity Dependence

Figure 5.15a shows the dependence of 21st order harmonic signal on the ellipticity of the laser for Ar and N₂ gasses. They both drop quickly with ellipticity. The signal from Ar gas was normalized to that from N₂ for linear polarization. It can be seen that the dependence on ellipticity for these two gases is almost identical.

In this measurement, the on-target laser intensity was $\sim 2.3 \times 10^{14}$ W/cm². The laser intensity was kept constant when the ellipticity was varied. The laser field decreases with the ellipticity. Since the tunneling ionization rate depends strongly on the field strength. The observed drop of high harmonic signal with ellipticity is the result of decrease of both the ionization rate in the first step and the recombination probability in the last step of the semiclassical model.

Figure 5.15b shows the 45th order harmonic signal dependence on the laser ellipticity for O₂, N₂, and Ar gasses, which is more interesting than the 21st order results. The estimated laser intensity on the target was $\sim 3.5 \times 10^{14}$ W/cm². For this order, the XUV signal from O₂ gas drops slower than N₂ gas, whereas that of Ar gas is in between. It indicates that N₂ is a better choice for generating isolated attosecond pulses with polarization gating than Ar or O₂. The question is why they have ellipticity dependence difference.

5.3.2 Numerical Simulations

To understand the ellipticity dependence difference for N₂ and O₂, The SFA for atoms was extended to simulate the harmonic generation from molecules. We start with the dipole moment of an atom or molecule in the time domain,

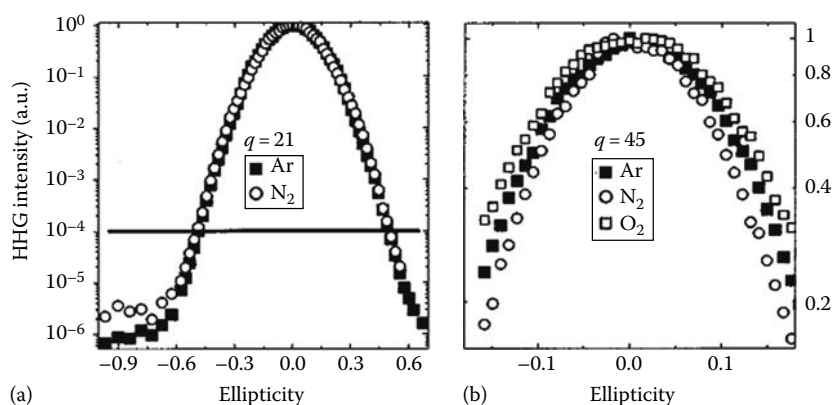


Figure 5.15 (a) Measured ellipticity dependence of 21st harmonics of Ar and N₂ gases. The laser intensity is 2.3×10^{14} W/cm². The region above the horizontal line has a signal/noise ratio, $(I_{\text{signal}} - I_{\text{background}})/(I_{\text{signal}} + I_{\text{background}})$, better than 30%. (b) Measured ellipticity dependence for the 45th order harmonic from N₂, O₂, and Ar gases. The laser intensity is 3.5×10^{14} W/cm². (From B. Shan, S. Ghimire, and Z. Chang, *Phys. Rev. A*, 69, 021404 (R), 2004. Copyright 2004 by the American Physical Society.)

$$\vec{r}(t) = i \int_0^\infty d\tau \left(\frac{\pi}{\epsilon + i\tau/2} \right)^{3/2} \cdot \vec{d}^* [\vec{p}_s(t, \tau) - \vec{A}_L(t)] e^{is(\vec{p}_s, t, \tau)} \vec{e}_L(t - \tau) \\ \cdot \vec{d} [\vec{p}_s(t, \tau) - \vec{A}_L(t - \tau)] + c.c. \quad (5.131)$$

The difference between the atoms and molecules originates from their dipole transition matrix elements.

For both atoms and molecules, the field-free dipole transition matrix element between the ground-state and the plane-wave continuum state is

$$\vec{d}(\vec{p}) = i \frac{2\vec{p}}{(p^2 + \alpha)} \tilde{\phi}(\vec{p}), \quad (5.132)$$

where $\alpha = 2I_p$ and $\tilde{\phi}(\vec{p})$ is the momentum space wave function for the ground state of the atom or molecules.

For the $1s$ state of an atom,

$$\vec{d}_{1s}(\vec{p}) = i \frac{2^{7/2}}{\pi} \alpha^{5/4} \frac{\vec{p}}{(\vec{p}^2 + \alpha)^3}. \quad (5.133)$$

5.3.2.1 Bonding Orbital and Antibonding Orbital

When the dipole transition matrix elements for molecules are also calculated using Equation 5.132, the ground states of molecules should be the highest occupied molecular orbitals (HOMOs) where the electrons are ionized by the intense laser field. The wave functions of the HOMOs for N_2 and O_2 are presented in Figure 5.16. The green and red colors indicate the phase of wave function in a given spatial point. The parity of N_2 is even, which is named bonding orbital. On the contrary, the parity of O_2 is odd. It is an antibonding orbital. The wave functions of the $3p$ state of Ar and the $5p$ state of Xe are also presented for comparisons. They are the outmost shells.

Instead of finding out the exact dipole transition matrix elements of N_2 and O_2 , two model diatomic, homonuclear molecules are used instead for simplicity. One has a bonding orbital and the other has an antibonding orbital to represent these two real molecules, respectively. The wave functions of the molecular orbitals are described as linear combinations of $1s$ atomic orbitals (LCAO). The $1s$ state is chosen also for simplicity.

In the configuration space, the LCAO molecular wave functions for bonding and antibonding orbitals are

$$\psi_b(\vec{r}) = \beta [\phi_{1s}(\vec{r} - \vec{R}_1) + \phi_{1s}(\vec{r} - \vec{R}_2)], \quad (5.134)$$

$$\psi_a(\vec{r}) = \gamma [\phi_{1s}(\vec{r} - \vec{R}_1) - \phi_{1s}(\vec{r} - \vec{R}_2)], \quad (5.135)$$

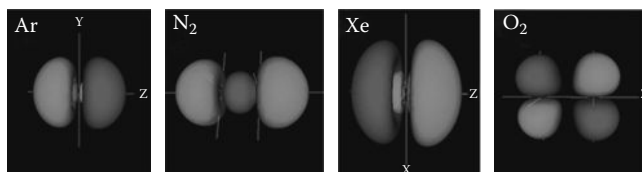


Figure 5.16 Wave functions of the electron in the outermost shells.

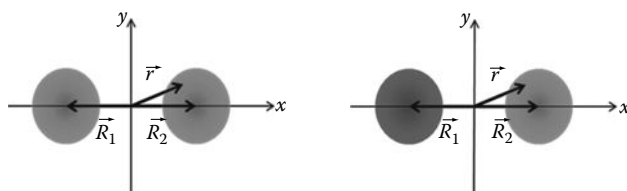


Figure 5.17 Wave functions of the model molecules.

respectively, where β and γ are factors from normalization of the wave functions. $\vec{R}_1, \vec{R}_2 = \vec{R}_1 + \vec{R}$ are the positions of the two nuclei, and $|\vec{R}|$ is the equilibrium internuclear separation. The wave functions are sketched in Figure 5.17. The plus sign in 5.134 indicates that the two atomic wave functions are in phase. The minus sign in 5.135 means that they are out of phase.

The wave function of the $1s$ atomic orbital that forms the molecules is

$$\phi_{1s}(\vec{r}) = \frac{1}{\pi^{1/2} r_0^{3/2}} e^{-r/r_0}, \quad (5.136)$$

where $r_0 = \frac{1}{\sqrt{\alpha}} = \frac{1}{\sqrt{2I_p}}$ is the size of the atom. It is assumed that $I_p = 15.8$ eV. The value is chosen so that the cutoff harmonic orders from the simulations are comparable to the measured ones. The corresponding atom size is $0.5 \text{ \AA}$. The internuclear distance is taken as $R = 2r_0$, which is $1 \text{ \AA}$. The internuclear distance is close to the values of N_2 ($1.098 \text{ \AA}$) and O_2 ($1.208 \text{ \AA}$).

In the momentum space, the wave functions of the molecular orbitals for the bonding and antibonding model molecules corresponding to $\psi_b(\vec{r})$ and $\psi_a(\vec{r})$ are

$$\tilde{\psi}_b(\vec{p}) = 2\beta \tilde{\phi}_{1s}(\vec{p}) \cos(\vec{p} \cdot \vec{R}/2), \quad (5.137)$$

$$\tilde{\psi}_a(\vec{p}) = i2\gamma \tilde{\phi}_{1s}(\vec{p}) \sin(\vec{p} \cdot \vec{R}/2), \quad (5.138)$$

where $\tilde{\phi}_{1s}(\vec{p})$ is the atomic $1s$ wave function in momentum space, which is the Fourier transform of Equation 5.127. The trigonometry function terms are due to the interference between the two atomic wave functions in configuration space. We see that the probability of finding electrons moving perpendicularly to the molecular axis direction ($\vec{p} \cdot \vec{R} = 0$) is maximized for the bonding orbital, whereas it is zero for the antibonding orbital.

The matrix elements for the dipole transition between the ground-state and the plane-wave continuum states $\vec{p}$ are

$$\vec{d}_b(\vec{p}) = i2\beta \vec{d}_{1s}(\vec{p}) \cos(\vec{p} \cdot \vec{R}/2), \quad (5.139)$$

$$\vec{d}_a(\vec{p}) = 2\gamma \vec{d}_{1s}(\vec{p}) \sin(\vec{p} \cdot \vec{R}/2), \quad (5.140)$$

for the bonding and antibonding molecules, respectively. $\vec{d}_{1s}(\vec{p})$ is given by Equation 5.133. Equations 5.139 and 5.140 reveal the origin of the difference between high harmonic generation from molecules and that from atoms. For $\vec{p} \perp \vec{R}$, the transition probability is the highest for the bonding orbital because the contributions from the two atoms add up constructively (the two atomic wave functions are in phase). On the contrary, the prob-

ability for the antibonding orbitals is zero due to the total destructive interference (the two atomic wave functions are out of phase).

5.3.2.2 Simulation Results

The simulations of the high harmonic spectra from molecules were done by inserting Equation 5.139 or 5.140 into Equation 5.131. The field-free dipole transition matrix elements of molecules depend on the orientation between the molecular axis and the direction of the freed electrons. In the strong laser field, the direction of free electron motion is almost the same as the laser field direction, thus, the dipole moment calculated with Equation 5.131 also depends on the angle between the molecular axis and the major axis of the ellipse of the NIR laser field, Θ .

The simulation results for $\Theta = 30^\circ, 50^\circ, 70^\circ, 90^\circ$ are shown in Figure 5.18 for the 45th harmonic order (~ 70 eV). For the 50° angle, the difference of ellipticity dependence between the two model molecules is rather small. For angles larger than 70° , the antibonding molecules show significantly slower decrease for the calculated ellipticity range. The largest difference occurs at 90° , where the antibonding signal drops to zero. For isolated attosecond polarization gating, bonding molecules oriented at 90° are preferred as the decrease of the harmonic signal with the ellipticity is the fastest.

5.3.2.3 Role of Interference

The difference in ellipticity dependence between an antibonding molecule and a bonding molecule oriented at $\Theta = 90^\circ$ can be explained with the semiclassical theory. For an antibonding molecule with its axis-oriented perpendicular to the electric field of linearly polarized laser (ellipticity is

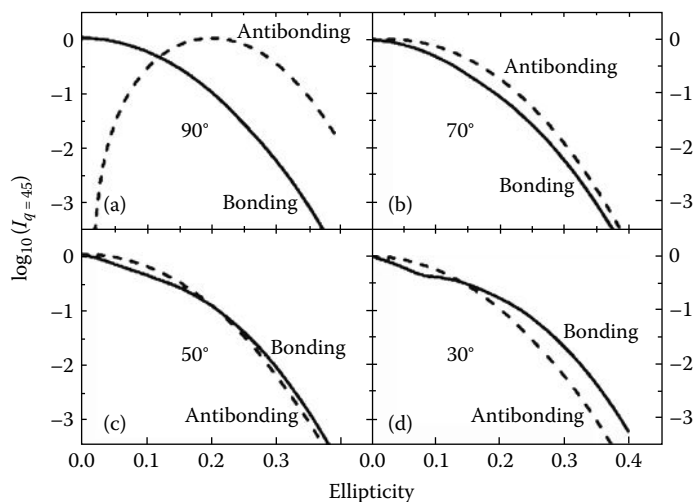


Figure 5.18 Calculated ellipticity dependence for the 45th order harmonic from bonding and antibonding molecules with different orientations. Θ is defined as the angle between the molecular axis and the major axis of the ellipse of the electric field. (a) $\Theta = 90^\circ$; (b) $\Theta = 70^\circ$; (c) $\Theta = 50^\circ$; (d) $\Theta = 30^\circ$. (From B. Shan, S. Ghimire, and Z. Chang, *Phys. Rev. A*, 69, 021404(R), 2004. Copyright 2004 by the American Physical Society.)

zero), the electron tunnels out with a certain initial transverse velocity due to the sine term of the wave function expressed by 5.138, i.e., $\sin(\vec{p} \cdot \vec{R}/2) = 0$ for $\vec{p} \perp \vec{R}$. In this case, the electron will drift away transversely from the parent molecular ions. This results in a very low XUV signal due to the small recombination probability for the re-collision process.

With an appropriate amount of ellipticity (~ 0.2 for the 45th harmonic order), the vertical component of the electric field compensates the effect of the transverse initial velocity. The elliptically polarized laser field drives the drifting electron right back to the parent ion, thus enhancing the recombination probability. Consequently, in the 0–0.2 ellipticity range, the XUV signal increases with the ellipticity for the antibonding molecule. If the ellipticity is too large, the returning electron is driven too far by the transverse component of the laser field, the high harmonic signals decreases with the ellipticity.

For a bonding molecule with its axis-oriented perpendicularly to the electric field of a linearly polarized laser field (ellipticity is zero), the initial velocity distribution of the tunneled out electron has the cosine term, as described by 5.139. Since $\cos(\vec{p} \cdot \vec{R}/2) = 1$ for $\vec{p} \perp \vec{R}$, the probability of an electron tunneling out with its initial velocity along the electric field is larger than in any other direction. In this case, the recombination probability is highest for a linearly polarized light field. Adding any amount of transverse field would drive the returning electron away from the parent ion. Consequently, the harmonic signal decreases monotonically with the driving laser ellipticity.

Although molecules can be aligned by another laser pulse, when the data in Figure 5.15 were taken, the molecules in the gas target were randomly oriented. The measured high harmonic signal was the coherent superposition of the radiation from all the molecules in the gas target. To compare with the measured data, Figure 5.19 shows the simulation results for the 45th order that summed up the contributions from molecules with random orientation angles. The ellipticity dependence difference between bonding and antibonding molecules still exists, although not as obvious as the aligned

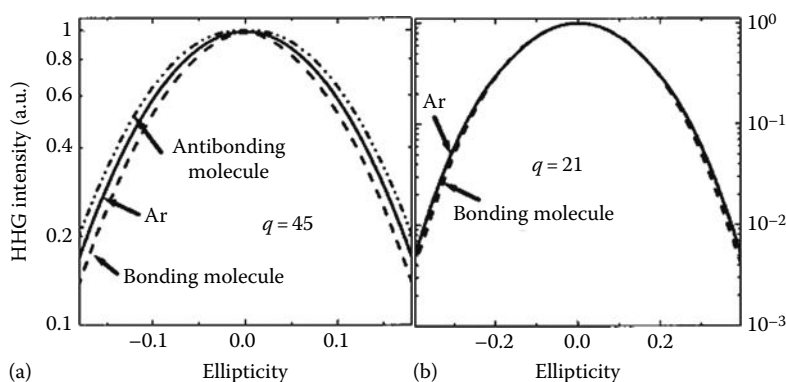


Figure 5.19 Calculated ellipticity dependence for (a) 45th and (b) 21st order harmonic from a bonding molecule, an antibonding molecule, and an atom. (From B. Shan, S. Ghimire, and Z. Chang, *Phys. Rev. A*, 69, 021404(R), 2004. Copyright 2004 by the American Physical Society.)

case. Again, bonding orbitals illustrated stronger ellipticity dependence, which is consistent with the measured results in Figure 5.19. The simulation results for the 21st harmonic from N₂ and Ar are shown in Figure 5.19b. The difference is very small, as was observed in the experiments.

High harmonic generation with aligned molecules leads to a powerful method for imaging the HOMO wave functions, as Corkum's group demonstrated. One of the future directions is to imaging the HOMO orbital when molecules are dissociating. This is important for visualizing chemical reactions.

5.4 Polarization Gating Revisit*

The principle of the polarization gating for generating single isolated attosecond pulses is introduced in Chapter 4. A simple expression is obtained for estimating the polarization gate width. Here, we study of the gating process by numerical simulations based on the SFA, which allows us to calculate the gated XUV spectrum and the duration of the attosecond pulses. The experiments that demonstrate polarization gating are presented in Chapter 8.

5.4.1 SFA for Polarization Gating

The laser field for polarization gating can be created by superimposing two counter-rotating circularly polarized pulses. By doing so, the opening time of the gate is so short that single attosecond pulses are generated. We assume that the peak field amplitude, E_0 , carrier frequency, ω_0 , pulse duration, τ_p , and carrier-envelope phase, φ_{CE} , are the same for the two pulses. The delay between them is T_d . The circularly polarized laser pulses are assumed to have a Gaussian shape with a carrier wavelength centered at 0.75 μm . The laser beam propagates in the z direction.

In experiments, the measured attosecond XUV light is the coherent superposition of the radiation from all the atoms in the laser-atom interaction region. To compare with the experiments, the attosecond pulse generation from the single atom driven by an NIR laser pulse with time-dependent ellipticity is simulated first under the SFA, then the macroscopic XUV signal is calculated by solving a 3D wave equation for the harmonic field. The second step is discussed in detail in Chapter 6.

5.4.1.1 Single Atom Response

The dipole moment of an atom in the time domain is calculated with the integral,

$$\begin{aligned} \vec{r}(t) = i \int_0^\infty d\tau \left(\frac{\pi}{\epsilon + i\tau/2} \right)^{3/2} \vec{d}^* [\vec{p}_s(t, \tau) - \vec{A}_L(t)] a^*(t) \\ \times e^{is(\vec{p}_s, t, \tau)} \vec{\epsilon}_L(t - \tau) \cdot \vec{d} [(\vec{p}_s(t, \tau) - \vec{A}_L(t - \tau))] a(t - \tau) + c.c. \end{aligned} \quad (5.141)$$

* This section is adapted from Chang, Z., Single attosecond pulse and xuv supercontinuum in the high-order harmonic plateau, *Phys. Rev. A* 70, 043802 (2004).

where

$\vec{\mathcal{E}}_L$ is the electric field of the laser pulse with a time-dependent ellipticity
 $\vec{\mathcal{A}}_L(t)$ is the vector potential
 I_p is the ionization potential of the generation target atom
 $a(t)$ is the ground-state amplitude, which is calculated by the ADK rate

The Neon atom is chosen for the simulation to avoid significant depletion of the ground state at the calculated intensity, 6×10^{14} W/cm².

Since the electrons only move in the plane perpendicular to the laser propagation direction, the dipole moment can be resolved into two components, $\vec{r}(t) = x(t)\hat{x} + y(t)\hat{y}$. The dipole moment along the $\hat{x}$ direction of the laser field is calculated by the integral,

$$\begin{aligned} x(t) \approx & i \int_0^\infty d\tau \left(\frac{\pi}{\varepsilon + i\tau/2} \right)^{3/2} d_x^* [\vec{p}_s(t, \tau) - \vec{A}_L(t)] e^{iS(\vec{p}_s, t, \tau)} \\ & \times \left\{ \varepsilon_x(t - \tau) \cdot d_x [\vec{p}_s(t, \tau) - \vec{A}_L(t - \tau)] + \varepsilon_y(t - \tau) \right. \\ & \left. \cdot d_y [\vec{p}_s(t, \tau) - \vec{A}_L(t - \tau)] \right\} |a(t)|^2 + c.c., \end{aligned} \quad (5.142)$$

and the dipole moment along the $\hat{y}$ direction is calculated by

$$\begin{aligned} y(t) \approx & i \int_0^\infty d\tau \left(\frac{\pi}{\varepsilon + i\tau/2} \right)^{3/2} d_y^* [\vec{p}_s(t, \tau) - \vec{A}_L(t)] e^{iS(\vec{p}_s, t, \tau)} \\ & \times \left\{ \varepsilon_{drive}(t - \tau) \cdot d_x [\vec{p}_s(t, \tau) - \vec{A}_L(t - \tau)] \right. \\ & \left. + \varepsilon_{gate}(t - \tau) \cdot d_y [\vec{p}_s(t, \tau) - \vec{A}_L(t - \tau)] \right\} |a(t)|^2 + c.c., \end{aligned} \quad (5.143)$$

where

$$\vec{\mathcal{E}}_L(t) = \varepsilon_x(t)\hat{x} + \varepsilon_y(t)\hat{y} = \varepsilon_{drive}(t)\hat{x} + \varepsilon_{gate}(t)\hat{y}, \quad (5.144)$$

$$\vec{\mathcal{A}}_L(t) = A_{drive}(t)\hat{x} + A_{gate}(t)\hat{y} \quad (5.145)$$

are the electric field and vector potential of the polarization gating field, which are resolved into the driving and gating components.

The dipole moment transition element is also a vector,

$$\vec{d}(\vec{p}) = d_x(\vec{p})\hat{x} + d_y(\vec{p})\hat{y}. \quad (5.146)$$

The x and y components of the dipole matrix elements

$$\begin{aligned} d_x^* (\vec{p}_s(t, \tau) - \vec{A}_L(t)) \\ = -i \frac{2^{7/2}}{\pi} \alpha^{5/4} \frac{p_{s,x}(t, \tau) - A_{drive}(t)}{\left\{ [p_{s,x}(t, \tau) - A_{drive}(t)]^2 + [p_{s,y}(t, \tau) - A_{gate}(t)]^2 + \alpha \right\}^3}, \end{aligned} \quad (5.147)$$

$$\begin{aligned} d_y^* (\vec{p}_s(t, \tau) - \vec{A}_L(t)) \\ = -i \frac{2^{7/2}}{\pi} \alpha^{5/4} \frac{p_{s,y}(t, \tau) - A_{gate}(t)}{\left\{ [p_{s,x}(t, \tau) - A_{drive}(t)]^2 + [p_{s,y}(t, \tau) - A_{gate}(t)]^2 + \alpha \right\}^3}. \end{aligned} \quad (5.148)$$

The expressions for the matrix elements $d_x[\vec{p}_s(t, \tau) - \vec{A}(t - \tau)]$ and $d_y[\vec{p}_s(t, \tau) - \vec{A}(t - \tau)]$ are similar to Equations 5.131 except the vector potential is at the time $t - \tau$.

The momentum corresponding to the stationary phase

$$\vec{p}_s(t, \tau) = p_{s,x}(t, \tau)\hat{x} + p_{s,y}(t, \tau)\hat{y}, \quad (5.149)$$

The two components of the momenta are calculated by

$$p_{s,x}(t, \tau) = \int_{t-\tau}^t dt'' A_{drive}(t'')/\tau, \quad (5.150)$$

and

$$p_{s,y}(t, \tau) = \int_{t-\tau}^t dt'' A_{gate}(t'')/\tau. \quad (5.151)$$

Finally, the action is calculated by

$$S(\vec{p}_s, t, \tau) = I_p \tau - \frac{1}{2} [p_{s,x}^2(t, \tau) + p_{s,y}^2(t, \tau)] + \frac{1}{2} \int_{t-\tau}^t dt'' [A_{drive}^2(t'') + A_{gate}^2(t'')]. \quad (5.152)$$

Calculations show that the amplitude of the XUV spectrum amplitude along $\hat{y}$ is much smaller than that along $\hat{x}$ because the major axis of the laser field ellipse is aligned to the $\hat{x}$ direction inside the polarization gate. Of course, if the ellipticity of the XUV pulse is the subject of study, the $\hat{y}$ component of the XUV field needed to be included.

5.4.1.2 Propagation Effects

The macroscopic XUV signal from all the atoms in the target is calculated by solving the electromagnetic-wave propagation equation for the XUV field that is introduced in Chapter 6. In the simulation, the laser is treated as a Gaussian beam with cylindrical symmetry about the z axis. The beam waist at the focus is $w_0 = 25 \mu\text{m}$, which gives a Rayleigh range of $z_R = 2.6 \text{ mm}$. A 1 mm long gas target is centered at 2 mm after the laser focus and the atomic density of the target is assumed to be a constant. The wave equation is solved numerically for each frequency in a spatial grid. The single atom dipole moments at the grid points are calculated first using Equations 5.142 and then are entered into wave equation. The output spectrum is calculated by adding up the power spectrum at each transverse point at the exit of the target.

To study the attosecond pulse in the time and spectral domain in the same time, one can apply the Gabor wavelet analysis. Here, we use a method that involves less computation. To obtain the attosecond pulse in the time domain, a square spectral window, instead of Gabor filter, is applied to the XUV spectrum at each transverse point at the exit of the target, and inverse Fourier transforms are performed to obtain the attosecond pulse for that point. The pulses of all the points are summed up to obtain the final pulse intensity. The width of the square window is $\Delta W = 9.9 \text{ eV}$ to reduce the effect of the window on the attosecond pulse duration.

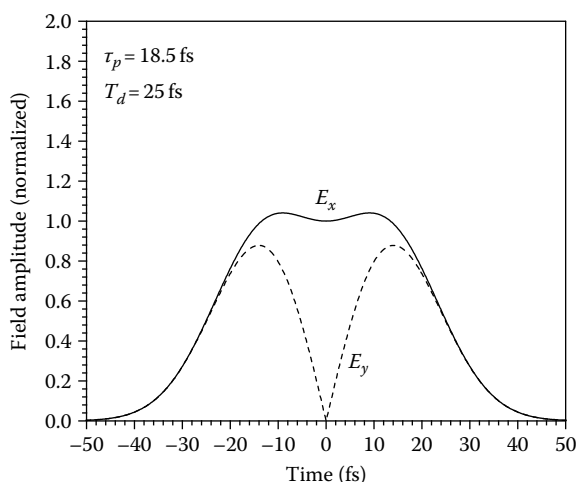


Figure 5.20 Amplitudes of the NIR laser-field components for polarization gating. The duration of each circularly polarized pulse is 18.5 fs.

5.4.2 Results of Simulations

5.4.2.1 Double Attosecond Pulses Generated with Multicycle NIR Lasers

The simulations are done for 18.75 fs laser pulses centered at 750 nm. The two circularly polarized pulses are separated by a 25 fs delay. The amplitudes of $\mathcal{E}_{drive}(t)$ and $\mathcal{E}_{gate}(t)$ are shown in Figure 5.20. The x field component is responsible for generating attosecond pulses whereas the y field suppresses the unwanted one. The carrier-envelope phase of the two pulses is 0 radians and the peak intensity of the linearly polarized portion is 6.4×10^{14} W/cm².

The XUV spectrum from a single atom calculated with the SFA is shown in Figure 5.21a. The complicated structure of the spectrum is a result of contribution of many quantum trajectories of the electron in the laser field. On the contrary, well-resolved high-order harmonic peaks are seen over the whole spectrum of the 3D propagation result, also shown in Figure 5.21a. This is because only radiation from the short trajectory survives.

The XUV pulses for two center frequencies are shown in Figure 5.21b. For a single atom, four attosecond pulses with comparable intensity are generated when their photon energies are centered at the 55th harmonic, which is in the plateau region. The spacing between two adjunct pulses is a quarter of a laser cycle. However, two of them correspond to the long trajectories that are suppressed by the phase matching. Consequently, only two attosecond pulses separated by half a laser cycle are left after the propagation, as the 3D simulation shows.

For the cutoff region around the 85th order, both the single atom calculation and the 3D simulation show that a pair of attosecond pulses are produced. They are separated by half a laser cycle. This is because the short and long trajectories merge into one trajectory for the generation of XUV light near the cutoff, as the semiclassical model predicted.

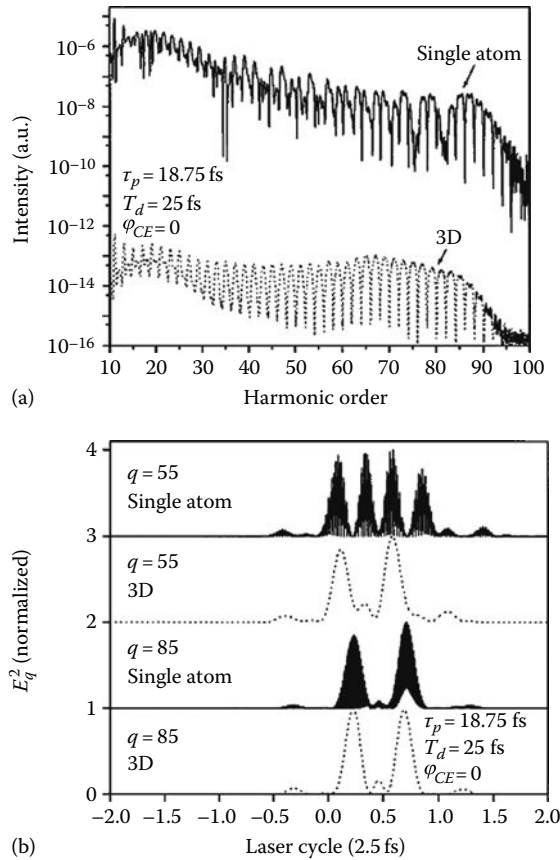


Figure 5.21 (a) The high-order harmonic spectra driving by a laser pulse with a time-dependent ellipticity. The pulse duration for both pulses is 18.75 fs and the delay between them is 25 fs. The peak intensity is 6×10^{14} W/cm². The carrier-envelope phase of the laser pulse is 0 rad. (b) The high harmonic pulses centered at two different frequencies: $55\hbar\omega_0$ and $85\hbar\omega_0$. The spectrum window is 9.9 eV. (From Z. Chang, *Phys. Rev. A*, 70, 043802, 2004. Copyright 2004 by the American Physical Society.)

For the given NIR laser pulse with a time-dependent ellipticity, the polarization gate width is $\delta t_{\xi < 0.2} \approx 0.3 \frac{18.75^2}{25} \approx 4.2$ fs, which is larger than the spacing between the two adjacent attosecond pulses, 1.25 fs. Consequently, more than one pulses are generated.

5.4.2.2 Isolated Attosecond Pulse Generated with Few-Cycle NIR Lasers

We then consider the case where the two counter-rotating circularly polarized pulses are 5 fs long. The delay between the two pulses is also 5 fs. The amplitudes of the two field components $\varepsilon_x(t)$ and $\varepsilon_y(t)$ are shown in Figure 5.22. For the given laser parameters, the polarization gate width in this case is $\delta t_{\xi < 0.2} \approx 1.5$ fs, which is close to the spacing between the two possible attosecond pulses from the short trajectory.

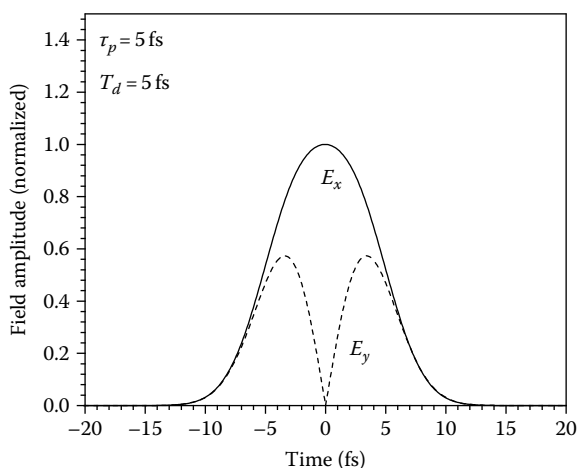


Figure 5.22 Amplitudes of the NIR laser-field components for polarization gating. The duration of each circularly polarized pulse is 5 fs.

The CE phase $\varphi_{CE} = \pi/2$ rad so that only a single attosecond pulse is generated. The peak intensity of the linearly polarized portion of the pulse inside the polarization gate is $6 \times 10^{14} \text{ W/cm}^2$.

Figure 5.23a compares the XUV spectrum calculated for a single atom to the spectrum that takes into account the macroscopic propagation effects. The single atom spectrum shows large modulations. This is the result of the interference between two pulses, as Figure 5.23b reveals. In the plateau region, two pulses with comparable intensities are emitted from a single atom. The first one corresponds to the short trajectory (labeled by τ_1) and the second one corresponds to the long trajectory (labeled by τ_2). This is true for all of the four center frequencies. The spectrum modulation period increases with the photon energy because the pulse separation decreases.

The emission time for both trajectories depends on the central frequency. It is clear that the long trajectory is negatively chirped. On the contrary, the XUV pulse from the short trajectory is positively chirped as the center energy increases with the emission time. The sign of the chirp is consistent with the predictions of the semiclassical model.

The 3D result shows that the XUV spectrum is a supercontinuum above the 25th harmonic photon energy. This is because only the short trajectory survives propagation, as shown in Figure 5.23c. Thus phase-matching plays an important role in the generation of single attosecond pulses.

The duration of the XUV pulses are 400 as for the four different central frequencies located in a range from the deep plateau to the cutoff. For the clearness of presentation, the pulse intensities are normalized in Figure 5.23b and c. Obviously, pulses with different center energy are generated at different time. When the whole spectrum is used to generate an isolated XUV pulse, the pulse is chirped. The sign of the chirp is the same as that of the short trajectory. In the next chapter, the chirp of the attosecond pulses is analyzed in detail.

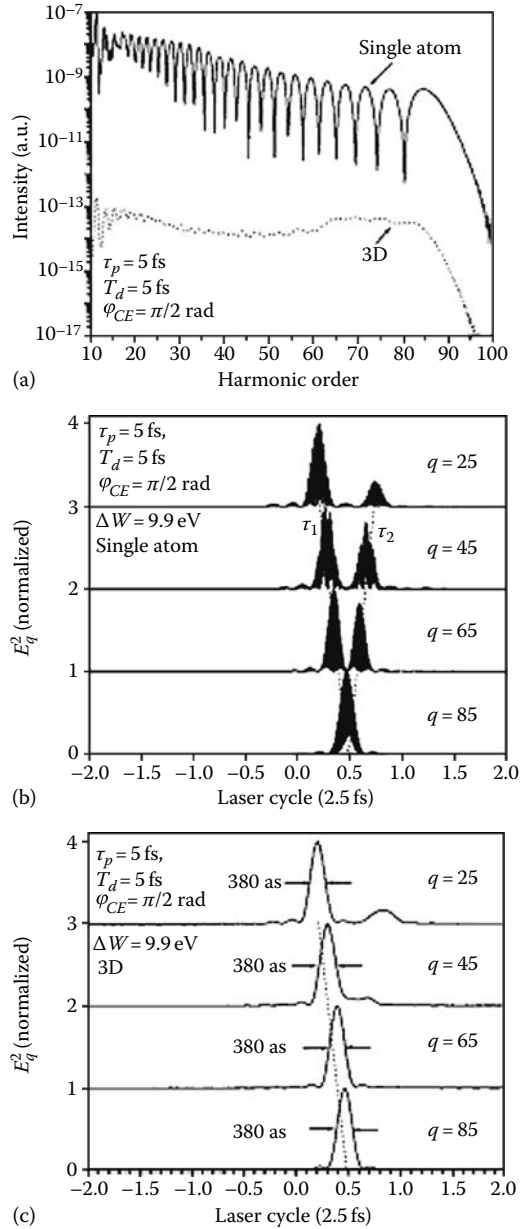


Figure 5.23 (a) The high-order harmonic spectra from Neon atoms driving by a laser pulse with a time-dependent ellipticity. The laser pulse is formed by the combination of a left-hand circularly polarized pulse and a right-hand circularly polarized pulse. The pulse duration for both pulses is 5 fs and the delay between them is 5 fs. The peak intensity is 6×10^{14} W/cm². The carrier-envelope phase of the laser pulse is $\varphi_{CE} = \pi/2$ rad. (b) The high harmonic pulses centered at four different frequencies: $25\hbar\omega_0$, $45\hbar\omega_0$, $65\hbar\omega_0$, and $85\hbar\omega_0$. The spectrum window is 9.9 eV. The results are obtained from the single atom calculation. (c) The attosecond pulses from the 3D propagation simulation. (From Z. Chang, *Phys. Rev. A*, 70, 043802, 2004. Copyright 2004 by the American Physical Society.)

5.4.2.3 Effects of Carrier-Envelope Phase

Figure 5.24a compares the XUV spectra after the propagation obtained under the same conditions as Figure 5.23a for two carrier-envelope phases: $\varphi_{CE} = \pi/2$ and $\varphi_{CE} = 0$. The spectrum is a supercontinuum in the former case. For $\varphi_{CE} = 0$, however, high-order harmonic peaks appear, as a result of interference between two pulses. Indeed, there are a pair of attosecond pulses for $\varphi_{CE} = 0$ as shown in Figure 5.24b. Their intensity is

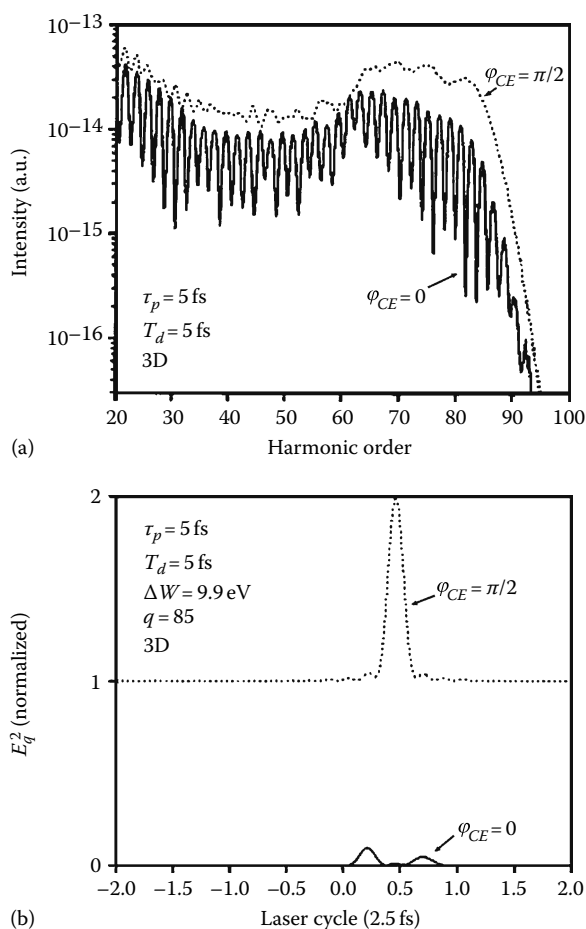


Figure 5.24 (a) The high-order harmonic spectra from Neon atoms driving by a laser pulse with a time-dependent ellipticity formed by the combination of a left-hand circularly polarized pulse and a right-hand circularly polarized pulse. The pulse duration for both pulses is 5 fs and the delay between them is 5 fs. The peak intensity is $6 \times 10^{14} \text{ W/cm}^2$. The dotted line is when the carrier-envelope phase of the laser pulse $\varphi_{CE} = \pi/2$ rad and the solid line is when the phase equals zero. (b) The high harmonic pulses centered at $85\hbar\omega$. The spectrum window is 9.9 eV. The upper curve (dotted line) is when the carrier-envelope phase is $\pi/2$ rad. The lower curve (solid line) is for the phase equaling zero and its intensity is normalized to the upper one. (From Z. Chang, *Phys. Rev. A*, 70, 043802, 2004. Copyright 2004 by the American Physical Society.)

much lower than that of the single attosecond pulse when $\varphi_{CE} = \pi/2$, because they are generated near the two edges of the polarization gate. Apparently, the carrier-envelope phase significantly affects the attosecond pulse generation process when the polarization gate width is this narrow. In order to produce single attosecond pulses under the given conditions, the carrier-envelope phase must be stabilized. Methods for stabilizing the CE phase of high-power laser amplifiers are discussed Chapter 3. One should keep in mind that the CE phase of a focused laser beam changes over the Rayleigh range. Thus the gas target in experiments should be much shorter than the confocal parameter of the laser.

The CE phase effects can be understood by examining the electron ionization and return as illustrated in Figure 5.25. The driving-field

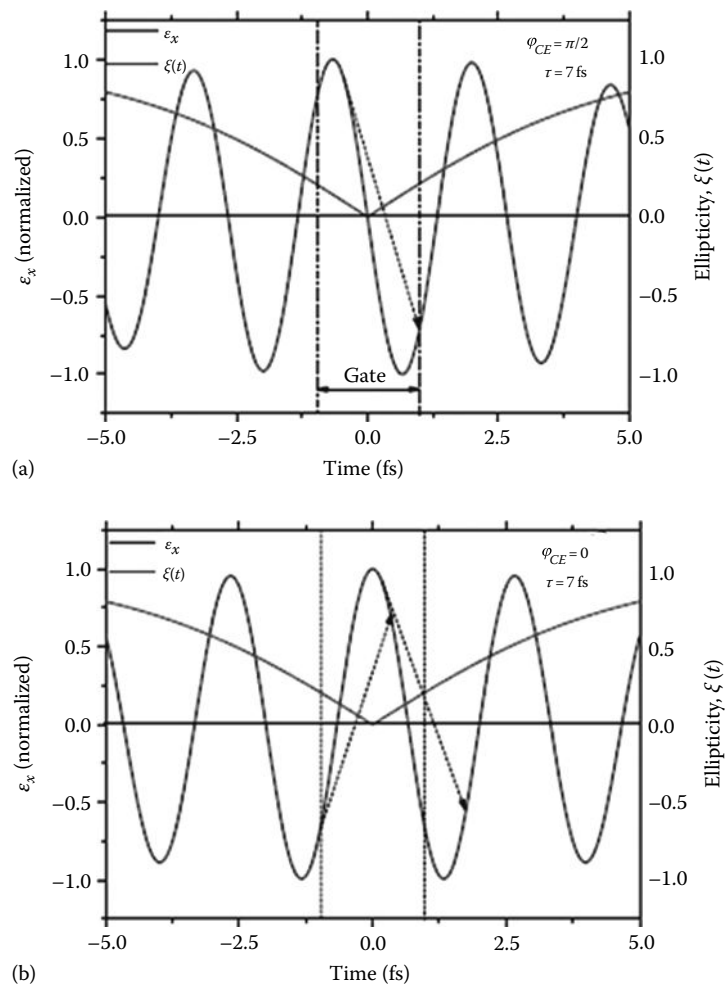


Figure 5.25 (a) The effect of the carrier-envelope phase of the driving laser pulse on the returning of the electron within the polarization gate. The carrier envelope phase is 90° in (a) and is 0° in (b). The solid line is the electric field of one component of the laser pulse. The dashed line is the ellipticity. The range within the two vertical dotted lines is the polarization gate width.

oscillation shifts in time relative to the polarization gate as the CE phase changes. At CE phase $\varphi_{CE} = \pi/2$, only electrons released between $t = -0.125$ fs and $t = 0$ fs can return within the gate as indicated by the arrow leading to the emission of an isolated attosecond pulse. On the contrary, at CE phase $\varphi_{CE} = 0$, as shown in Figure 5.25b, two groups of electrons with short trajectories are released within the gate. One returns within the gate, and the other returns outside the gate but the ellipticity is still small. In this case, two weaker attosecond pulses are produced.

5.5 Complete Reconstruction of Attosecond Burst

The characterization of the XUV attosecond pulses can be performed using an extension of the frequency resolved optical gating technique developed for characterizing femtosecond laser pulses, which is introduced in Chapter 2. The technique is called complete reconstruction of attosecond burst, or CRAB. CRAB is based on the photoionization of atoms by the XUV pulse, in the presence of an intense near infrared laser field. The XUV pulse ionizes a gas, by single photon absorption, thus, generating an attosecond electron pulse that is a replica of the attosecond photon pulse.

The photoelectrons are momentum shifted by the NIR laser field, which is the foundation of the attosecond streak camera described in Chapter 1. The photoelectron spectra modified by the laser are measured as a function delay between the attosecond pulse and the laser field. This provides a two-dimensional (2D) set of data, called a spectrogram or FROG-CRAB trace. The XUV pulse shape and duration can be retrieved by using various iterative reconstruction algorithms. Details on the experimental implementation and pulse reconstruction are discussed in Chapters 7 and 8. Here, we derive the equations that express the CRAB trace.

5.5.1 Approximations

5.5.1.1 Strong Field Approximation

The key approximation in the SFA discussed in Section 5.1 is the neglect of the effects of excited states during the ionization of the ground state. It is a good approximation for the NIR lasers ($\hbar\omega_{IR} \ll I_p$) when the Keldysh parameter is much smaller than one, i.e., in the tunneling ionization regime. Strong field is required because more than one photons are needed to liberate one electron. For photon energy $\hbar\omega_X > I_p$, which is the case of attosecond light, ionization occurs even if XUV field is weak. Single photon ionization of atoms by XUV and x-ray has been studied by using synchrotrons since 1960s. It is known that excited states do play important roles in explaining the autoionization resonance peaks of photo-absorption cross section. If we choose the photon energy range where the cross is smooth, then the effects of excited states can be ignored.

The SFA also ignores the effects of the parent ionic potential on the electron motion after ionization. It is valid for NIR field in the tunneling region because the NIR laser field is much stronger than the Coulomb field when the freed electron is not too close to the nucleus. When the

photoionization is induced by an XUV field with photon energy $\hbar\omega_X \gg I_p$, the kinetic energy of the electron is so large that it quickly moves away from the nucleus. In the spatial region where the electron momentum is changed by the NIR field, the ionic potential can be neglected. In this regime, the conditions on the laser field intensity to neglect the ionic potential are much less stringent than the ionization by the NIR alone.

Despite its name, the SFA for CRAB can be expected to be satisfactory even for moderate NIR laser intensities and very weak XUV field. In CRAB measurements, the laser intensity is on the order of 10^{12} W/cm² or higher. As a comparison, for attosecond pulse generation, the NIR laser intensity is $\sim 10^{14}$ W/cm².

5.5.1.2 Single Active-Electron Approximation

Although almost all atoms used as the detection gases contain more than one electron, for mathematical simplicity we assume that only one electron is active. The effects of electron correlation are considered in Chapter 9 when the autoionization process and double excitation are studied. The XUV photon energy should be smaller than the banding energy of the inner shells otherwise the mixture of the electron signals from different shells makes it difficult to retrieve the attosecond pulses.

5.5.2 Ionization in Two-Color Field

The physical process of the CRAB measurement is the ionization of atom in simultaneous action of the attosecond XUV field and the NIR laser field, which is a type of two-color field.

5.5.2.1 XUV Field

In the time domain, an attosecond XUV pulses can be described as

$$\mathcal{E}_X(t) = E_X(t)e^{i[\omega_X t + \phi_X(t)]}, \quad (5.153)$$

where the pulse envelope $E_X(t)$ and the temporal phase $\phi_X(t)$ need to be determined by the CRAB.

In the frequency domain, the XUV field can be described by the Fourier transform

$$\tilde{E}_X(\omega) = \int_{-\infty}^{+\infty} \mathcal{E}_X(t)e^{-i\omega t} dt = |\tilde{E}_X(\omega)|e^{i\varphi(\omega)}, \quad (5.154)$$

where $\varphi(\omega)$ is the spectral phase. The power spectrum of an XUV pulse can be measured with an XUV spectrometer. It is

$$I(\omega) = |\tilde{E}_X(\omega)|^2 = \left| \int_{-\infty}^{+\infty} \mathcal{E}_X(t)e^{-i\omega t} dt \right|^2. \quad (5.155)$$

The Fourier transform of $\sqrt{I(\omega)}$ gives the shortest pulses that can be supported by the measured spectrum. The pulse duration determined by the FROG-CRAB should be equal to or larger than the transform-limited value.

5.5.2.2 Photoelectron Wave Packet

We consider an atom with ionization potential I_p , photoionized by an XUV electric field $\vec{\varepsilon}_X(t)$, in the presence of a low-frequency laser field $\vec{\varepsilon}_L(t) = -\frac{\partial \mathcal{A}_L}{\partial t}$ shifted by a variable delay τ_d where $\mathcal{A}_L(t)$ is the vector potential of this laser field. In experiments, both the XUV and NIR fields are focused to the gas target. For mathematical simplicity, we assume both of the fields are plane waves.

Under the single active-electron approximation and the dipole approximation, the Schrödinger equation (in atomic unit) corresponding to the photoionization in the two-color field is

$$i \frac{\partial}{\partial t} \Psi(\vec{r}, t) = \left[-\frac{1}{2} \nabla^2 + V(\vec{r}) - \vec{r} \cdot \vec{\varepsilon}(t) \right] \Psi(\vec{r}, t), \quad (5.156)$$

where

$V(\vec{r})$ is the potential of the atom

$\vec{\varepsilon}(t) = \vec{\varepsilon}_X(t) + \vec{\varepsilon}_L(t)$ is the total field

The equation can be solved analytically by applying the SFA. As a result, the Coulomb continuum eigenfunctions may be substituted by plane waves. The solution can be expressed by

$$\Psi(\vec{r}, t) = e^{-iI_p t} \left[|0\rangle + \int d^3 v \, b(\vec{v}, t) |\vec{v}\rangle \right], \quad (5.157)$$

where $|0\rangle$ denotes the ground state with ionization potential I_p , which has the same form as Equation 5.15 for attosecond pulse generation.

The plane wave component with momentum $\vec{v}$ is given by

$$|\vec{v}\rangle = e^{-i\vec{v} \cdot \vec{r}}, \quad (5.158)$$

where $\vec{v}$ is the electron momentum.

$b(\vec{v}, t)$ is related to the momentum space wave function of free electrons. In other words, it is related to the amplitude and phase of each momentum components. It is a function of time because the amplitude and phase are changed by the time dependent external field $\vec{\varepsilon}(t)$.

Without the laser field, the transition amplitude to the final continuum state with momentum $\vec{v}$ at a time t is given by

$$b(\vec{v}, t) \approx i \int_{-\infty}^t dt' \vec{\varepsilon}_X(t') \cdot \vec{d}[\vec{v} - \vec{\mathcal{A}}_X(t) + \vec{\mathcal{A}}_X(t')] e^{-i \left(\frac{[\vec{v} - \vec{\mathcal{A}}_X(t) + \vec{\mathcal{A}}_X(t')]^2}{2} + I_p \right) t}, \quad (5.159)$$

which is essentially expression 5.46 except that the field now is the XUV field.

For the weak XUV field considered, $\vec{\mathcal{A}}_X(t) \ll \vec{v}$, thus when the attosecond pulse passed the detection gas target ($t = \infty$)

$$\begin{aligned} b(\vec{v}) &= i \int_{-\infty}^{\infty} \vec{\varepsilon}_X(t) \cdot \vec{d}(\vec{v}) e^{-i \left(\frac{v^2}{2} + I_p \right) t} dt \\ &= i \vec{d}(\vec{v}) \cdot \int_{-\infty}^{\infty} \vec{\varepsilon}_X(t) e^{-i \left(\frac{v^2}{2} + I_p \right) t} dt, \end{aligned} \quad (5.160)$$

When the XUV field is a monochromatic, i.e., $\varepsilon_X(t) = E_X e^{i\omega_X t}$,

$$\begin{aligned} b(\vec{v}) &= i\vec{d}(\vec{v}) \cdot \vec{E}_X \int_{-\infty}^{\infty} e^{i\omega_X t} e^{-i\left(\frac{v^2}{2} + I_p\right)t} dt \\ &= i\vec{d}(\vec{v}) \cdot \vec{E}_X \delta\left[\omega_X - \left(\frac{v^2}{2} + I_p\right)\right], \end{aligned} \quad (5.161)$$

which yields $\frac{v^2}{2} = \omega_X - I_p$, consistent with Einstein's law of photoelectric effect.

In general, $\frac{v^2}{2} + I_p = \omega$, where ω is the frequency of the XUV pulse, we have

$$b(\vec{v}) = i\vec{d}(\vec{v}) \cdot \vec{E}_X \left(\frac{v^2}{2} + I_p\right) = i\vec{d}(\vec{v}) \cdot \vec{E}_X(\omega), \quad (5.162)$$

where $d(\vec{v})$ is the dipole transition-matrix element between the ground state and the continuum state. The transition is shown in Figure 5.26.

Equation 5.162 shows that the photoelectron spectrum is directly related to the attosecond field spectrum, both in phase and amplitude. Thus, when $\vec{d}(\vec{v}) \approx \text{constant}$ the photoelectron pulse is considered as a replica of the XUV attosecond pulse. The phase and amplitude of attosecond pulses can be determined by measuring the electron pulse.

5.5.2.3 Effects of Dipole Matrix Elements

The dipole transition-matrix element $\vec{d}(\vec{v}) = \langle \vec{v} | \vec{r} | 0 \rangle$ is determined by the detection atom, which is related to the photoionization cross sections. For simplicity, consider the case that $\vec{d}(\vec{v})$ and $\vec{E}_X$ are pointing to the same direction, then

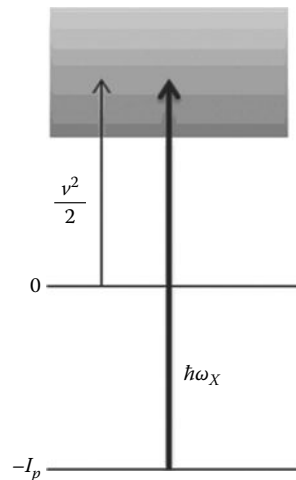


Figure 5.26 Photoionization in XUV field.

$$\frac{b^2(K)}{d^2(K)} = |\tilde{E}_X(\omega)|^2, \quad (5.163)$$

where the kinetic energy of the electron is $K = \frac{v^2}{2}$. In other words, the XUV power spectrum can be obtained by dividing the measured spectrum by the square of the dipole transition element.

The two spectra might differ in shape if $\vec{d}(\vec{v})$ depends on $\vec{v}$. The ionization cross section is generally well known, which allows one to correct for this dependence. If this is not the case, the attosecond field spectral amplitude can be measured independently, using an XUV spectrometer. The two spectra might differ in phase because of a possible phase dependence of $\vec{d}(\vec{v})$ on $\vec{v}$, which can, for instance, be expected to occur near some resonances. If this phase dependence is known either from theory or experiment, the spectral phase of the attosecond pulse can be directly deduced from the spectral phase of the electron wave packet. For attosecond fields with limited bandwidth (<10 eV), this correction can be neglected.

5.5.2.4 Photoelectron Wave Packet Produced by the Two-Color Field

We now turn to the effect of a low-frequency laser field on the photoelectron spectrum. We consider the situation where there is no time delay between the XUV pulse and the NIR field. When the laser is turned on, within the SFA, the amplitude of populating a state with kinetic momentum v at a moment t is

$$b(\vec{v}, t) = i \int_{-\infty}^t \vec{\mathcal{E}}(t') \cdot \vec{d}[\vec{v} - \vec{A}(t) + \vec{A}(t')] e^{-i \int_{t'}^t \frac{1}{2} [\vec{v} - \vec{A}(t) + \vec{A}(t'')]^2 dt'' - i l_p t'} dt'. \quad (5.164)$$

The field in dot product is $\vec{\mathcal{E}}(t) \approx \vec{\mathcal{E}}_X(t)$ because the bound to continuum transition probability is dominated by the XUV field. The vector potential contribution from the XUV field is much smaller than that of laser and can therefore be neglected. Thus,

$$b(\vec{v}, t) = i \int_{-\infty}^t \vec{\mathcal{E}}_X(t') \cdot \vec{d}[\vec{v} - \vec{A}_L(t) + \vec{A}_L(t')] e^{-i \int_{t'}^t \frac{1}{2} [\vec{v} - \vec{A}_L(t) + \vec{A}_L(t'')]^2 dt'' - i l_p t'} dt'. \quad (5.165)$$

The electron spectrum is measured at $t = \infty$. Considering $\vec{A}_L(\infty) = 0$ for pulse laser field, we have

$$b(\vec{v}) = i \int_{-\infty}^{\infty} \vec{\mathcal{E}}_X(t') \cdot \vec{d}(\vec{v} + \vec{A}_L(t')) e^{-i \int_{t'}^{\infty} \frac{1}{2} [\vec{v} - \vec{A}_L(t'')]^2 dt'' - i l_p t'} dt', \quad (5.166)$$

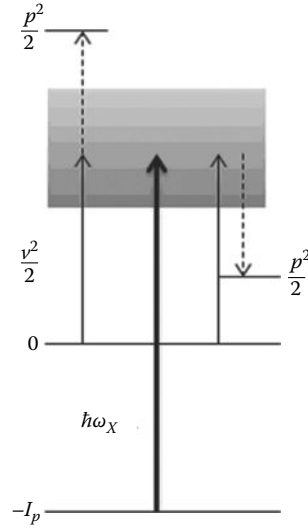


Figure 5.27 Photoionization in two-color field $\vec{\varepsilon}(t) = \vec{\varepsilon}_X(t) + \vec{\varepsilon}_L(t)$.

which is equivalent to

$$b(\vec{v}) = i \int_{-\infty}^{\infty} \vec{\varepsilon}_X(t) \cdot \vec{d}[\vec{v} + \vec{A}_L(t)] e^{-i \int_t^{\infty} \frac{1}{2} [\vec{v} + \vec{A}_L(t')]^2 dt' - i I_p t} dt. \quad (5.167)$$

The transition is shown in Figure 5.27.

5.5.2.5 Time Delay between the Two Fields

When there is a time delay, τ_d , between the XUV and the intense NIR laser pulses

$$\vec{\varepsilon}(t, \tau_d) = \vec{\varepsilon}_X(t - \tau_d) + \vec{\varepsilon}_L(t), \quad (5.168)$$

which means that

$$\Psi(\vec{r}, t, \tau_d) = e^{-i I_p t} \left[|0\rangle + \int d^3 v \, b(\vec{v}, t, \tau_d) |\vec{v}\rangle \right] \quad (5.169)$$

and

$$b(\vec{v}, \tau_d) = i \int_{-\infty}^{+\infty} \vec{\varepsilon}_X(t - \tau_d) \cdot \vec{d}[\vec{v} + \vec{A}_L(t)] e^{-i \int_t^{\infty} \left(\frac{(\vec{v} + \vec{A}_L(t'))^2}{2} + I_p \right) dt'} dt. \quad (5.170)$$

For simplicity, we consider the case that the XUV pulse shape is Gaussian. This consideration means that the $1/e^2$ pulse width is τ_X . The XUV pulse can be expressed as

$$\vec{\varepsilon}_X(t) = \vec{E}_X(t) e^{-\frac{t^2}{2\tau_X^2} + i\omega_X t}, \quad (5.171)$$

where the complex quantity $\vec{E}_X(t)$ contains both the peak field amplitude and the temporal phase.

In this case,

$$b(\vec{v}, \tau_d) = i \int_{-\infty}^{\infty} \vec{E}_X(t - \tau_d) \cdot \vec{d}[\vec{v} + \vec{A}_L(t)] e^{-\frac{(t-\tau_d)^2}{2\tau_X^2} + i\omega_X(t-\tau_d) - i \int_t^{\infty} \frac{1}{2} [\vec{v} + \vec{A}_L(t')]^2 dt' - i p t} dt. \quad (5.172)$$

5.5.3 Saddle Point Approximation

The integral in Equation 5.172 can be performed by making the saddle point approximation, similar to the approach introduced in [Section 5.1.3](#) when attosecond pulse generation is considered.

When $\left. \frac{d}{dx} f(x) \right|_{x_0} = 0, \left. \frac{d^2}{dx^2} f(x) \right|_{x_0} = 0$, the integral

$$\int_a^b e^{f(x)} dx \approx \sqrt{\frac{2\pi}{f''(x_0)}} e^{f(x_0)}. \quad (5.173)$$

For simplicity, we introduce a quantity $\vec{p}(t) = \vec{v} + \vec{A}_L(t)$, which is the momentum of the electron at the time when it is ionized while $\vec{v}$ is the momentum of the electron at the time of measurement. The saddle point of the function,

$$f(t) = -\frac{(t - \tau_d)^2}{2\tau_X^2} + i\omega_X(t - \tau_d) - i \int_t^T \left(\frac{p^2(t')}{2} + I_p \right) dt', \quad (5.174)$$

is defined as

$$\frac{d}{dt} f(t) = -\frac{t - \tau_d}{\tau_X^2} + i\omega_X - i \left(\frac{p^2(t)}{2} + I_p \right) = 0, \quad (5.175)$$

or

$$-\frac{t_s - \tau_d}{\tau_X^2} + iK_0 - i \left(\frac{p^2(t_s)}{2} \right) = 0, \quad (5.176)$$

where $K_0 = \omega_X - I_p$ is the center energy of the photoelectron.

Equation 5.176 can be rewritten as

$$t_s - \tau_d = i \left[K_0 - \frac{p^2(t_s)}{2} \right] \tau_X^2. \quad (5.177)$$

The second derivative of this function is

$$\begin{aligned} \frac{d^2}{dt^2} f(t) &= -\frac{1}{\tau_X^2} - ip(t) \frac{dp}{dt} = -\frac{1}{\tau_X^2} - ip(t) \frac{d}{dt} [v + A_L(t)] \\ &= -\frac{1}{\tau_X^2} + ip(t) \varepsilon_L(t). \end{aligned} \quad (5.178)$$

Thus,

$$\begin{aligned}\frac{d^2 f(t)}{dt^2}\bigg|_{t_s} &= -\frac{1}{\tau_X^2} + ip(t_s)\varepsilon_L(t_s) = -\frac{1}{\tau_X^2} [1 - ip(t_s)\varepsilon_L(t_s)\tau_X^2] \\ &= -\frac{1}{\tau_X^2} \sqrt{1 + [p(t_s)\varepsilon_L(t_s)\tau_X^2]^2} e^{-ia \tan[p(t_s)\varepsilon_L(t_s)\tau_X^2]}.\end{aligned}\quad (5.179)$$

Considering

$$\begin{aligned}\frac{d}{dt} \vec{d}[\vec{p}(t)] &= \frac{d}{dp} d[\vec{p}(t)] \frac{d}{dt} p(t) \approx \vec{d}[\vec{p}(t)] \frac{d}{dt} [v + \mathcal{A}_L(t)] \\ &= -\vec{d}[\vec{p}(t)] \varepsilon_L(t_s),\end{aligned}\quad (5.180)$$

and let $\mu = \sqrt{1 + [p(t_s)\varepsilon_L(t_s)\tau_X^2]^2}$. This leads to

$$\begin{aligned}b(\vec{v}, \tau_d) &\approx \tau_X^2 \sqrt{\frac{2\pi}{\mu}} e^{i\frac{1}{2}a \tan[p(t_s)\varepsilon_L(t_s)\tau_X^2]} \vec{E}_X(t_s - \tau_d) \vec{d}[\vec{p}(t_s)] \\ &\times e^{\frac{\mu}{2\tau_X^2}} e^{-\frac{(t_s - \tau_d)^2}{2\tau_X^2} + i\omega_X(t_s - \tau_d) - i \int_{t_s}^{\infty} \left(\frac{p^2(t')}{2} + I_p\right) dt'}.\end{aligned}\quad (5.181)$$

Finally, the measured photoelectron spectrum

$$|b(\vec{v}, K_0)|^2 = 2\pi\tau_X^2 \frac{|\vec{E}_X(t_s - \tau_d) \vec{d}[\vec{p}(t_s)]|^2}{\sqrt{1 + [p(t_s)\varepsilon_L(t_s)\tau_X^2]^2}} e^{-\frac{(t_s - \tau_d)^2}{2\tau_X^2}}.\quad (5.182)$$

Now, if we insert

$$t_s - \tau_d = i \left[K_0 - \frac{p^2(t_s)}{2} \right] \tau_X^2\quad (5.183)$$

into Equation 5.182, we have

$$|b(\vec{v}, \tau_d)|^2 = 2\pi\tau_X^2 \frac{|\vec{E}_X(t_s - \tau_d) \vec{d}[\vec{p}(t_s)]|^2}{\mu} e^{-\left[K_0 - \frac{p^2(t_s)}{2}\right]^2 \tau_X^2}.\quad (5.184)$$

When the XUV pulse is sufficiently short, $t_s = \tau_d$, which corresponds to the peak of the XUV pulse.

$$|b(\vec{v}, \tau_d)|^2 = 2\pi\tau_X^2 \frac{|\vec{E}_X(0) \vec{d}[\vec{p}(\tau_d)]|^2}{\sqrt{1 + [p(\tau_d)\varepsilon_L(\tau_d)\tau_X^2]^2}} e^{-\left[K_0 - \frac{p^2(\tau_d)}{2}\right]^2 \tau_X^2}\quad (5.185)$$

or

$$\begin{aligned}|b(\vec{v}, \tau_d)|^2 &= 2\pi\tau_X^2 \frac{|\vec{E}_X(0) \vec{d}[\vec{v} + \vec{A}_L(\tau_d)]|^2}{\sqrt{1 + [\vec{v} + \vec{A}_L(\tau_d)] \varepsilon_L(\tau_d) \tau_X^2]^2}} \\ &\times e^{-\left[K_0 - \frac{[\vec{v} + \vec{A}_L(\tau_d)]^2}{2}\right]^2 \tau_X^2}.\end{aligned}\quad (5.186)$$

5.5.4 FROG-CRAB Trace

The measurements are always done at a time when both pulses are completely gone, so $t = +\infty$, implying

$$b(\vec{v}, \tau_d) = i \int_{-\infty}^{+\infty} \vec{\mathcal{E}}_X(t - \tau_d) \cdot \vec{d}[\vec{v} + \vec{\mathcal{A}}_L(t)] e^{-i \int_t^{\infty} \left(\frac{(\vec{v} + \vec{\mathcal{A}}_L(t'))^2}{2} + I_p \right) dt'} dt. \quad (5.187)$$

Another way of expressing Equation 5.187 is

$$b(\vec{v}, \tau_d) = i \int_{-\infty}^{\infty} \vec{\mathcal{E}}_X(t - \tau_d) \cdot \vec{d}[\vec{v} + \vec{\mathcal{A}}_L(t)] e^{i\Phi_G(t)} e^{-i\left(\frac{v^2}{2} + I_p\right)t} dt, \quad (5.188)$$

which implies

$$\Phi_G(t) = - \int_t^{\infty} \left[\vec{v} \cdot \vec{\mathcal{A}}_L(t') + \frac{1}{2} \mathcal{A}_L^2(t') \right] dt', \quad (5.189)$$

where $\Phi_G(t)$ is the quantum phase acquired by the electron due to its interaction with the laser field. The phase it accumulates along this trajectory is thus temporally modulated by the dressing laser field.

Equation 5.188 has a very intuitive interpretation. $b(\vec{v}, \tau)$ is the sum of the probability amplitudes of all electron trajectories leading to the same final velocity $\vec{v}$.

5.5.4.1 Electron Phase Modulator

The main effect of the laser field is to induce a temporal phase modulation $\Phi_G(t)$ on the electron wavepacket generated in the continuum by the XUV field. Because of the scalar product $\vec{v} \cdot \vec{\mathcal{A}}_L(t')$ in Equation 5.189, the photoelectrons have to be observed in a given direction for the phase modulation to be well defined. With these conditions, the laser field acts on electron wave packets just as a conventional phase modulator used in optics acts on light pulses. We therefore consider it as an ultrafast electron phase modulator.

5.5.4.2 FROG-CRAB Trace

The transition amplitude cannot be measured directly. The photon electron spectrum is the power spectrum, not amplitude, which can be represented by the expression

$$S_e(v, \tau_d) = \left| \int_{-\infty}^{\infty} \vec{\mathcal{E}}_X(t - \tau_d) \cdot \vec{d}[\vec{v} + \vec{\mathcal{A}}_L(t)] e^{i\Phi_G(\vec{v}, t)} e^{-i(K + I_p)t} dt \right|^2, \quad (5.190)$$

where the kinetic energy $K = \frac{1}{2} v^2$.

The linear temporal phase, $iI_p t$, corresponds to a frequency offset in the frequency domain, which can be dropped. Thus,

$$S_e(\vec{v}, \tau_d) = \left| \int_{-\infty}^{\infty} \varepsilon_X(t - \tau_d) d[\vec{v} + \vec{A}_L(t)] e^{i\Phi_G(\vec{v}, t)} e^{-iKt} dt \right|^2, \quad (5.191)$$

$$\varphi(\vec{v}, t) = - \int_t^{\infty} \left[\vec{v} \cdot \vec{A}_L(t') + \frac{1}{2} A_L^2(t') \right] dt'. \quad (5.192)$$

$S_e(\vec{v}, \tau_d)$ is the FROG-CRAB trace that can be measured experimentally. The setups for measuring the trace are described in Chapters 7 and 8.

5.5.4.3 Dipole Correction

It is possible to reconstruct the phase and shape of the XUV pulse from the trace described in Equation 5.191. However, we need to reduce the effects of the frequency response of the photo-emission process, which is described by the dipole transition-matrix element.

In conventional FROG, the measured trace can be expressed as

$$S(\omega, \tau_d) = \left| \int_{-\infty}^{+\infty} E_L(t - \tau_d) G(t) e^{-i\omega t} dt \right|^2. \quad (5.193)$$

The temporal gate $G(t)$ depends on time, not on frequency. However, the dipole transition-matrix element, $d[\vec{v} + \vec{A}_L(t)]$, is a function of momentum and time, which makes it difficult to reconstruct the phase of the XUV pulse using the FROG method.

The dipole transition-matrix element can be taken out of the integral by inserting the saddle point value of time into the vector potential, i.e.,

$$S_e(v, \tau_d) = \left| \int_{-\infty}^{\infty} \varepsilon_X(t - \tau_d) d[\vec{v} + \vec{A}_L(t_s)] e^{i\Phi_G(\vec{v}, t)} e^{-iKt} dt \right|^2, \quad (5.194)$$

or

$$S_e(v, \tau_d) = d^2[\vec{v} + \vec{A}_L(t_s)] \left| \int_{-\infty}^{\infty} \varepsilon_X(t - \tau_d) e^{i\Phi_G(\vec{v}, t)} e^{-iKt} dt \right|^2. \quad (5.195)$$

From the saddle point approximation, we have

$$t_s = \tau + i \left[K_0 - \frac{[\vec{v} + \vec{A}_L(t_s)]^2}{2} \right] \tau_x^2 \approx \tau_d. \quad (5.196)$$

Thus

$$S_e(v, \tau_d) = d^2[\vec{v} + \vec{A}_L(\tau_d)] \left| \int_{-\infty}^{\infty} \varepsilon_X(t - \tau_d) e^{i\Phi_G(\vec{v}, t)} e^{-iKt} dt \right|^2. \quad (5.197)$$

5.5.4.4 Central Momentum Approximation

By making the central momentum approximation, that is, by replacing $\Phi_G(\vec{v}, t)$ with $\Phi_G(\vec{v}_0, t)$, where $\vec{v}_0 = \omega_X - I_p$ is the central momentum of the photoelectron, the XUV attosecond pulse can be reconstructed from the trace

$$S(v, \tau_d) = \frac{S_e(v, \tau_d)}{d^2 [\vec{v} + \vec{A}_L(\tau_d)]} = \left| \int_{-\infty}^{\infty} \mathcal{E}_X(t - \tau_d) e^{i\Phi_G(\vec{v}_0, t)} e^{-iKt} dt \right|^2, \quad (5.198)$$

which now has similar form as the optical FROG trace.

5.6 Summary

The analytical solution of the Schrödinger equation obtained under the SFA is useful for performing semiquantitative calculations. It can be done much faster than solving the Schrödinger equation numerically. This is important when the radiations from many atoms or molecules are needed, such as the case when phase matching is considered, which is discussed in the next chapter.

Problems

- 5.1 The amplitude of the dipole moment is the same for two frequency components, ω and 2ω , what is the ratio between the electric field amplitudes of the dipole radiation? How about the intensity ratio?
- 5.2 Plot the potential energy curve and the ground state wave function of the hydrogen atom in atomic units.
- 5.3 Assuming that the ground state helium atom wave function can be described by Equation 5.10 what would be value of α ?
- 5.4 For hydrogen atom, plot the ground-state population $a(t)$ when it interacts with a 20 fs Gaussian laser pulses centered at 800 nm. The peak intensity is $1 \times 10^{14} \text{ W/cm}^2$.
- 5.5 Plot the dipole transition-matrix elements of 1s state for $\alpha = 0.5 \text{ a.u.}$ and $\alpha = 1.0 \text{ a.u.}$ on the same figure. Discuss the difference between them.
- 5.6 Find the conditions that the electric dipole approximation is valid.
- 5.7 Compare the wave function of a plane electron wave, $|\vec{v}\rangle = e^{-i\vec{v}\cdot\vec{r}}$, with the wave function of a plane light wave. $\vec{v}$ is the electron momentum.
- 5.8 An electron with 100 eV kinetic energy leaves the center of the potential well of the hydrogen atom due to absorption of XUV photon. Calculate the time it takes for the electron to reach a position where the potential energy of the nucleus is 10% of the ionization potential ($I_p = 13 \text{ eV}$).
- 5.9 Calculate the electric field amplitude when the laser intensity is 10^{12} W/cm^2 . Compare it with the field amplitude at 10^{14} W/cm^2 .

- 5.10** For the hydrogen atom, what is the Coulomb field strength when the electron is 1 nm away from the proton? Compare it with the laser field amplitude at 10^{14} W/cm².
- 5.11** In a field-free region, how long does it take for an electron with 10 eV kinetic energy to travel 1 nm?
- 5.12** Plot the four Gegenbauer polynomial terms given in Equation 5.98 for $\lambda = 2$.

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Phase Matching

The attosecond/high-harmonic field emitted from a single atom is too weak for applications. To produce sufficient XUV photons, many atoms are placed in the interaction region, as depicted in [Figure 6.1](#).

The density of the gas in the generation target is typically on the order of $10^{17}/\text{cm}^3$. The typical value of the volume with high enough driving laser intensity is a cylinder that is ~ 5 mm long and ~ 50 μm in diameter when lasers with millijoule pulse energy are used. Therefore, the total number of atoms that may contribute to the high-harmonic light is 10^{12} . If each atom emits one XUV photon, then 10^{12} high-harmonic photons are produced per laser shot, which corresponds to 3 μJ pulse energy at 20 eV photon energy. High-harmonic (the 11th–19th order) pulses with up to 10 μJ energy have indeed been generated experimentally by Midorikawa's group. The conversion efficiency from laser to the 15th harmonic was estimated to be 6.4×10^{-5} . In general, the conversion efficiency decreases with the harmonic order.

The energy of single isolated attosecond pulses generated with a few millijoule femtosecond lasers is on the order of 1 nJ. Thus the conversion efficiency is about 10^{-6} . This is rather low as compared to second harmonic (SH) generation in nonlinear crystals, which can reach more than 10%. It is also much lower than pump to laser pulse energy conversion in laser oscillators and amplifiers, which is typically higher than 10%. Finding schemes that can significantly increase the conversion efficiency is one of the biggest challenges in attosecond optics research.

There are several approaches for scaling up the XUV pulse energy so that it can be used to study attosecond nonlinear physics. Firstly, one has to find schemes so that fields emitted by atoms located at different positions in the propagation direction are in phase so that they add up constructively on the detector; such a condition is called phase matching. Improving the length over which the attosecond fields are phase matched, which is named coherent length, is one of the active areas of high-harmonic and attosecond pulse generation research.

Secondly, use femtosecond driving lasers with higher pulse energy, which would allow increasing the transverse size and thus the volume and the number of interacting atoms for a given coherent length. Chirped pulse amplifiers with petawatt power have already been developed. It is expected that much higher XUV attosecond flux be generated by implementing subcycle gating on such lasers.

To know whether the fields emitted by atoms are in phase, we need to know the phases of the XUV fields in each point of the generation target.

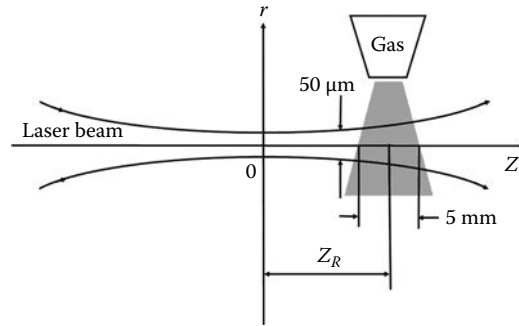


Figure 6.1 Gas target delivered from a nozzle located near the laser focus. Z_R is the Rayleigh range of the laser beam.

In principle, the phase can be found by solving the wave equations of the high-harmonic/attosecond field.

6.1 Wave-Propagation Equation

First, we will derive the wave equations for the combination of the driving laser field and the high-harmonic field. We choose to work with the harmonic field because each of them can be considered as a monochromatic field, which is much easier to deal with in the frequency domain than the attosecond pulses. Once we know the field of each harmonic (frequency), then we can work out the attosecond field by using the Fourier theory.

6.1.1 Wave Equations for the Total Fields

When a laser beam interacts with an atomic gas target, the medium can be considered as homogeneous, isotropic, and nonmagnetic ($\mu = \mu_0$, μ_0 is the magnetic permeability). Anisotropic media, such as aligned and oriented molecules, are not considered in this section. The propagation of the fundamental laser field and the generated high-harmonic field in the medium can be described by the Maxwell equations.

6.1.1.1 Maxwell Equations

In SI units for homogeneous, isotropic, and nonmagnetic media containing no free charges and no free currents, the equations are expressed by four basic laws:

Faraday's law of induction,

$$\nabla \times \vec{E}(\vec{r}, t) = -\frac{\partial \vec{B}(\vec{r}, t)}{\partial t}, \quad (6.1)$$

Ampère's circuital law,

$$\nabla \times \vec{B}(\vec{r}, t) = \mu_0 \frac{\partial \vec{D}(\vec{r}, t)}{\partial t}, \quad (6.2)$$

Gauss's law for electricity,

$$\nabla \cdot \vec{D}(\vec{r}, t) = 0, \text{ and} \quad (6.3)$$

Gauss's law for magnetism,

$$\nabla \cdot \vec{B}(\vec{r}, t) = 0, \quad (6.4)$$

in which $\vec{E}(\vec{r}, t)$ is the total electric field and $\vec{B}(\vec{r}, t)$ is the total magnetic induction, i.e., they include both the fundamental and high-harmonic field contributions.

The electric displacement is

$$\vec{D}(\vec{r}, t) = \epsilon_0 \vec{E}(\vec{r}, t) + \vec{P}(\vec{r}, t). \quad (6.5)$$

where $\vec{P}(\vec{r}, t)$ is the electric polarization, or dipole moments per unit volume. Also, ϵ_0 is the electric permittivity.

In principle, the high-harmonic field at the exit of the media can be found out by solving these four equations. However, Maxwell's equations are coupled differential equations for the electric and magnetic fields. It is extremely difficult to solve the equations directly.

6.1.1.2 Wave Equation for Electric Field

To find the solutions, the Maxwell equations are decoupled into two wave equations first; one for the electric field and the other for the magnetic field. The laser intensity used for high-harmonic generation is typically less than 10^{16} W/cm², which is much weaker than the so called relativistic intensity (10^{18} W/cm²). Under such circumstances, we only need to consider the electric fields.

From the first two equations, we have

$$\nabla \times [\nabla \times \vec{E}(\vec{r}, t)] = -\frac{\partial}{\partial t} [\nabla \times \vec{B}(\vec{r}, t)] = -\mu_0 \frac{\partial^2 \vec{D}(\vec{r}, t)}{\partial t^2}. \quad (6.6)$$

Using an identity from vector calculus, we arrive at

$$\nabla [\nabla \cdot \vec{E}(\vec{r}, t)] - \nabla^2 \vec{E}(\vec{r}, t) = -\mu_0 \frac{\partial^2 \vec{D}(\vec{r}, t)}{\partial t^2}. \quad (6.7)$$

We only consider the situation when $\nabla \cdot \vec{E}(\vec{r}, t)$ is zero (no net free charges in the gas target). In this case, we obtain the wave equation for the electric field

$$\nabla^2 \vec{E}(\vec{r}, t) - \frac{1}{c^2} \frac{\partial^2 \vec{E}(\vec{r}, t)}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}(\vec{r}, t)}{\partial t^2}, \quad (6.8)$$

where $c = 1/\sqrt{\epsilon_0 \mu_0}$ is the speed of light in vacuum.

6.1.2 Wave Equations for High-Harmonic Fields

The contribution of the nonlinearity in the wave equation can be made clearer by separating the linear polarization from the nonlinear polarization, $\vec{P}^{NL}(\vec{r}, t)$, i.e.,

$$\vec{P}(\vec{r}, t) = \vec{P}^{(1)}(\vec{r}, t) + \vec{P}^{NL}(\vec{r}, t). \quad (6.9)$$

For dispersionless media, the linear polarization is $\vec{P}^{(1)}(\vec{r}, t) = \epsilon_0 \chi^{(1)} \vec{E}(\vec{r}, t)$, where $\chi^{(1)}$ is the linear susceptibility, which is related to the gas density. Using the definition of the dielectric constant, $\epsilon^{(1)} = 1 + \chi^{(1)}$, the wave equation becomes

$$\nabla^2 \vec{E}(\vec{r}, t) - \frac{\epsilon^{(1)}}{c^2} \frac{\partial^2 \vec{E}(\vec{r}, t)}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}^{NL}(\vec{r}, t)}{\partial t^2}. \quad (6.10)$$

The right-hand side is the source of the high-harmonic radiation.

6.1.2.1 Monochromatic Driving Laser

Equation 6.10 is still difficult to solve because it is a four-dimension problem (three spatial variables and a time variable). It is a common practice to deal with this problem in the frequency domain. The wave equation in the frequency domain can be obtained by performing Fourier transforms to the wave equation in the time domain. Here, we use Fourier series approach, which can better express the high-harmonic process for the case when the fundamental laser is an infinitely long monochromatic light.

The electric field and the nonlinear polarization can be expanded as a Fourier series,

$$\vec{E}(\vec{r}, t) = \sum_{q=1}^{q_c} \vec{E}_q(\vec{r}, t), \quad (6.11)$$

$$\vec{P}^{NL}(\vec{r}, t) = \sum_{q=2}^{q_c} \vec{P}_q(\vec{r}, t), \quad (6.12)$$

where q_c is the cutoff harmonic order. The q th harmonic components are

$$\vec{E}_q(\vec{r}, t) = \vec{E}_q(\vec{r}) e^{i\omega_q t}, \quad (6.13)$$

$$\vec{P}_q^{NL}(\vec{r}, t) = \vec{P}_q(\vec{r}) e^{i\omega_q t}, \quad (6.14)$$

where $\omega_q = q\omega_1$ and ω_1 is the angular frequency of the fundamental laser.

The wave equations obtained for the phasor amplitudes of each harmonic component are

$$\nabla^2 \vec{E}_q(\vec{r}) + \frac{\epsilon^{(1)}(\omega_q) \omega_q^2}{c^2} \vec{E}_q(\vec{r}) = -\mu_0 \omega_q^2 \vec{P}_q(\vec{r}). \quad (6.15)$$

These are a set of coupled wave equations because $\vec{P}_q(\vec{r})$ is determined by the total electric field, $\vec{E}(\vec{r}, t)$, not the q th harmonic field. In reality, the conversion efficiency to high harmonics is rather low, and the nonlinear polarization is almost entirely produced by the fundamental field. In this case, one can solve the wave equation for the laser field first and then calculate the nonlinear polarization to feed the wave equations. In other words, the wave equations can be decoupled and solved one by one.

6.1.3 Linearly Polarized Fields

When the laser is linearly polarized, the fields and the polarizations are scalar quantities, which are much easier to deal with. The wave equations take the form

$$\nabla_T^2 E_q(\vec{r}) + \frac{\partial^2 E_q(\vec{r})}{\partial z^2} + k_q^2 E_q(\vec{r}) = -\mu_0 \omega_q^2 P_q(\vec{r}), \quad (6.16)$$

where $k_q = \frac{\omega_q}{c} \sqrt{\epsilon^{(1)}(\omega_q)}$ is the propagation constant of the q th harmonic field. Here, the Laplacian operator is split into the transverse part, ∇_T^2 , and the part in the propagation direction, $\frac{\partial^2}{\partial z^2}$.

6.1.3.1 Paraxial Approximation

The equation can be further simplified by making the paraxial approximation. For that, we represent the field and polarization components as

$$E_q(\vec{r}) = \tilde{A}_q(\vec{r}) e^{-ik_q z}, \quad (6.17)$$

$$P_q(\vec{r}) = \tilde{P}_q(\vec{r}) e^{-iqk_1 z}, \quad (6.18)$$

where k_1 is the propagation constant of the driving laser. We insert them into the wave equation and drop the $\frac{\partial^2 \tilde{A}_q}{\partial z^2}$ term because its module is much smaller than the other terms. Finally, the wave equation for the complex amplitudes is

$$\nabla_T^2 \tilde{A}_q(\vec{r}) - 2ik_q \frac{\partial}{\partial z} \tilde{A}_q(\vec{r}) = -\mu_0 \omega_q^2 \tilde{P}_q(\vec{r}) e^{i(k_q - qk_1)z}. \quad (6.19)$$

The solution of this equation, $\tilde{A}_q(\vec{r})$, gives the phase and amplitude of the harmonic field at point $\vec{r}$. Various mechanisms that affect the coherent build up of a particular harmonics can be understood by using this equation.

6.2 Phase Matching for Plane Waves

The wave equation we derived is valid for monochromatic, paraxial waves. Several phase-matching mechanisms can be revealed more clearly for plane waves. High harmonics can be generated in gas filled hollow-core fiber, the wavefront of the laser and harmonic beam is considered as plane waves. When petawatt (10^{15} W) lasers are used to generate attosecond pulses in xenon gas, it is possible to use collimated, not focus beam because the ionization saturation intensity is 10^{13} W/cm².

For plane waves, $\nabla_T^2 \tilde{A}_q(\vec{r}) = 0$, thus, the wave equation becomes

$$2ik_q \frac{\partial}{\partial z} \tilde{A}_q(z) = \mu_0 \omega_q^2 \tilde{P}_q(\tilde{A}_1) e^{i(k_q - qk_1)z}. \quad (6.20)$$

Here, we write the propagation constant as complex form to include the absorption, i.e.,

$$\tilde{k}_q = \frac{\omega_q}{c} \sqrt{\epsilon^{(1)}(\omega_q)} = \frac{\omega_q}{c} [n_R(\omega_q) - in_I(\omega_q)],$$

where n_R and n_I are the real and the imaginary parts of the linear index of refraction, respectively.

The variation of the q th harmonic field amplitude in the propagation direction can be calculated by

$$\tilde{A}_q(z) = -i \frac{\mu_0 \omega_q^2}{2k_q} \int_0^z \tilde{P}_q(A_1) e^{i\Delta k_q z} e^{-\alpha_q z} dz, \text{ where } \Delta k_q = \frac{\omega_q}{c} n_R(\omega_1) - qk_1,$$

The absorption coefficient is $\alpha_q = \frac{\omega_q}{c} n_I(\omega_q)$.

Since the conversion efficiency from the laser power to the harmonic power is very low, it is safe to assume that for the fundamental laser field, $2ik_1 \frac{\partial}{\partial z} \tilde{A}_1(z) = 0$, thus $\tilde{A}_1$ and its phase do not change with z . In this case, we can set the phase of $\tilde{A}_1$ to zero, and replace $\tilde{A}_1$ with a real quantity A_1 . The variation of the q th harmonic field amplitude in the propagation direction can be calculated by

$$\tilde{A}_q(z) = -i \frac{\mu_0 \omega_q^2}{2k_q} \tilde{P}_q(A_1) \int_0^z e^{i\Delta k_q z} e^{-\alpha_q z} dz.$$

We first discuss some special phase-matching conditions.

6.2.1 Perfect Phase Matching in Lossless Media

For high harmonics generated below the ionization threshold of the target gas, the absorption due to linear photoionization is zero. For example, helium is not ionized by light with photon energy below 24.6 eV.

When absorption and other losses are zero, $\alpha_q = 0$. The propagation constant becomes $\tilde{k}_q = k_q = \frac{\omega_q}{c} n_R(\omega_q)$. Considering a general situation where Δk_q may vary with z , the harmonic-field amplitude can be expressed by a simple expression

$$\tilde{A}_q(z) = -i \frac{\mu_0 \omega_q^2}{2k_q} \tilde{P}_q(A_1) \int_0^z e^{i\Delta k_q(z)z} dz. \quad (6.21)$$

The perfect phase-matching condition is

$$\frac{d}{dz} [\Delta k_q(z)z] = \frac{d}{dz} [\varphi_{P,q}(z) - \varphi_q(z)] = 0, \quad (6.22)$$

where $\varphi_{P,q}(z) = \frac{\omega_q}{c} n_R(\omega_q, z)z$ is the phase of the polarization responsible for generating the q th harmonic by the dipoles at position z . $\varphi_q(z) = \frac{\omega_q}{c} z$ is the phase of the harmonic field generated in front of z when it arrives at z . The phase velocities of the polarization and the harmonic field are $\frac{\omega_q}{\frac{d}{dz} \varphi_{P,q}(z)}$ and $\frac{\omega_q}{\frac{d}{dz} \varphi_q(z)}$.

Obviously, the two velocities are the same when $\frac{d}{dz} [\varphi_{P,q}(z) - \varphi_q(z)] = 0$. Then, the newly generated harmonic wave by the nonlinear polarization at position z is always in phase with the one

produced in front of z . The constructive superposition leads to the linear increase of the wave amplitude, i.e.,

$$|\tilde{A}_q(z)| = -i \frac{\mu_0 \omega_q^2}{2k_q} |\tilde{P}_q(A_1)| z.$$

In some cases, Δk_q does not change in the propagation direction. In this special case, the perfect phase-matching condition becomes

$$\Delta k_q = 0. \quad (6.23)$$

The phase velocity of q th harmonic wave is $v_q = \omega_q/k_q = q\omega_1/k_q$. While for the driving laser, the phase velocity is $v_1 = \omega_0/k_1 = q\omega_1/(qk_1)$. If the perfect phase-matching condition is fulfilled, $\Delta k_q = k_q - qk_1 = 0$, the two phase velocities are the same.

By definition, the nonlinear polarization is the number of dipole moments per unit volume, $\tilde{P}_q(A_1) = \tilde{d}_q(A_1)N_a$. Here $\tilde{d}_q(A_1)$ is the dipole of a single atom and N_a is the atomic density in the gas target. We can rewrite the wave amplitude as

$$\tilde{A}_q(N_a z) = -i \frac{\mu_0 \omega_q^2}{2k_q} \tilde{d}_q(A_1) N_a z = -i \frac{k_q}{2\epsilon_0} \tilde{d}_q(A_1) N_a z. \quad (6.24)$$

In high-harmonic generation, ionization of gas is unavoidable, which turns the medium into a mixture of plasma and neutral atoms. The atomic density is $N_a = N(1 - p)$. Here N is the field-free gas density and p is the ionization probability.

The intensity of the q th harmonics is

$$I_q(NL) = \frac{c\epsilon_0}{2} \tilde{A}_q^* \tilde{A}_q = \frac{ck_q^2}{8\epsilon_0} |\tilde{d}_q(A_1)|^2 (1 - p)^2 (NL)^2. \quad (6.25)$$

where L is the length of the gas target. Evidently, the intensity increases quadratically with the density-length product. Unlike conventional nonlinear optics in solid media where the density is fixed, the gas target density is a controllable parameter in high-harmonic generation. Experimentally, it is much easier to change the gas density than varying the target length. The strong field approximation gives $|\tilde{d}_q(A_1)|^2 \approx 10^{-10}$ atomic unit, which yields 10^7 W/cm² for $N = 10^8$ /cm³ and $L = 1$ cm. One needs to keep in mind that the value of dipole moment calculated with the strong field approximation is not accurate.

Gas pressure can be measured directly in experiments. Since gas density is proportional to pressure, the harmonic signal can be expressed as a function of pressure-length product $I_q(PL) \propto (PL)^2$, as depicted in Figure 6.2.

6.2.1.1 Plasma Dispersion

The index of refraction of plasma is

$$n_p(\omega_q) = \sqrt{1 - \left(\frac{\omega_p}{\omega_q}\right)^2}, \quad (6.26)$$

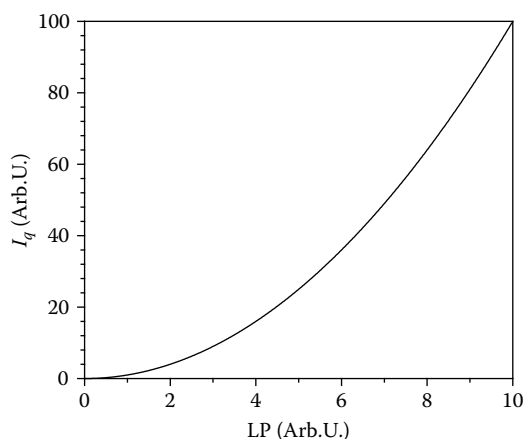


Figure 6.2 Harmonic signals in the perfect phase-matching condition. Absorption is zero.

in which ω_p is the plasma angular frequency such that

$$\omega_p^2 = \frac{N_e e^2}{\epsilon_0 m_e}, \quad (6.27)$$

where m_e is the mass of an electron and $N_e = Np$ is the free electron density, which is less than $10^{18}/\text{cm}^3$ in most experiments. The plasma frequency is much lower than the laser frequency and the high-harmonic frequency, which implies

$$n_p(\omega_q) \approx 1 - \frac{e^2}{2\epsilon_0 m_e} \frac{N_e}{\omega_q^2}. \quad (6.28)$$

For harmonic order $q \gg 1$, $n_p(\omega_q) \approx 1$ and the phase velocity $v_q \approx c$. For the driving laser, $v_1 > c$.

The index of refraction of the neutral atoms is

$$n_a(\omega_q) \approx 1 + \frac{e^2}{2\epsilon_0 m_e} \frac{N_a}{\omega_r^2 - \omega_q^2} = 1 + \Delta n_a, \quad (6.29)$$

where ω_r is the resonance frequency of the atom, which is determined by its excitation energy between the ground state and the first excitation energy level, I_e . For harmonics with photon energy much higher than I_e , $n_a(\omega_q) \approx 1$. For the driving laser, the values of the index of refraction for noble gases in one atmosphere pressure are listed in Table 6.1. The

TABLE 6.1 Plasma Parameters of Noble Gases

Gas	He	Ne	Ar	Kr	Xe
I_e (eV)	19.8	16.6	11.6		8.39
$\Delta n_a \times 10^4$ (1 atm)	0.36	0.67	2.8	4.27	7.02
I'_e (eV)	21.9		8		
p	5×10^{-3}	9×10^{-3}	0.038	0.054	0.086

resonant frequency can be treated as a fitting parameter to match the measured index of refraction.

Finally, the total index of the refraction of the gas medium for the driving laser is

$$\begin{aligned} n_R(\omega_1) &\approx 1 + \frac{e^2}{2\epsilon_0 m_e} \left(\frac{N_a}{\omega_r^2 - \omega_1^2} - \frac{N_e}{\omega_1^2} \right) \\ &= 1 + \frac{e^2 N}{2\epsilon_0 m_e} \left(\frac{1-p}{\omega_r^2 - \omega_1^2} - \frac{p}{\omega_1^2} \right). \end{aligned} \quad (6.30)$$

For high-order harmonics, $n_R(\omega_q) \approx 1$, to fulfill the phase-matching condition, $n_R(\omega_1) = n_R(\omega_q)$, one can control the ionization probability so that $\frac{1-p}{\omega_r^2 - \omega_1^2} - \frac{p}{\omega_1^2} = 0$. This can be done by properly setting the laser inten-

sity so that the ionization probability becomes $p = \left(\frac{\hbar\omega_1}{\hbar\omega_r} \right)^2 = \left(\frac{\hbar\omega_1}{I_e'} \right)^2$.

$I_e' = \hbar\omega_r$ is the effective excitation energy between the ground state and the first excited state calculated from the resonance frequency. Alternatively, the ionization probability can be calculated from $p = 1 / \left[1 + \frac{N_0 \omega_1^2}{\Delta n_a(\omega_1)} \frac{2\epsilon_0 m_e}{e^2} \right]$, where $N_0 = 2.5 \times 10^{19}$ atoms cm^{-3} is the gas density of a standard gas at room temperature and atmosphere pressure.

The values for a Ti:Sapphire laser centered at 800 nm are given in Table 6.1 for several noble gases. It shows that the neutral gas can only compensate the plasma induced dephasing if the ionization probability is less than 10%. For longer wavelength driving lasers, such as 1600 nm, very high harmonic orders can be reached with low ionization probability of helium; thus, it is possible to phase match harmonic radiations in the soft x-ray wavelength range.

For xenon gas, the phase-matching ionization probability is 8.6%. As Figure 4.14 indicates, a 40 fs laser pulse 8×10^{13} W/cm² intensity would lead to such an ionization probability. The corresponding high-harmonic cutoff photon energy is 27 eV when Ti:Sapphire laser is used.

6.2.1.2 Pressure (Plasma) Gradient Gas Target

When the ionization probability is high, the phase matching can be achieved by varying the gas density in the propagation direction. The index of refraction of the plasma for the driving laser is $n_R(\omega_1, z) \approx$

$$1 - \frac{e^2}{2\epsilon_0 m_e} \frac{N(z)p}{\omega_1^2}. \text{ Therefore,}$$

$$\Delta k_q(z) = \frac{\omega_q}{c} n_R(\omega_1) - qk_1 = \frac{\omega_q}{c} n_R(\omega_1) - \frac{\omega_q}{c} = \frac{\omega_q}{c} \frac{e^2}{2\epsilon_0 m_e} \frac{N(z)p}{\omega_1^2}.$$

We can design such a gas target where $N(z) = N_0 \frac{1}{z}$, so that $\Delta k_q z = \frac{\omega_q}{c} \frac{e^2}{2\epsilon_0 m_e} \frac{N_0 p}{\omega_1^2}$, which is independent of z . Then

$$\tilde{A}_q(z) = i \frac{\mu_0 \omega_q^2}{2\tilde{k}_q} \tilde{P}_q(A_1) e^{\frac{\omega_q}{c} \frac{e^2 N_0 p}{2\epsilon_0 m_e \omega_1^2} (1 - e^{-\alpha_q z})}.$$

The phase matching is perfect for all harmonics. In this case, the phase velocities of the polarization

$$\begin{aligned} \frac{\omega_q}{\frac{d}{dz} \varphi_{P,q}(z)} &= \frac{\omega_q}{\frac{d}{dz} \left[\frac{\omega_q}{c} \left(1 - \frac{e^2 N(z)p}{2\epsilon_0 m_e \omega_1^2} \right) z \right]} \\ &= \frac{c}{\frac{d}{dz} \left[\left(1 - \frac{e^2 N_0 p}{2\epsilon_0 m_e \omega_1^2} \right) z \right]} = c. \end{aligned} \quad (6.31)$$

Interestingly, in such an ionized target, the phase velocity of the polarization wave is the same as the speed of light in vacuum.

When PW (10^{15} W) level lasers are used for generating attosecond pulses, the laser beam size on the target is about 1 cm. Such a beam can be considered as collimated in the interaction region. Phase matching with the pressure-gradient gas target is feasible. The gas density decreases in the propagation direction. Experimentally, an array of targets (gas cell or jet) in the propagation direction can be used. By individually adjusting the pressure of each target, it is possible to form a density distribution close to $N(z) = \frac{N_0}{Z}$.

6.2.2 Effect of Absorption

Under the perfect phase-matching condition, when the absorption of gas to the harmonics is taken into account, the amplitude is

$$\begin{aligned} \tilde{A}_q(z) &= -i \frac{\mu_0 \omega_q^2}{2\tilde{k}_q} \tilde{P}_q(A_1) \int_0^z e^{-\alpha_q z} dz \\ &= i \frac{\mu_0 \omega_q^2}{\tilde{k}_q} \tilde{P}_q(A_1) L_a \left(1 - e^{-z/(2L_a)} \right). \end{aligned} \quad (6.32)$$

It reaches an asymptotic maximum value when the propagation distance is much longer than the absorption length, $L_a = \frac{1}{2\alpha_q} = \frac{1}{N\sigma_q}$. Here, σ_q is the absorption cross section. The absorption length is defined as the distance over which the transmission of light power is $\frac{1}{e} = 0.368$.

6.2.2.1 Absorption Limit

The maximum value of the harmonic intensity is called the absorption limit, which is given by

$$I_{max} = \frac{ck_q^2}{8\epsilon_0} |\tilde{d}_q(A_1)|^2 (1-p)^2 N^2 L_a^2 = \frac{ck_q^2}{8\epsilon_0} |\tilde{d}_q(A_1)|^2 (1-p)^2 \frac{1}{\sigma_q^2}. \quad (6.33)$$

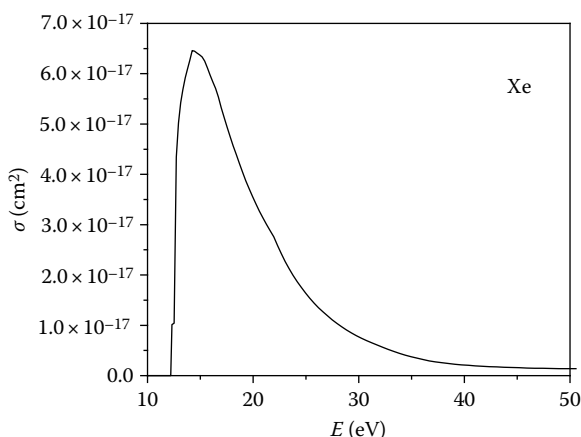


Figure 6.3 Photoionization cross section of xenon.

This value does not depend on the target gas density. Rather, it is determined by the atomic species, laser intensity, and the harmonic photon energy.

The photoionization cross section $\sigma_q = 2r_e \lambda_q f_2(\lambda_q)$, as discussed in Chapter 1. f_2 is the imaginary part of the atomic scattering factor. r_e is the classical electron radius. The cross section of xenon is shown in Figure 6.3.

It is even more meaningful to introduce a parameter called absorption length–density product, $L_a N = \frac{1}{\sigma_q}$, or absorption length–pressure product,

$L_a P = L_a \frac{N}{N_0} = \frac{1}{N_0 \sigma_q}$, beyond which the harmonic intensity does not increase significantly. At room temperature, $N_0 \approx 3.3 \times 10^{16}$ atoms/(cm³ torr). Figure 6.4 shows the absorption length–pressure product for (a) argon and (b) neon gas in the 25–200 eV photon energy range. The

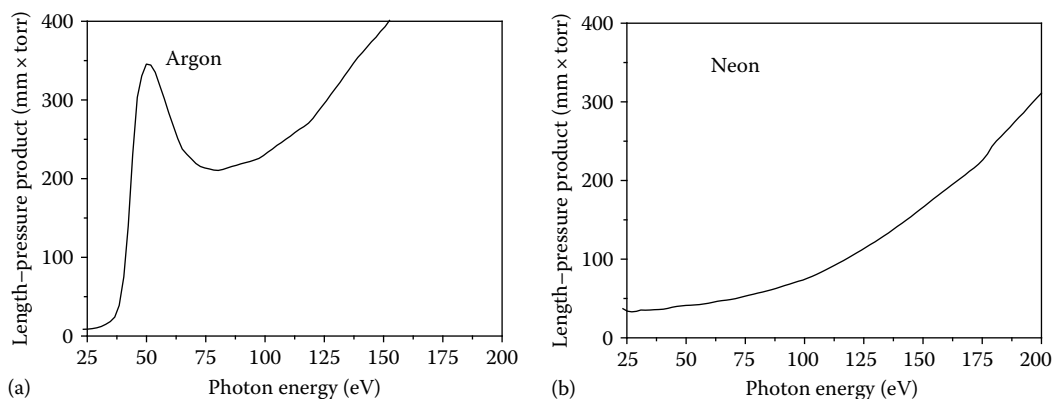


Figure 6.4 Absorption length–pressure product: (a) argon; (b) neon.

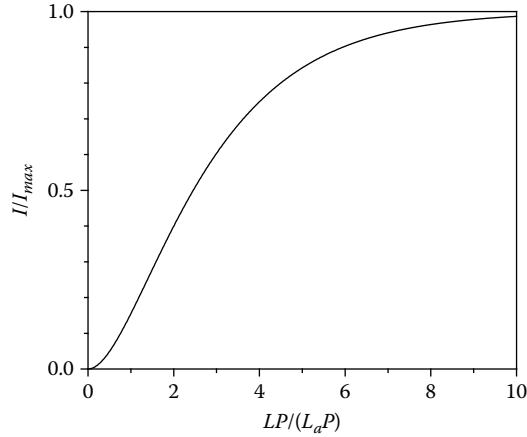


Figure 6.5 Effects of the absorption on high-harmonic signal when the phase matching is perfect.

product is only determined by the type of the atom and the harmonic order (wavelength). It does depend on any other target parameters such as the size and pressure.

The dependence of the harmonic intensity on the length–pressure product is

$$I(LP) = I_{max} \left(1 + e^{-\frac{LP}{L_a P}} - 2e^{-\frac{LP}{2L_a P}} \right), \quad (6.34)$$

as shown in Figure 6.5.

Since $\frac{I(10LP_a)}{I_{max}} = 0.987$, it is not necessary to use length–pressure product more than ten times of the absorption length–density product. Apparently the absorption length–pressure product depends on the photon energy. For xenon gas, $L_a P = 9$ mm torr at 20 eV. To generate attosecond pulses at centered at 20 eV photo energy, for 1 mm long gas target, the highest pressure is 90 torr. For argon gas, $L_a P = 8$ mm torr at 20 eV, which is similar to the Xe case.

For $LP \ll LP_a$, $I(LP) \approx \frac{1}{4} I_{max} \left(\frac{LP}{LP_a} \right)^2$, the harmonic signal increases quadratically with the density–length product, like the lossless case.

6.2.3 Maker Fringes

In lossless media, when $\Delta k_q \neq 0$ and Δk_q does not change with z , the harmonic amplitude is

$$\begin{aligned} \tilde{A}_q(z) &= -i \frac{\mu_o \omega_q^2}{2k_q} \tilde{P}_q(A_1) \int_0^z e^{i\Delta k_q z} dz \\ &= -i \frac{\mu_o \omega_q^2}{2k_q} \tilde{P}_q(A_1) z \left(\frac{e^{i\Delta k_q z} - 1}{i\Delta k_q} \right), \end{aligned} \quad (6.35)$$

$$\text{where } \Delta k_q = \frac{\omega_q}{c} \frac{e^2}{2\epsilon_0 m_e} \left(\frac{p}{\omega_1^2} - \frac{1-p}{\omega_r^2 - \omega_1^2} \right) N.$$

The corresponding intensity is

$$I_q(NL) = \frac{ck_q^2}{8\epsilon_0} \tilde{d}_q(A_1) \tilde{d}_q^*(A_1) (1-p)^2 (NL)^2 \sin^2\left(\frac{\Delta k_q L}{2}\right). \quad (6.36)$$

The dependence of the normalized harmonic intensity on the density–length product called the Maker Fringe in conventional nonlinear optics. The dephasing effects show up when the medium length is longer than the coherent length, $L_c = \pi/\Delta k_q$. Since the Maker Fringe is proportional to the gas density, we can introduce a quantity called the coherent length–pressure product,

$$(LP)_c = \frac{\pi}{\frac{\omega_q}{c} \frac{e^2}{2\epsilon_0 m_e} \left(\frac{p}{\omega_0^2} - \frac{1-p}{\omega_r^2 - \omega_1^2}\right) N_0}.$$

The harmonic intensity can then be expressed as

$$I_q(LP) = \frac{ck_q^2}{8\epsilon_0} |\tilde{d}_q(A_1)|^2 (1-p)^2 N_0^2 (LP)^2 \sin^2\left(\frac{\pi}{2} \frac{LP}{(LP)_c}\right). \quad (6.37)$$

Maker fringes were observed in SH generation crystals long time ago. When one looks at the scattered SH light from the side of the crystal, one sees the intensity changes periodically along the propagation direction. In high-harmonic generation, the detected harmonic signal should oscillate with the pressure while keeping the target length fixed as the sinc^2 function in Equation 6.37 indicates.

6.2.4 Rule of Thumb for Optimizing XUV Photon Flux

When both phase mismatch and absorption are considered at the same time, assuming Δk_q is independent of z .

$$\begin{aligned} \tilde{A}_q(z) &= -i \frac{\mu_0 \omega_q^2}{2\tilde{k}_q} \tilde{P}_q(A_1) \int_0^z e^{i\Delta k_q z} e^{-\alpha_q z} dz \\ &= -i \frac{\mu_0 \omega_q^2}{2\tilde{k}_q} \tilde{P}_q(A_1) \frac{e^{i\Delta k_q z} e^{-\alpha_q z} - 1}{i\Delta k_q - \alpha_q}. \end{aligned} \quad (6.38)$$

The harmonic intensity is a function of the coherent length, the absorption length, and the medium length. Since

$$\begin{aligned} &\left(e^{i\pi L/L_c} e^{-L/(2L_a)} - 1\right) \left(e^{-i\pi L/L_c} e^{-L/(2L_a)} - 1\right) \\ &= 1 + e^{-L/L_a} - 2 \cos(\pi L/L_c) e^{-L/(2L_a)}, \end{aligned} \quad (6.39)$$

and

$$\frac{1}{i\frac{\pi}{L_c} - \frac{1}{2L_a}} \frac{1}{-i\frac{\pi}{L_c} - \frac{1}{2L_a}} = \frac{4L_a^2}{1 + 4\pi^2 \left(\frac{L_a}{L_c}\right)^2},$$

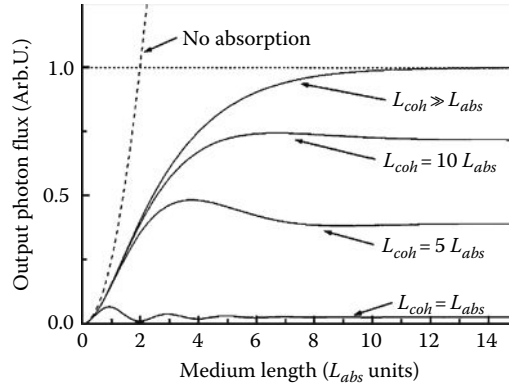


Figure 6.6 Dependence of output on gas parameters. The quantity L_{coh} and L_{abs} in the figure are the coherent and absorption lengths, respectively. (Reprinted with permission from E. Constant, D. Garzella, P. Breger, E. Mevel, Ch. Dorrer, C. Le Blanc, F. Salin, and P. Agostini, *Phys. Rev. Lett.*, 82, 1668, 1999. Copyright 1999 by the American Physical Society.)

we have

$$I_q(LP) = I'_{max} \left[1 + e^{-\frac{LP}{L_a P}} - 2 \cos\left(\frac{\pi LP}{(LP)_c}\right) e^{-\frac{LP}{L_a P}} \right], \quad (6.40)$$

and

$$I'_{max} = I_{max} \frac{1}{1 + 4\pi^2 \left(\frac{L_a P}{(LP)_c}\right)^2}. \quad (6.41)$$

For a given target gas density, the absorption length is determined by the high-harmonic photon energy. The dependence of the XUV photon flux on the medium length for several coherent lengths is shown Figure 6.6. It is clear from the figure that the medium length should be set to $L \approx L_c$. Apparently, the coherent length should be much larger than the absorption length. As a rule of thumb, to generate more than 50% of the maximum photon flux, the phase matching should be improved so that $L_c > 5L_a$.

In experiments, if the pressure between the target and the detector is P' , and the distance is L' , then the measurement of the harmonic intensity becomes

$$I_q(LP) = I'_{max} \left[1 + e^{-\frac{LP}{L_a P}} - 2 \cos\left(\frac{\pi LP}{(LP)_c}\right) e^{-\frac{LP}{L_a P}} \right] e^{-\frac{L'}{L'_a(P')}}, \quad (6.42)$$

where L'_a is the absorption length determined by the pressure P' .

6.2.5 Effects of Intensity Distribution in the Propagation Direction

For uniform neon and helium gas targets, the ionization probability at which the phase-matching conditions can be fulfilled by slowing down

the phase velocity with neutral gas is very low. Another parameter can be used to realize phase matching for highly ionized gases. According to the theory under the strong field approximation, the dipole moment can be expressed as

$$\tilde{d}_q(I_1) = |\tilde{d}_q(I_1)|e^{i\varphi_{in}(I_1)}, \quad (6.43)$$

where

I_1 is the intensity of the excitation laser intensity

φ_{in} is the intrinsic dipole phase that depends on the laser intensity

It is worthwhile to point out that for conventional nonlinear optics, the dipole phase is not a function of the laser intensity. If the laser intensity can be controlled in the propagation direction, then the phase variation of the polarization can compensate for the phase change due to the ionization.

In a lossless medium, when the variation of the laser intensity in the propagation direction is taken into account, the complex amplitude of the q th harmonic can be written as

$$\begin{aligned} \tilde{A}_q(z) &= -i \frac{\mu_0 \omega_q^2}{2\tilde{k}_q} \int_0^z \tilde{P}_q[I_1(z)] e^{i\Delta k_q z} dz \\ &= -i \frac{\mu_0 \omega_q^2}{2\tilde{k}_q} N \int_0^z [1 - p(z)] \tilde{d}_q[I_1(z)] e^{i\Delta k_q z} dz \\ &= -i \frac{\mu_0 \omega_q^2}{2\tilde{k}_q} N \int_0^z [1 - p(z)] |\tilde{d}_q[I_1(z)]| e^{i\varphi_{in}[I_1(z)]} e^{i\Delta k_q z} dz. \end{aligned} \quad (6.44)$$

Perfect phase matching happens when the total phase, $\varphi_{in}[I_1(z)] + \Delta k_q z$, does not depend on z . In that case $\varphi_{in}[I_1(z)] + \Delta k_q z$ is a constant, which can be taken out of the integral, which maximize $|\tilde{A}_q(z)|^2$.

The intrinsic phase can then be expressed as

$$\varphi_{in}(I_1) = \alpha I_1, \quad (6.45)$$

in which the proportional coefficient is different for the two quantum trajectories. For the short trajectory, $\alpha \approx 1 \times 10^{-14} \text{ cm}^2/\text{W}$ while for the long trajectory, $\alpha \approx 25 \times 10^{-14} \text{ cm}^2/\text{W}$. In highly ionized gases, the index of refraction of the gas for the fundamental laser is predominately

from plasma contribution $n_p(\omega_1) \approx 1 - \frac{e^2}{2\epsilon_0 m_e} \frac{N_e}{\omega_1^2}$. Thus, $\Delta k_q = \frac{q}{c} \frac{e^2}{2\epsilon_0 m_e} \frac{p}{\omega_1} N$.

For perfect phase matching of harmonic order q , the laser intensity is controlled so that it decreases linearly with z , i.e., $I_1(z) = I_1(0)(1 - \beta z)$, and $\alpha \beta I_1(0) = \Delta k_q$. In other words,

$$\beta(q) = \frac{q}{c} \frac{e^2}{2\epsilon_0 m_e} \frac{pN}{\omega_1} \frac{1}{\alpha I_1(0)}. \quad (6.46)$$

In this case, the phase velocities of the polarization

$$\begin{aligned} \frac{\omega_q}{\frac{d}{dz} \varphi_{P,q}(z)} &= \frac{\omega_q}{\frac{d}{dz} \left[\alpha I_1(z) + \frac{\omega q}{c} \left(1 - \frac{e^2 N p}{2 \epsilon_0 m_e \omega_1^2} \right) z \right]} \\ &= \frac{\omega_q}{\frac{d}{dz} \left[\alpha I_1(0)(1 - \beta(q)z) + \frac{\omega q}{c} \left(1 - \frac{e^2 N p}{2 \epsilon_0 m_e \omega_1^2} \right) z \right]} = c. \quad (6.47) \end{aligned}$$

Here, we neglected the variation of the ionization probability with z .

6.2.5.1 Quasiphase Matching

It is possible to change the laser intensity over a distance comparable to the laser wavelength by adding the laser beam with a weaker counter propagating beam. The superposition of the two waves creates a partial standing wave along the z direction, which changes the dipole phase periodically. When the phase modulation and the coherent length–pressure product are controlled properly, the harmonics from many zones can add constructively, which is very similar to the quasiphase matching in conventional SH generation in PPLN crystals.

6.3 Phase Matching for Gaussian Beams

The laser intensity for generating high-order harmonics and attosecond pulses is on the order of 10^{14} W/cm². Such intensity has been achieved by focusing laser beams onto the gas target. Thus in all experiments, the transverse size of the laser beam is always finite. For such beams, the three-dimensional (3D) wave equations should be used. For beams with small divergence angles, under the Paraxial Approximation and assuming the laser light is monochromatic, the equation for the fundamental wave is

$$\nabla_T^2 \tilde{A}_1(\vec{r}) - 2ik_1 \frac{\partial}{\partial z} \tilde{A}_1(\vec{r}) = 0. \quad (6.48)$$

For convenience, we use the Gaussian beam to describe the fundamental laser propagation and neglect the plasma defocusing. The electric field is expressed by

$$\tilde{A}_1(r, z) = A_0 \frac{w_0}{w(z)} e^{-\frac{r^2}{w(z)^2}} e^{-i\frac{z^2}{2R(z)}} e^{i\psi(z)}, \quad (6.49)$$

where

A_0 is the amplitude of the field at the laser focus and on axis

w_0 and $w(z)$ are the radius of the beam at the focus and at position z , respectively

$R(z)$ is the radius of curvature of the wave front

$\psi(z)$ is the Gouy phase, as introduced in Chapter 2

By introducing the Rayleigh length

$$z_R = \frac{\pi w_0^2}{\lambda_1}, \quad (6.50)$$

we have

$$w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_R}\right)^2}, \quad (6.51)$$

$$R(z) = z + \frac{z_R^2}{z}, \quad (6.52)$$

$$\psi(z) = \tan^{-1} \left(\frac{z}{z_R} \right). \quad (6.53)$$

When the laser beam is Gaussian, the harmonic field should have axial symmetry. The wave equation for the q th harmonic can be rewritten as

$$\frac{\partial}{\partial z} \tilde{A}_q(r, z) = -\frac{i}{2k_q} \nabla_T^2 \tilde{A}_q(r, z) - \frac{i\omega_q^2}{2k_q} \tilde{P}_q(r, z) e^{i\Delta k_q z}, \quad (6.54)$$

or

$$\frac{\partial}{\partial z} \tilde{A}_q(r, z) = -\frac{i}{2k_q} \nabla_T^2 \tilde{A}_q(r, z) - \frac{i\mu_0 \omega_q^2}{2k_q} N(1-p) \tilde{d}_q [\tilde{A}_1(r, z)] e^{i\Delta k_q z}. \quad (6.55)$$

The dipole moment is

$$\tilde{d}_q [\tilde{A}_1(r, z)] = |\tilde{d}_q [\tilde{A}_1(r, z)]| e^{i\varphi_m [I_1(r, z)]} e^{-iqk_1 \frac{r^2}{2R(z)}} e^{iq\psi(z)}. \quad (6.56)$$

6.3.1 On-Axis Phase Matching without Plasma and Gas Dispersion

For simplicity, we discuss the atoms placed on the axis. The phase difference between the polarization wave and the harmonic field is

$$\varphi_{P,q}(z) - \varphi_q(z) = \varphi_{in}[I_1(z)] + q\psi(z) + \Delta k_q(z)z. \quad (6.57)$$

For the Gaussian beam

$$\varphi_{P,q}(z) - \varphi_q(z) = \frac{\alpha I_0}{1 + \left(\frac{z}{z_R}\right)^2} + q \tan^{-1} \left(\frac{z}{z_R} \right) + \Delta k_q(z)z, \quad (6.58)$$

where

$$\Delta k_q(z) = \frac{\omega_q}{c} \frac{e^2}{2\epsilon_0 m_e} \left(\frac{p(z)}{\omega_1^2} - \frac{1-p(z)}{\omega_r^2 - \omega_1^2} \right) N.$$

The Gouy phase increases the phase velocity of the polarization wave since $\frac{d}{dz} \left[q \tan^{-1} \left(\frac{z}{z_R} \right) \right] > 0$.

The degree of phase matching can be evaluated by the magnitude of $\frac{d}{dz} [\varphi_{P,q}(z) - \varphi_q(z)]$. When the phase mismatch caused by the plasma

and neutral atom dispersion is ignored, i.e., $\Delta k_q(z) = 0$, the derivative of the phase is

$$\frac{d}{dz} [\varphi_{P,q}(z) - \varphi_q(z)] = \frac{2\alpha I_0 z}{\left[1 + \left(\frac{z}{z_R}\right)^2\right]^2 z_R^2} + q \frac{\left[1 + \left(\frac{z}{z_R}\right)^2\right] z_R}{\left[1 + \left(\frac{z}{z_R}\right)^2\right]^2 z_R^2}, \quad (6.59)$$

which equals zero when

$$\left(\frac{z}{z_R}\right)^2 - \frac{2\alpha I_0}{q} \left(\frac{z}{z_R}\right) + 1 = 0. \quad (6.60)$$

The solution is

$$\frac{z}{z_R} = \frac{\alpha I_0}{q} \pm \sqrt{\left(\frac{\alpha I_0}{q}\right)^2 - 1}. \quad (6.61)$$

It gives the location of gas target for phase matching.

Equation 6.61 requires $I_0 > q/\alpha$ for the solution to be real. For the short trajectory, $\alpha \approx 1 \times 10^{-14} \text{ cm}^2/\text{W}$. Thus, the peak intensity must be higher than $q \times 10^{14} \text{ W/cm}^2$ for a working solution. For harmonic order $q > 20$, it requires the intensity at the laser focus $I_0 > 2 \times 10^{15} \text{ W/cm}^2$. This intensity is applicable to neon and helium gas, but is too high for Xe, Kr, and Ar atoms because they will be completely ionized at this laser intensity.

For the long trajectory, $\alpha \approx 25 \times 10^{-14} \text{ cm}^2/\text{W}$, the peak intensity must be higher than $q \times 10^{13} \text{ W/cm}^2$. For harmonic order $q > 20$, it requires the intensity at the laser focus $I_0 > 2 \times 10^{14} \text{ W/cm}^2$. This intensity is applicable to all noble gases. At the same laser intensity, the order at which the harmonics is phase matching is different for the two trajectories, which provides a scheme for selecting the contribution of a particular quantum trajectory.

When the intensity is chosen to the lowest value for perfect phase matching, i.e., $I_1 = q/\alpha$, we have $z = z_R$. It means that the target should be located one Rayleigh range away after the laser focus. When a finite gas target is located after the laser focus, the intrinsic dipole phase decreases with z , whereas the Gouy phase increases with z , as shown in Figure 6.7. These phases tend to cancel each other on the contributions to the total polarization phase, which results in good phasing matching.

When a thin target is placed at the laser focus, $z = 0$, we have

$$\frac{d}{dz} [\varphi_{P,q}(z) - \varphi_q(z)] = q \frac{1}{\left[1 + \left(\frac{z}{z_R}\right)^2\right] z_R}. \quad (6.62)$$

The results of Equation 6.62 are the same for the long and the short trajectory implying that $z = 0$ is the best location to observe emissions from both trajectories.

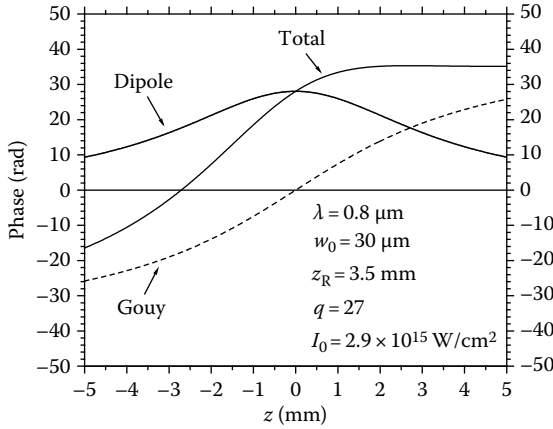


Figure 6.7 Effects of target location on phase matching of the short-trajectory contribution.

For $z < 0$,

$$\frac{d}{dz} [\varphi_{P,q}(z) - \varphi_q(z)] = \frac{\alpha I_0 |z|}{\left[1 + \left(\frac{z}{z_R}\right)^2\right]^2 z_R^2} + q \frac{\left[1 + \left(\frac{z}{z_R}\right)^2\right] z_R}{\left[1 + \left(\frac{z}{z_R}\right)^2\right]^2 z_R^2} > 0, \quad (6.63)$$

which is larger than $z \geq 0$. Consequently, the harmonic generation is not efficient in this region.

6.3.2 On-Axis Phase Matching without Neutral Gas Dispersion

We consider the case that the ionization probability is high enough so that the effect of plasma dispersion is much larger than the neutral gas, or,

$$\Delta k_q(z)z = \frac{\omega_q}{c} \frac{e^2}{2\epsilon_0 m_e} \frac{N}{\omega_1^2} p(z)z, \quad (6.64)$$

which leads to

$$\frac{d}{dz} [\Delta k_q(z)z] = q \frac{e^2}{2c\epsilon_0 m_e} \frac{N}{\omega_1} \frac{d}{dz} [p(z)z]. \quad (6.65)$$

The ionization probability depends strongly on the laser intensity, $I_1(z)$. For $z > 0$, $I_1(z)$ decreases with z , so does the product $p(z)z$, which results in $\frac{d}{dz} [\Delta k_q(z)z] < 0$. Therefore, the plasma dispersion and the intrinsic phase work together to cancel out the phase mismatch effects caused by the Gouy phase. This means that when the plasma contribution is included, the required laser intensity for phase matching is lower.

We can compare the effects of the plasma dispersion to that of the Gouy phase by taking the derivative of the Gouy phase at $z = z_R$:

$$\frac{d\psi}{dz} = q \frac{1/z_R}{1 + \left(\frac{z}{z_R}\right)^2} = q \frac{1}{2z_R}. \quad (6.66)$$

A typical value in the Rayleigh range in experiments is 2.5 mm, which yields $\frac{d\psi}{dz} \approx 0.2q/\text{mm}$.

For $\lambda_1 = 800 \text{ nm}$, and for 1 torr gas pressure, $\frac{e^2}{2c\epsilon_0 m_e} \frac{N}{\omega_1} = 0.074/\text{mm}$. Therefore $\frac{d}{dz} [\Delta k_q(z)z] = 0.074qP \frac{d}{dz} [p(z)z]/\text{mm}$, where the unit of the pressure is in torr. If $P \frac{d}{dz} [p(z)z] < -1$, then the plasma dispersion can effectively cancel the Gouy phase effects.

For simplicity, we assume

$$p(z) = \left(\frac{I_1(z) - I_{th}}{I_s - I_{th}} \right)^m, \quad (6.67)$$

where I_{th} and I_s are the threshold and saturation ionization intensities, respectively. When $m \approx 4$, we find that

$I_{th} \approx I_s/10$. For $z \approx z_R$,

$$I_1(z) \approx \frac{I_0}{2} + \frac{dI_1(z)}{dz} \Big|_{z_R} (z - z_R) = I_0 \left(1 - \frac{z}{2z_R} \right).$$

We then have

$$p(z) \approx \left(\frac{I_0 - 0.1I_s}{0.9I_s} - \frac{I_0 z / 2z_R}{0.9I_s} \right)^m = \left(\frac{I_0}{0.9I_s} \right)^m \left(1 - \frac{0.1I_s}{I_0} - \frac{z}{2z_R} \right)^m, \quad (6.68)$$

$$\left(\frac{d}{dz} p(z)z \right) \Big|_{z_R} = \left(\frac{I_0}{0.9I_s} \right)^m \left[\left(\frac{1}{2} - \frac{0.1I_s}{I_0} \right)^m - \frac{m}{2} \left(\frac{1}{2} - \frac{0.1I_s}{I_0} \right)^{m-1} \right]. \quad (6.69)$$

As it turns out, the value of Equation 6.69 is > -0.1 . Thus the effects of plasma dispersion are comparable to the Gouy phase effects for $P < 10 \text{ torr}$.

6.3.3 Off-Axis Phase Matching

For the atoms located off the axis, we need to take into account the dipole phase change in the radial direction:

$$\varphi_{in}(r, z) = \alpha I_1(r, z) = \alpha I_0 \frac{w_0^2}{w^2(z)} e^{-\frac{r^2}{w^2(z)}}. \quad (6.70)$$

For the atoms near the axis, Equation 6.70 simplifies to

$$\varphi_q(r, z) \approx \alpha I_0 \frac{w_0^2}{w^2(z)} - \alpha I_0 \frac{w_0^2}{w^4(z)} r^2. \quad (6.71)$$

While the effects of the first term have already been discussed, the second term is a parabolic phase, which makes the harmonic beam diverge. This term bears resemblance to the optical Kerr effect in conventional non-linear optics. Because the α value of the short trajectory is more than 20 times smaller than that of the long trajectory, the beam emitted by the short trajectory is more concentrated near the axis and can be filtered out by using an iris.

We assume that the modulus of the wave vector of the harmonic beam is a constant, $k_q = q \frac{\omega_0}{c}$, but its direction depends on the location. This assumption means that the harmonic beam is close to a plane wave. If this is the case, then the total phase difference between the harmonic field and the polarization is

$$\begin{aligned} \varphi_{P,q}(r, z) - \varphi_q(r, z) = & \frac{\alpha I_0}{1 + \left(\frac{z}{z_R}\right)^2} - \frac{\alpha I_0}{\left[1 + \left(\frac{z}{z_R}\right)^2\right]^2} r^2 + q \tan^{-1} \\ & \times \left(\frac{z}{z_R}\right) + \Delta k_q(r, z)z + \left[-qk_1 \frac{1}{2R(z)}\right] r^2. \end{aligned} \quad (6.72)$$

The phase matching is typically shown as a two-dimensional (2D) contour diagram.

When the gas is significantly ionized by the laser, the on-axis plasma density is higher than the off-axis region. Such a transverse plasma density variation produces a diverging lens for the near-infrared (NIR) laser. The variation of the index of refraction due to the plasma also changes $\varphi_{P,q}(r, z) - \varphi_q(r, z)$. In the cases where TW or PW lasers are used to generate high flux attosecond pulses, the focal spot is sufficiently large. One can vary the gas density distribution in both transverse and propagation directions to optimize $\varphi_{P,q}(r, z) - \varphi_q(r, z)$ for phase matching over a large spatial region.

6.4 Phase Matching for Pulsed Lasers

The above analysis was done for CW lasers while almost all high-harmonic generation and attosecond pulse generation experiments have been carried out using pulsed lasers because of the required high intensity. It is very challenging to develop analytical theories for pulsed lasers, although numerical simulations have been used to understand the propagation effects. For this, we need to start with the time-dependent wave equations.

6.4.1 Wave Equation

The wave equation

$$\nabla^2 \vec{\mathcal{E}}(\vec{r}, t) - \frac{1}{c^2} \frac{\partial^2 \vec{\mathcal{E}}(\vec{r}, t)}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}(\vec{r}, t)}{\partial t^2}$$

is valid for pulse laser too. When the laser is linearly polarized, scalar quantities can be used, i.e.,

$$\nabla^2 \mathcal{E}(\vec{r}, t) - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}(\vec{r}, t)}{\partial t^2} = \mu_0 \frac{\partial^2 \mathcal{P}(\vec{r}, t)}{\partial t^2}. \quad (6.73)$$

Assuming the fields are along the x direction, and the medium is ionized by the laser field, then at a given location $\vec{r}$, the polarization

$$\mathcal{P}(t) = N_e(t)[-e \cdot x(t)], \quad (6.74)$$

where N_e is the electron density in the medium. The derivative is

$$-\frac{\partial \mathcal{P}}{\partial t} = ex \frac{\partial N_e}{\partial t} + eN_e \frac{\partial x}{\partial t} = ex_0 \frac{\partial N_e}{\partial t} + eN_e \frac{\partial x}{\partial t}, \quad (6.75)$$

where x_0 is the electron position after it is freed. $\frac{\partial N_e}{\partial t} \neq 0$ only when an electron is born.

We are interested in the second derivative, which is

$$-\frac{1}{e} \frac{\partial^2 \mathcal{P}}{\partial t^2} = x_0 \frac{\partial^2 N_e}{\partial t^2} + \frac{\partial x}{\partial t} \frac{dN_e}{dt} + N_e \frac{\partial^2 x}{\partial t^2} = x_0 \frac{\partial^2 N_e}{\partial t^2} + N_e \frac{\partial^2 x}{\partial t^2}, \quad (6.76)$$

where $N_e(\vec{r}, t)$ is the free electron density as the result of the field ionization, which changes slowly with time, i.e., $\frac{\partial^2 N_e}{\partial t^2} \approx 0$. The free electron density is changed only when an electron is born due to ionization and the birth velocity is $\left. \frac{\partial x}{\partial t} \right|_{t=0} = 0$. Thus,

$$\begin{aligned} \frac{\partial^2 \mathcal{P}(\vec{r}, t)}{\partial t^2} &\approx -eN_e(\vec{r}, t) \frac{\partial^2 x}{\partial t^2} = -eN_e(\vec{r}, t) \frac{-e\mathcal{E}(\vec{r}, t)}{m_e} \\ &= \epsilon_0 \frac{e^2 N_e(\vec{r}, t)}{m_e} \mathcal{E}(\vec{r}, t) = \epsilon_0 \omega_p^2(\vec{r}, t) \mathcal{E}(\vec{r}, t), \end{aligned} \quad (6.77)$$

where $\omega_p(\vec{r}, t)$ is the plasma frequency. Therefore, the wave equation in plasma is

$$\nabla^2 \mathcal{E}(\vec{r}, t) - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}(\vec{r}, t)}{\partial t^2} = \frac{\omega_p^2(\vec{r}, t)}{c^2} \mathcal{E}(\vec{r}, t). \quad (6.78)$$

6.4.1.1 Beams with Axial symmetry

In most of the experiments, the laser and harmonic beams are axially symmetric. It is more convenient to use cylindrical coordinates in which the operator

$$\nabla^2 \mathcal{E} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \mathcal{E}}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \mathcal{E}}{\partial \phi^2} + \frac{\partial^2 \mathcal{E}}{\partial z^2} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \mathcal{E}}{\partial r} \right) + \frac{\partial^2 \mathcal{E}}{\partial z^2}, \quad (6.79)$$

where

r and z are radial and propagation axes

ϕ is the azimuthal angle

The wave equation is then

$$\begin{aligned} \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \mathcal{E}(r, z, t)}{\partial r} \right) + \frac{\partial^2 \mathcal{E}(r, z, t)}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}(r, z, t)}{\partial t^2} \\ = \frac{\omega_p^2(r, z, t)}{c^2} \mathcal{E}(r, z, t). \end{aligned} \quad (6.80)$$

A possible solution of the wave equation is a plane wave and Gaussian pulse in vacuum so that

$$\mathcal{E}(z,t) = e^{-a\left(t-\frac{z}{c}\right)^2} e^{i\omega_0\left(t-\frac{z}{c}\right)}, \quad (6.81)$$

where $a = 2 \ln 2 \frac{1}{\tau^2}$ and τ is the pulse width.

6.4.1.2 Retarded Coordinate

In a retarded coordinate frame, $z' = z$, $t' = t - \frac{z}{c}$, the field $\mathcal{E}(z',t') = e^{-at'^2} e^{i\omega_0 t'}$. Perform the following substitutes:

$$\begin{aligned} \frac{\partial^2 \mathcal{E}(z,t)}{\partial t^2} &= \frac{\partial}{\partial t} \left(\frac{\partial \mathcal{E}(z,t)}{\partial t} \right) = \frac{\partial}{\partial t} \left(\frac{\partial \mathcal{E}(z',t')}{\partial t'} \frac{\partial t'}{\partial t} + \frac{\partial \mathcal{E}(z',t')}{\partial z'} \frac{\partial z'}{\partial t} \right) \\ &= \frac{\partial}{\partial t} \left(\frac{\partial \mathcal{E}(z',t')}{\partial t'} \right) \\ &= \frac{\partial}{\partial t'} \left(\frac{\partial \mathcal{E}(z',t')}{\partial t'} \right) \frac{\partial t'}{\partial t} + \frac{\partial}{\partial z'} \left(\frac{\partial \mathcal{E}(z',t')}{\partial t'} \right) \frac{\partial z'}{\partial t} = \frac{\partial^2 \mathcal{E}(z',t')}{\partial t'^2}, \end{aligned} \quad (6.82)$$

i.e.,

$$-\frac{1}{c^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = -\frac{1}{c^2} \frac{\partial^2 \mathcal{E}}{\partial t'^2}, \quad (6.83)$$

and

$$\begin{aligned} \frac{\partial^2 \mathcal{E}}{\partial z^2} &= \frac{\partial}{\partial z} \left(\frac{\partial \mathcal{E}}{\partial z} \right) = \frac{\partial}{\partial z} \left(\frac{\partial \mathcal{E}}{\partial z'} \frac{\partial z'}{\partial z} + \frac{\partial \mathcal{E}}{\partial t'} \frac{\partial t'}{\partial z} \right) = \frac{\partial}{\partial z} \left(\frac{\partial \mathcal{E}}{\partial z'} - \frac{1}{c} \frac{\partial \mathcal{E}}{\partial t'} \right) \\ &= \frac{\partial}{\partial z'} \left(\frac{\partial \mathcal{E}}{\partial z'} - \frac{1}{c} \frac{\partial \mathcal{E}}{\partial t'} \right) \frac{\partial z'}{\partial z} + \frac{\partial}{\partial t'} \left(\frac{\partial \mathcal{E}}{\partial z'} - \frac{1}{c} \frac{\partial \mathcal{E}}{\partial t'} \right) \frac{\partial t'}{\partial z} \\ &= \frac{\partial^2 \mathcal{E}}{\partial z'^2} - \frac{2}{c} \frac{\partial^2 \mathcal{E}}{\partial z' \partial t'} + \frac{1}{c^2} \frac{\partial^2 \mathcal{E}}{\partial t'^2}, \end{aligned} \quad (6.84)$$

we have the wave equation for the electric field

$$\begin{aligned} \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \mathcal{E}(r,z',t')}{\partial r} \right) + \frac{\partial^2 \mathcal{E}(r,z',t')}{\partial z'^2} - \frac{2}{c} \frac{\partial^2 \mathcal{E}(r,z',t')}{\partial t' \partial z'} \\ = \frac{\omega_p^2(r,z',t')}{c^2} \mathcal{E}(r,z',t'). \end{aligned} \quad (6.85)$$

It is interesting that the second-order time derivative does not show up in the retarded frame.

6.4.1.3 Plane Waves

An example of the plane wave solution from Equation 6.85 in the retarded frame is

$$\mathcal{E}(z',t') = e^{-at'^2} e^{i\omega_0 t'}, \quad (6.86)$$

which does not change with z' , implying that the time-dependent field is identical for any spatial location.

For a Gaussian beam whose waist size is much larger than the center wavelength, the change of the field along z' is much slower than the change in the r direction, thus, $\frac{\partial^2 \varepsilon}{\partial z'^2} \ll \frac{1}{r} \frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial \varepsilon}{\partial r} \right)$. We drop the $\frac{\partial^2 \varepsilon}{\partial z'^2}$ term to obtain the *paraxial* wave equation in time domain

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \varepsilon(r, z', t')}{\partial r} \right) - \frac{2}{c} \frac{\partial^2 \varepsilon(r, z', t')}{\partial t' \partial z'} = \frac{\omega_p^2(r, z', t')}{c^2} \varepsilon(r, z', t'). \quad (6.87)$$

This equation is still too hard to solve, because it contains derivatives of three variables. The time derivative can be removed by transforming the equation into the frequency domain.

6.4.2 Paraxial Wave Equation in the Frequency Domain

Applying a Fourier transform to the wave Equation 6.85 yields

$$\begin{aligned} \hat{F} \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \varepsilon(r, z', t')}{\partial r} \right) \right] + \hat{F} \left[\frac{\partial^2 \varepsilon(r, z', t')}{\partial z'^2} \right] - \hat{F} \left[\frac{2}{c} \frac{\partial^2 \varepsilon(r, z', t')}{\partial t' \partial z'} \right] \\ = \hat{F} \left[\frac{\omega_p^2(r, z', t')}{c^2} \varepsilon(r, z', t') \right]. \end{aligned} \quad (6.88)$$

The Fourier transform of the electric field

$$\hat{F}[\varepsilon(t')] = \int_{-\infty}^{+\infty} \varepsilon(t') e^{-i\omega t'} dt' = \tilde{E}(\omega), \quad (6.89)$$

where $\tilde{E}(\omega)$ is the electric field in the frequency domain and in the retarded frame. This field is different from that in the lab frame, $\tilde{E}_{lab}(\omega) = \tilde{E}(\omega) e^{i\omega \tilde{z}}$.

The Fourier transform

$$\begin{aligned} \hat{F} \left[\frac{\partial}{\partial t'} \varepsilon(t') \right] &= \int_{-\infty}^{+\infty} \left[\frac{\partial}{\partial t'} \varepsilon(t') \right] e^{-i\omega t'} dt' \\ &= \int_{-\infty}^{+\infty} \left[\frac{\partial}{\partial t'} \int_{-\infty}^{+\infty} \tilde{E}(\omega) e^{i\omega t'} d\omega \right] e^{-i\omega t'} dt' \\ &= \int_{-\infty}^{+\infty} \left\{ \int_{-\infty}^{+\infty} \frac{\partial}{\partial t'} [\tilde{E}(\omega) e^{i\omega t'}] d\omega \right\} e^{-i\omega t'} dt' \\ &= \int_{-\infty}^{+\infty} \left\{ \int_{-\infty}^{+\infty} (i\omega) \tilde{E}(\omega) e^{i\omega t'} d\omega \right\} e^{-i\omega t'} dt' \\ &= \hat{F} \left\{ \hat{F}^{-1} [i\omega \tilde{E}(\omega)] \right\} = i\omega \tilde{E}(\omega). \end{aligned} \quad (6.90)$$

Define

$$\hat{F} \left[\frac{\omega_p^2(r, z', t')}{c^2} \varepsilon(r, z', t') \right] = \tilde{G}(r, z', \omega), \quad (6.91)$$

we finally obtain the wave equation in frequency domain

$$\begin{aligned} \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \tilde{E}(r, z', \omega)}{\partial r} \right) + \frac{\partial^2 \tilde{E}(r, z', \omega)}{\partial z'^2} - \frac{2i\omega}{c} \frac{\partial \tilde{E}(r, z', \omega)}{\partial z'} \\ = \tilde{G}(r, z', \omega). \end{aligned} \quad (6.92)$$

Making the *paraxial* approximation gives

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \tilde{E}(r, z', \omega)}{\partial r} \right) - \frac{2i\omega}{c} \frac{\partial \tilde{E}(r, z', \omega)}{\partial z'} = \tilde{G}(r, z', \omega), \quad (6.93)$$

i.e., we assume that $\frac{\partial^2 E}{\partial z'^2}$ is much smaller than other terms in Equation 6.92.

6.4.3 Carrier-Envelope Phase

The time domain description of a transform-limited pulse with carrier-envelope phase φ_{CE} is

$$\varepsilon(t) = e^{-at^2} e^{i(\omega_0 t + \varphi_{CE})}. \quad (6.94)$$

In the frequency domain,

$$\begin{aligned} \tilde{E}(\omega) &= \int_{-\infty}^{+\infty} \varepsilon(t) e^{-i\omega t} dt = \int_{-\infty}^{+\infty} e^{-a^2} e^{i(\omega_0 t + \varphi_{CE})} e^{-i\omega t} dt \\ &= e^{i\varphi_{CE}} \int_{-\infty}^{+\infty} e^{-at^2} e^{i\omega_0 t} e^{-i\omega t} dt = e^{i\varphi_{CE}} e^{-\frac{(\omega - \omega_0)^2}{4a}} \end{aligned} \quad (6.95)$$

meaning that the carrier-envelope phase, φ_{CE} , corresponds to a phase shift to all the frequency components.

6.4.4 Propagation of Few-Cycle Pulses

Single isolated attosecond pulses can be generated by using few-cycle lasers compressed by hollow-core fiber/chirped mirrors. If we assume that at the exit of the fiber the transverse beam profile is a Gaussian (which is a good approximation of the actual Bessel profile) and the spot size is the same for all frequency components, then when the exit of the fiber is imaged to the target with a perfect lens (or mirror), the transverse beam profile is also a Gaussian and the spot size is the same for all frequency components at the focus.

The time domain description of the linearly polarized pulse at the focus is

$$\varepsilon(t, r, z = 0) = E_0 e^{-at^2} e^{i(\omega_0 t + \varphi_{CE})} e^{-\frac{r^2}{w_0^2}}. \quad (6.96)$$

The peak intensity at the focus is $I_0(t = 0, r = 0, z = 0) = \frac{c\epsilon_0}{2} E_0^2$. Thus, the frequency domain description of the pulse at the focus is

$$\begin{aligned}\tilde{E}(\omega, r, z = 0) &= \int_{-\infty}^{+\infty} \varepsilon(t, r, z = 0) e^{-i\omega t} dt \\ &= E_0 \sqrt{\frac{\pi}{a}} w_0 e^{i\varphi_{CE}} e^{-\frac{(\omega - \omega_0)^2}{4a}} \frac{1}{w_0} e^{-\frac{r^2}{w_0^2}}.\end{aligned}\quad (6.97)$$

The integral was done as

$$\begin{aligned}\int_{-\infty}^{\infty} e^{-at^2 + i\omega_0 t} e^{-i\omega t} dt &= \int_{-\infty}^{\infty} e^{-[at^2 + i(\omega - \omega_0)t]} dt \\ &= \frac{1}{\sqrt{a}} \int_{-\infty}^{+\infty} e^{-\left[(\sqrt{at})^2 + i\frac{(\omega - \omega_0)}{\sqrt{a}}(\sqrt{at})\right]} d(\sqrt{at}) \\ &= \frac{1}{\sqrt{a}} e^{-\frac{(\omega - \omega_0)^2}{4a}} \int_{-\infty}^{+\infty} e^{-\left[t'^2 + i\frac{(\omega - \omega_0)}{\sqrt{a}}t' - \frac{(\omega - \omega_0)^2}{4a}\right]} dt' \\ &= \frac{1}{\sqrt{a}} e^{-\frac{(\omega - \omega_0)^2}{4a}} \int_{-\infty}^{+\infty} e^{-\left(t' + i\frac{\omega - \omega_0}{2\sqrt{a}}\right)^2} d \\ &\quad \times \left(t' + i\frac{\omega - \omega_0}{2\sqrt{a}}\right) \\ &= \sqrt{\frac{\pi}{a}} e^{-\frac{(\omega - \omega_0)^2}{4a}}.\end{aligned}\quad (6.98)$$

After the focus, at point (r, z) , the field becomes

$$\begin{aligned}\tilde{E}(\omega, r, z) &= \varepsilon_0 \sqrt{\frac{\pi}{a}} w_0 e^{i\varphi_{CE}} e^{-\frac{(\omega - \omega_0)^2}{4a}} \frac{1}{w(\omega, z)} e^{-\frac{r^2}{w(\omega, z)^2}} e^{-i\frac{\omega}{c} \frac{r^2}{2R(\omega, z)}} e^{i\psi(\omega, z)} \\ &= \varepsilon_0 \sqrt{\frac{\pi}{a}} e^{i\varphi_{CE}} \\ &\quad \times \frac{1}{\sqrt{1 + \left(\frac{z}{z_R(\omega)}\right)^2}} e^{-\frac{(\omega - \omega_0)^2}{4a}} e^{-\frac{r^2}{w(\omega, z)^2}} e^{-i\frac{\omega}{c} \frac{r^2}{2R(\omega, z)}} e^{i\psi(\omega, z)},\end{aligned}\quad (6.99)$$

where the frequency-dependent Gaussian beam parameters

$$\begin{aligned}w(\omega, z) &= w_0 \sqrt{1 + \left(\frac{z}{z_R(\omega)}\right)^2} \\ R(\omega, z) &= z + \frac{z_R^2(\omega)}{z} \\ \psi(\omega, z) &= \tan^{-1} \left(\frac{z}{z_R(\omega)}\right) \\ z_R(\omega) &= \frac{\pi w_0^2}{\lambda} = \frac{1}{2} k w_0^2 = \frac{\omega w_0^2}{2c}.\end{aligned}\quad (6.100)$$

The time domain description of the pulse at (r, z) is

$$\begin{aligned}\varepsilon(t, r, z) &= \frac{1}{2\pi} \int_{-\infty}^{+\infty} \tilde{E}(\omega, r, z) e^{i\omega t} d\omega \\ &= \frac{E_0}{2\pi} \sqrt{\frac{\pi}{a}} e^{i\varphi_{CE}} \int_{-\infty}^{+\infty} \\ &\quad \times \frac{1}{\sqrt{1 + \left(\frac{z}{z_R(\omega)}\right)^2}} e^{-\frac{(\omega - \omega_0)^2}{4a}} e^{-\frac{z^2}{w(\omega, z)^2}} e^{-i\frac{\omega}{c} \frac{z^2}{2R(\omega, z)}} e^{i\eta(\omega, z)} e^{i\omega t} d\omega. \quad (6.101)\end{aligned}$$

6.4.5 Integral Approach

The differential wave equations discussed so far have been widely used in simulating high-harmonic and attosecond pulse generation. Here, we briefly introduce the integral approach which is more intuitive in explaining the physics.

For the given electric field at the source plane, $\varepsilon(t, r_0, z_0)$ we want to find out the field at the observation plane, $\varepsilon(t, r, z)$. These fields can be expressed by their Fourier components:

$$\varepsilon(t, r_0, z_0) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \tilde{E}(\omega, r_0, z_0) e^{i\omega t} d\omega, \quad (6.102)$$

$$\varepsilon(t, r, z) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \tilde{E}(\omega, r, z) e^{i\omega t} d\omega. \quad (6.103)$$

By the change of variable $\omega' = -\omega$ the equations are transformed into

$$\varepsilon(t, r_0, z_0) = -\frac{1}{2\pi} \int_{-\infty}^{+\infty} \tilde{E}(-\omega', r_0, z_0) e^{-i\omega' t} d\omega', \quad (6.104)$$

$$\varepsilon(t, r, z) = -\frac{1}{2\pi} \int_{-\infty}^{+\infty} \tilde{E}(-\omega', r, z) e^{-i\omega' t} d\omega'. \quad (6.105)$$

For each frequency component, the field $\tilde{E}(-\omega', r, z)$ can be calculated from $\tilde{E}(-\omega', r_0, z_0)$ using the Rayleigh–Sommerfeld integral

$$\begin{aligned}\tilde{E}(-\omega', r, z) &= \frac{1}{i\lambda} \iint \tilde{E}(-\omega', r_0, z_0) \frac{e^{ik\rho}}{\rho} \cos \theta ds \\ &= -i \frac{\omega'}{2\pi c} \iint \tilde{E}(-\omega', r_0, z_0) \frac{e^{i\omega' \rho / (2\pi c)}}{\rho} \cos \theta ds. \quad (6.106)\end{aligned}$$

where $\rho = \sqrt{(r - r_0)^2 + (z - z_0)^2}$. Thus,

$$\begin{aligned}\varepsilon(t, r, z) &= -\frac{1}{2\pi} \int_{-\infty}^{+\infty} \left[-i \frac{\omega'}{2\pi c} \iint \tilde{E}(-\omega', r_0, z_0) \frac{e^{i\omega'(\rho/2\pi c)}}{\rho} \cos \theta ds \right] e^{-i\omega' t} d\omega' \\ &= \iint \frac{\cos \theta}{2\pi c \rho} \int_{-\infty}^{+\infty} (-i2\pi v') \tilde{E}(-\omega', r_0, z_0) e^{-i2\pi v'(t-\rho/c)} dv' ds.\end{aligned}\quad (6.107)$$

Since

$$\begin{aligned}\frac{d\varepsilon(t, r_0, z_0)}{dt} &= \frac{d}{dt} \int_{-\infty}^{+\infty} \tilde{E}(-\omega', r_0, z_0) e^{-i2\pi v' t} dv' \\ &= \int_{-\infty}^{+\infty} (-i2\pi v') \tilde{E}(-\omega', r_0, z_0) e^{-i2\pi v' t} dv',\end{aligned}\quad (6.108)$$

we have

$$\begin{aligned}\varepsilon(t, r, z) &= \iint \frac{\cos \theta}{2\pi c \rho} \frac{d}{dt} \varepsilon\left(t - \frac{\rho}{c}, r_0, z_0\right) ds \\ &= \int_0^\infty \frac{\cos \theta}{2\pi c \rho} \frac{d}{dt} \varepsilon\left(t - \frac{\rho}{c}, r_0, z_0\right) 2\pi r dr.\end{aligned}\quad (6.109)$$

Assuming

$$\varepsilon(t, r, z = 0) = E_0 e^{-at^2} e^{i(\omega_0 t + \phi_0)} e^{-\frac{r^2}{w_0^2}}, \quad (6.110)$$

we arrive at

$$\begin{aligned}\frac{d}{dt} \varepsilon\left(t - \frac{\rho}{c}, r_0, z_0 = 0\right) &= E_0 e^{i\phi_0} e^{-\frac{r^2}{w_0^2}} \frac{d}{dt} e^{-a\left(t - \frac{\rho}{c}\right)^2 + i2\pi v_0\left(t - \frac{\rho}{c}\right)} = E_0 e^{i\phi_0} e^{-\frac{r_0^2}{w_0^2}} \\ &\quad \times \left[-2a\left(t - \frac{\rho}{c}\right) + i2\pi v_0 \right] e^{-a\left(t - \frac{\rho}{c}\right)^2 + i2\pi v_0\left(t - \frac{\rho}{c}\right)}.\end{aligned}\quad (6.111)$$

Finally,

$$\begin{aligned}\varepsilon(t, r, z) &= \int_0^\infty \int_{\phi=0}^{2\pi} \frac{\cos \theta}{2\pi c \rho} \frac{d}{dt} \varepsilon\left(t - \frac{\rho}{c}, r_0, z_0\right) r_0 dr_0 d\phi \\ &= \frac{E_0 e^{i\phi_0}}{2\pi c} \int_{r_0=0}^\infty \int_0^{2\pi} \frac{\cos \theta}{\rho} e^{-\frac{r_0^2}{w_0^2}} \\ &\quad \times \left[-2a\left(t - \frac{\rho}{c}\right) + i2\pi v_0 \right] e^{-a\left(t - \frac{\rho}{c}\right)^2} e^{i(\omega_0 t - k_0 \rho)} r_0 dr_0 d\phi.\end{aligned}\quad (6.112)$$

At this point, it is necessary to introduce the Fraunhofer condition which means three requirements:

1. For the phase term we require that $\rho = R - \frac{r_0 r \cos(\phi - \Phi)}{R}$, where $R = \sqrt{r^2 + (z - z_0)^2}$, ϕ and Φ are the azimuthal angles in the source and observation planes respectively.

Here, we will take $\Phi = 0$ due to the axial symmetry. Therefore,

we are left with $\rho = R - \frac{r_0 r \cos(\phi)}{R}$.

2. For the other terms we assume that $\rho = R$.

3. Also, we must set $\theta = 0$.

After these three conditions are met,

$$\begin{aligned} \varepsilon(t, r, z) = & \frac{E_0 e^{i\phi_0}}{2\pi c R} \left[-2a \left(t - \frac{R}{c} \right) + i\omega_0 \right] e^{-a \left(t - \frac{R}{c} \right)^2} e^{i(\omega_0 t - k_0 R)} \\ & \times \int_{r_0=0}^{\infty} \int_0^{2\pi} e^{-\frac{r_0^2}{w_0^2}} e^{i(k_0 r_0 r / R) \cos(\phi)} r_0 dr_0 d\phi. \end{aligned} \quad (6.113)$$

The integral associated with the variable ϕ can be expressed by the Bessel function

$$J_0(u) = \frac{1}{2\pi} \int_0^{2\pi} e^{iu \cos v} dv. \quad (6.114)$$

Thus,

$$\begin{aligned} \varepsilon(t, r, z) = & \frac{E_0 e^{i\phi_0}}{c R} \left[-2a \left(t - \frac{R}{c} \right) + i\omega_0 \right] e^{-a \left(t - \frac{R}{c} \right)^2} e^{i\omega_0 \left(t - \frac{R}{c} \right)} \\ & \times \int_0^{\infty} e^{-\frac{r_0^2}{w_0^2}} J_0(k_0 r_0 r / R) r_0 dr_0. \end{aligned} \quad (6.115)$$

For the on axis point $r = 0$, and $R = z$

$$\begin{aligned} \varepsilon(t, r, z) = & \frac{E_0 e^{i\phi_0}}{c Z} \left[-2a \left(t - \frac{z}{c} \right) + i\omega_0 \right] e^{-a \left(t - \frac{z}{c} \right)^2} e^{i\omega_0 \left(t - \frac{z}{c} \right)} \\ & \times \int_0^{\infty} e^{-\frac{r_0^2}{w_0^2}} r_0 dr_0. \end{aligned} \quad (6.116)$$

In the retarded frame, $t' = t - \frac{z}{c}$, so

$$\varepsilon(t', r, z) = -\frac{E_0 e^{i\phi_0}}{c z} \left[-2 \left(\frac{2 \ln 2}{\tau_p^2} \right) t' + i\omega_0 \right] e^{-a(t')^2} e^{i\omega_0 t'} \frac{w_0^2}{2}. \quad (6.117)$$

6.4.6 Calculating the Electric Field in the Far-Field

Given the complex electric field in the near-field, $\tilde{E}(r)$, the field in the far-field is the Fourier transform of $\tilde{E}(r)$, i.e.,

$$\varepsilon(\kappa) = \int_{r=0}^{\infty} \tilde{E}(r) \left[\int_{\theta=0}^{2\pi} e^{i\kappa r \cos \theta} d\theta \right] r dr = 2\pi \int_0^{\infty} \tilde{E}(r) J_0(\kappa r) r dr. \quad (6.118)$$

This transform is also called the Fourier–Bessel transform, or the Hankel transform of zeroth order. The angular spatial frequency can be expressed as

$$\kappa = \frac{2\pi}{\lambda} \tan \theta, \quad (6.119)$$

where

λ is the wavelength of the field

θ is the diffraction angle

For high-harmonic generation, $\theta \leq 10$ mrad. Under the small angle approximation, $\tan \theta \approx \theta$. Thus, $\kappa = \frac{2\pi}{\lambda} \theta$, and

$$\kappa r = 2\pi \theta \frac{r}{\lambda} = q 2\pi \theta \frac{r}{\lambda_0}, \quad (6.120)$$

where

q is the harmonic order

$\lambda_0 = 0.8 \mu\text{m}$ is the driving laser wavelength when Ti:Sapphire lasers are used

In many experiments, the maximum value of the laser focal spot size is $r \approx 100 \mu\text{m}$. For $q = 100$, the maximum value of $\kappa r \approx 10,000$. Thus, the transform can be estimated by

$$\varepsilon(\kappa) \approx \sum_{i=1}^{10,000} \tilde{E}(r_i) J_0(\kappa r_i) r_i \frac{r_{\max}}{10,000} \approx 2\pi \int_0^{\infty} \tilde{E}(r) J_0(\kappa r) r dr. \quad (6.121)$$

6.5 Compensating the Chirp of Attosecond Pulses*

As discussed in Chapter 4, the attosecond pulse from the short trajectory is positively chirped. The chirp must be removed to produce transform-limited XUV pulses. The chirp is inversely proportional to the laser intensity and wavelength. Thus, it can be reduced by using long wavelength driving lasers or using intensity close to the ionization saturation.

Once the attosecond pulse is produced, materials with negative group velocity dispersions can be placed after the gas target to compensate the chirp. Other schemes based on XUV prisms, gratings, chirped mirrors, and phase modulators like what have been used in femtosecond lasers are yet to be developed.

The chirp of the XUV pulse from a single atom depends on the laser peak intensity. Since the laser intensity varies along both transverse and propagation direction inside the target, the final pulse on the detector is a superposition of many pulses with different chirp. The contribution of each atom is affected by the phase matching. In this section, the spectral phase of the XUV supercontinuum generated by the polarization gating on the detector is investigated, which includes the effects of phase matching.

* This section is adapted from Chang, Z., Chirp of the attosecond pulses generated by a polarization gating, *Phys. Rev. A* 71, 023813 (2005).

6.5.1 Numerical Simulation Method

Helium is chosen as the target gas. Due to its large ionization potential, $I_p = 24.59$ eV, the saturation ionization intensity of helium is the highest among neutral atoms and molecules. Thus, one expects to generate the shortest attosecond pulse from helium. The chirp is relatively small due to the high intensity, which needs thinner filter to compensate the chirp.

First, the polarization gating of the high-harmonic generation from a single atom was simulated with the strong field approximation. Then the macroscopic attosecond signal was calculated by solving the 3D wave equation for the attosecond XUV field.

6.5.1.1 NIR Laser Field

The NIR laser is a Gaussian beam propagating in the z direction. Cylindrical symmetry with respect to the z axis is assumed, which converts the 3D problem into a 2D one to reduce the numerical simulation time. The beam waist at the focus is $w_0 = 25$ μm , which gives a Rayleigh range of $z_R = 2.6$ mm. The focal spot size is chosen for femtosecond lasers with a few millijoule pulse energy to reach the required peak intensity on the target. A 1 mm long gas target is centered at 2 mm after the laser focus. The target length is shorter than the Rayleigh range to avoid the averaging due to the carrier envelope (CE) phase variation inside the target. The position of the target is chosen for the best phase matching, as discussed in the next section.

It is assumed that the laser pulse with a time-dependent ellipticity is formed by the superposition of a left and a right-circularly polarized Gaussian pulse, as explained in Chapter 4. The center wavelength is 750 nm, which corresponds to a 2.5 fs optical period. The peak field amplitude, E_0 , carrier frequency (ω_0), pulse duration ($\tau_p = 5$ fs) and carrier-envelope phase ($\varphi_{CE} = \pi/2$) are the same for the two circularly polarized pulses. The delay between them is $T_d = 5$ fs, which is two optical cycles. The two-cycle pulse is chosen to reduce the ionization of the target atom by the laser field in front of the polarization gate. The high-power two-cycle pulses can be generated by compressing the laser pulses from Ti:Sapphire chirped pulse amplifier with gas-filled hollow-core fiber and chirped mirrors, as discussed in Chapter 2. The chosen center wavelength is close to that from the hollow-core fiber.

The electric field of the combined pulse is $\vec{\epsilon}(t) = \epsilon_{drive}(t)\hat{x} + \epsilon_{gate}(t)\hat{y}$ where $\hat{x}$ and $\hat{y}$ are unit vectors in the x and y directions, respectively. The x component is the driving field that generates the attosecond XUV radiation whereas the y component is the gating field for suppressing all the XUV pulse except one. The two field components of the laser field at a given spatial point are

$$\begin{aligned}\epsilon_{drive}(t) &= E_0\hat{x} \left[e^{-2\ln(2)\left(\frac{t-T_d/2}{\tau_p}\right)^2} + e^{-2\ln(2)\left(\frac{t+T_d/2}{\tau_p}\right)^2} \right] \cos(\omega t + \varphi_{CE}), \\ \epsilon_{gate}(t) &= E_0\hat{y} \left[e^{-2\ln(2)\left(\frac{t-T_d/2}{\tau_p}\right)^2} - e^{-2\ln(2)\left(\frac{t+T_d/2}{\tau_p}\right)^2} \right] \sin(\omega t + \varphi_{CE}).\end{aligned}\tag{6.122}$$

As explained in Chapter 4, the laser field is linearly polarized at $t=0$, where the polarization gate is centered. The peak laser intensity there is 1.4×10^{15} W/cm². The field strength E_0 and the CE phase φ_{CE} are functions of the position.

6.5.1.2 Single-Atom Response

The single-atom response is calculated by the strong field approximation discussed in Chapter 5. The calculation for each atom takes much less time than numerically solving the time-dependent Schrödinger equation. This is very important because the calculations need to be done for many atoms at different positions in the gas target.

When the ellipticity of the driving pulse changes with time, the two transverse components of the dipole moment were calculated separately. Calculations showed that the amplitude of the XUV spectrum along the gating field direction $\hat{y}$ was much smaller than that along the driving field direction $\hat{x}$. This is because the laser field is polarized in the $\hat{x}$ inside the polarization gate where the XUV field is emitted. To save calculation time, only the dipole moment along the $\hat{x}$ direction of the laser field was calculated, which was done by performing the integral,

$$\begin{aligned} x(t) \approx & i \int_0^\infty d\tau \left(\frac{\pi}{\epsilon + i\tau/2} \right)^{3/2} d_x^* [\vec{p}_s(t, \tau) - \vec{A}(t)] e^{-iS(\vec{p}_s, t, \tau)} \\ & \times \left\{ \epsilon_x(t - \tau) \cdot d_x [\vec{p}_s(t, \tau) - \vec{A}(t - \tau)] + \epsilon_y(t - \tau) \right. \\ & \left. \cdot d_y [\vec{p}_s(t, \tau) - \vec{A}(t - \tau)] \right\} |a(t)|^2 + c.c., \end{aligned} \quad (6.123)$$

where

$\vec{A}(t)$ is the vector potential of the laser field

ϵ is a small number

$a(t)$ is the ground-state amplitude as calculated by the ADK rate

When the NIR laser field is elliptically polarized, the field-free dipole transition matrix elements between the ground state and the plane wave continuum state is a vector, which can be resolved into two components,

$$\begin{aligned} & d_x^* [\vec{p}_s(t, \tau) - \vec{A}(t)] \\ & = -i \frac{2^{7/2}}{\pi} \alpha^{5/4} \frac{p_{s,x}(t, \tau) - A_x(t)}{\{ [p_{s,x}(t, \tau) - A_x(t)]^2 + [p_{s,y}(t, \tau) - A_y(t)]^2 + \alpha \}^3}, \end{aligned} \quad (6.124)$$

$$\begin{aligned} & d_y^* [\vec{p}_s(t, \tau) - \vec{A}(t)] \\ & = -i \frac{2^{7/2}}{\pi} \alpha^{5/4} \frac{p_{s,y}(t, \tau) - A_y(t)}{\{ [p_{s,x}(t, \tau) - A_x(t)]^2 + [p_{s,y}(t, \tau) - A_y(t)]^2 + \alpha \}^3}, \end{aligned} \quad (6.125)$$

where $\alpha = 2I_p$. I_p is the ionization potential of the atom.

The two components of the canonical momentum of the electron corresponding to a stationary phase are calculated by $p_{s,x}(t, \tau)$

$= \int_{t-\tau}^t dt'' A_x(t'')/\tau$, and $p_{s,y}(t,\tau) = \int_{t-\tau}^t dt'' A_y(t'')/\tau$. Finally, the quasiclassical action of the electron is calculated by

$$S(\vec{p}_s, t, \tau) = I_p \tau - \frac{1}{2} [p_{s,x}^2(t, \tau) + p_{s,y}^2(t, \tau)] + \frac{1}{2} \int_{t-\tau}^t dt'' [A_x^2(t'') + A_y^2(t'')]. \quad (6.126)$$

Since S oscillates very rapidly with τ , care must be taken when the integral in Equation 6.123 is performed numerically.

6.5.1.3 Macroscopic Attosecond Signal

The macroscopic attosecond signal from all the atoms in the target was calculated by solving the wave propagation equation of the XUV field. It turns out that it is easier to solve the equation numerically in the frequency domain than in the time domain. Furthermore, it is also more convenient to express the field in the reference frame that moves with the XUV pulse.

In the moving frame, Equation 6.16 for the XUV field component $E(\omega_X, r, z)$ with frequency, ω_X , can be cast into

$$\nabla_T^2 E(\omega_X, r, z) - \frac{2i\omega_X}{c} \frac{\partial E(\omega_X, r, z)}{\partial z} = -\mu_0 \omega_X^2 P_{nl}(\omega_X, r, z), \quad (6.127)$$

where

r is the transverse coordinate

z is the propagation coordinate in the moving frame

The nonlinear polarization is $P_{nl}(\omega_X, r, z) \propto x(\omega_X, r, z)$, where $x(\omega_X, r, z)$ is the dipole moment of an atom at point (r, z) in the frequency domain. It is the Fourier transform of the $x(t, r, z)$ calculated with Equation 6.123. The strong field approximation gives the correct temporal shape and phase of, but not the peak amplitude value of the dipole moment, which is good enough for calculating the chirp of the attosecond pulse.

In experiments, the gas density changes with z . For simplicity, the atomic density of the target is assumed to be a constant. Equation 6.127 is solved numerically for each frequency in a spatial grid. The single-atom dipole moments at the grid points, $x(\omega_X, r, z)$, are calculated first and then are entered into Equation 6.127 through the polarization $P_{nl}(\omega_X, r, z)$.

The output XUV spectrum from the target is calculated by adding up the power spectrum at each transverse point at the exit of the target. This corresponds to the case when the detector is located at the exit of the target, or at the imaging location of the target, as in streak camera measurement described in Chapter 8.

To see the attosecond pulse in the time domain, a square spectral window is applied to the XUV spectrum at each transverse point at the exit of the target, and inverse Fourier transforms are performed to obtain the attosecond pulse for that point. The pulses of all the points are summed up to yield the final pulse intensity. Of course, other types of spectral window can also be used, which would give similar results. The square window is chosen just for convenience.

6.5.2 Simulation Results

6.5.2.1 Ground-State Depletion

To illustrate the effects of the NIR laser pulse duration on the polarization gating, the ground-state population was calculated for two cases. In the first case, the duration of the two circularly polarized pulses is 5 fs. Their delay is also 5 fs. For a comparison, the ground-state population with 10 fs lasers was also calculated. The delay between the two 10 fs pulses is 15 fs, which was chosen to yield similar spectrum profile as that from the 5 fs lasers.

The attosecond pulses are generated inside the polarization gate around $t = 0$. The ionization probability at $t = 0$ is 13.4% for the 5 fs pulses that is much less than that with the 10 fs pulses (93.4%). The intensity of attosecond pulse generated by the 5 fs pulses is more than two orders of magnitude higher than that produced by the 10 fs pulses, as the single-atom calculation results shows in Figure 6.8. The lower XUV intensity in the 10 fs laser case is the result of larger ground-state population of the atom by the laser field in front of the polarization gate. There are fewer neutral atoms left (6.6% in the 10 fs case as compared to the 86% in the 5 fs case) to emit XUV light. One should keep in mind that the XUV photon flux is proportional to the square of the neutral atom population. Therefore, it is critical to use two-cycle lasers for polarization gating.

6.5.2.2 Gated XUV Spectrum

The simulation of attosecond pulse generation by polarization gating that includes the propagation effects is done for only the 5 fs laser pulse because of the higher XUV photon flux as compared to longer driving laser pulses. The plasma induced defocusing of the NIR laser and phase mismatch are not included. The phase mismatch caused by the Gouy phase of the driving laser is partially corrected by the intrinsic phase variation in space. Both the single-atom spectrum and the macroscopic spectrum are shown in

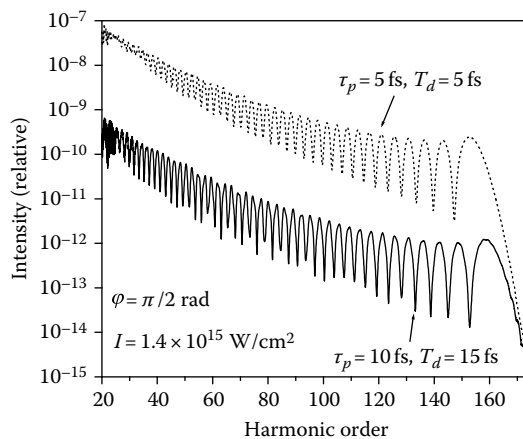


Figure 6.8 Single-atom polarization gated high-order harmonic spectra. The dotted line and the solid line are the results obtained with 5 and 10 fs lasers pulses, respectively. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

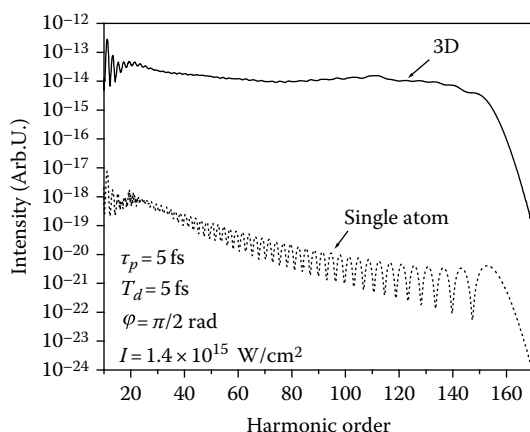


Figure 6.9 3D high-order harmonic spectra. The dotted line and the solid line are the results of the single-atom calculation and three dimensional simulations, respectively. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

Figure 6.9. Obviously, there is a drastic difference between the attosecond spectrum from the 3D propagation and that of the single atom. The intensity of the single-atom spectrum is modulated throughout the plateau and the cutoff region, whereas the macroscopic spectrum is rather smooth.

6.5.2.3 Modulation in the Single-Atom Spectrum

We focus on the single-atom XUV spectrum first. Both the separation of peaks and the modulation depth increase with frequency. To understand this feature, a square window with a width of 33 eV was applied to the XUV spectrum in Figure 6.9. The selected section of spectrum is Fourier-transformed to yield attosecond pulses in the time domain. This process is repeated for seven center frequency in the unit of NIR laser photon energy. This technique allows us to observe the variation of the emission time of attosecond pulses with the center frequency. The results are shown in Figure 6.10. For the clearness of presentation, the pulse intensities are normalized. It is clear that in the plateau region, two attosecond pulses are emitted in the laser cycle after $t = 0$. Near the cutoff, the two pulses merge to one.

Although the NIR laser pulses contain many cycles of oscillation, the fast change of ellipticity allows attosecond generation occurs only in one laser cycle. The XUV emissions in other laser cycles are suppressed by the large ellipticity of the laser field. This is the reason that there are two attosecond pulses. The one on left (emitted earlier) corresponds to the short trajectory (labeled by τ_1) and the one on the right corresponds to the long trajectory (labeled by τ_2). The modulation in the single-atom spectrum is the result of interference between the two pulses when they arrive at the same position on the detector. The spatial analogy is the Young's double-slit interference. The period of the modulation is inversely proportional to the time spacing between the two pulses, just like the relationship between the Young's fringe and the slits.

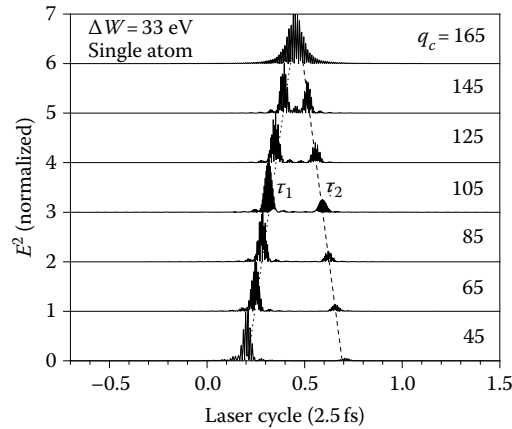


Figure 6.10 High-harmonic pulses centered at different frequencies emitted from a single atom. τ_1 and τ_2 represent the short and long trajectories, respectively. The spectrum window is 33 eV. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

The modulation depth is determined by the intensity ratio of the two interfering attosecond pulses. When the delay between the pulses from the two trajectories is comparable to the gate width (half laser cycle), the emission of the long trajectory pulse is suppressed because it falls on the edge of the gate. For pulses centered at $45\hbar\omega_1$, the ellipticity of the laser at the time when the second pulses are generated ($t = 0.7$ cycle) is significantly larger than that of the first pulses. This leads to the absence of the second pulses. The intensity of the second pulse diminishes with the decrease of central frequency. This leads to the reduction of modulation depth at low photon energy.

6.5.2.4 Comparison with the Semiclassical Results

The driving field inside the polarization gate is close to linearly polarized. Thus, the gated attosecond generation should be similar to what happens in one cycle of a perfectly linearly polarized laser field. To quantitatively compare the two cases, the classical return times of electrons with different energy gains in the laser field with $\varepsilon_x(t)$ expressed by Equation 6.122 were calculated ($\varepsilon_y(t) = 0$ was assumed, i.e., a linearly polarized field). The results are shown in Figure 6.11. In the same figure, the quantum return time in the gated case calculated with the strong field approximation is also shown, which is the time of attosecond pulse emission shown in Figure 6.10. The good agreement between the two results indicates that attosecond pulse generation process in the laser cycle around $t = 0$ is indeed similar to that in a linearly polarized field. This is an important conclusion because it provides a simple approach to understand the attosecond generation process in the somewhat complicated gating field.

6.5.2.5 Chirp of Attosecond Pulses

One can figure out the chirp of the two pulses in Figure 6.10 by connecting the emission time with straight lines. The slope of a line is

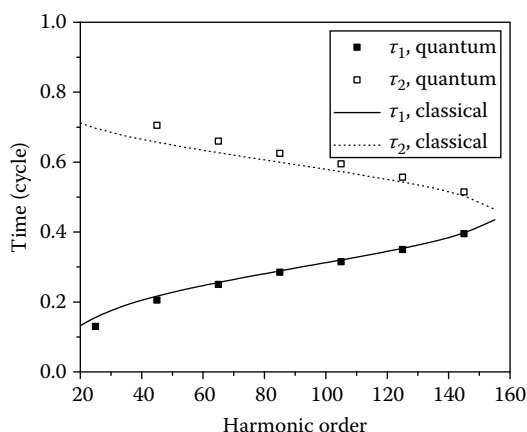


Figure 6.11 Recombination time of the electrons with short and long trajectories. Recombination in other laser cycles is suppressed by the large ellipticity of the field. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

proportional to the chirp. The sign of the chirp is different for the short and long trajectory as indicated by the two slopes. The XUV pulses corresponding to short trajectories are positively chirped, since the emission time increases with the central frequency. On the contrary, the pulses corresponding to the long trajectories are negatively chirped because the emission time decreases with the central frequency.

The XUV beam that comes out from a gas target is diverging. The divergence angle of the light from the long trajectory is larger than that of the short trajectory. The contributions from the long trajectory can be blocked by using an aperture to let only the short-trajectory beam reaching the detector. The attosecond pulses obtained including the effects of propagation are shown in Figure 6.12, which are from the short trajectory.

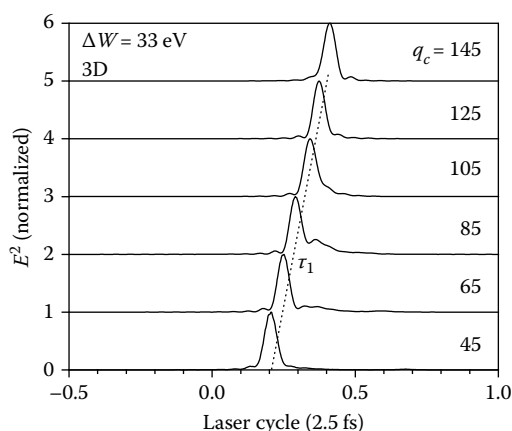


Figure 6.12 Pulses from the 3D propagation simulations. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

This explains why the modulation depth of the 3D spectrum in [Figure 6.9](#) is so much smaller.

The attosecond pulse on the detector is positively chirped. Thus, it is anticipated that the chirp can be compensated by an optical device that introduces a negative chirp in the $45\hbar\omega_1$ to $145\hbar\omega_1$ photon energy, in order to generate transform-limited attosecond pulses corresponding to a 150 eV wide spectral, which would be less than 25 as.

6.5.2.6 Chirp Compensation

Nam's group discovers that many thin-film filters exhibit negative dispersion in the XUV region. Ideally, the second and higher order phases of the filter should be the opposite of the attosecond spectral phase. The attosecond spectral phase varies with the radial position at the exit of the target, as shown in [Figure 6.13](#). The linear phases are not included in the figure because they do not affect the pulse duration. The phases are dominated by the second order terms, indicated by the quasiparabolic shapes. This means that the chirp is close to linear.

The XUV intensity decreases with the off-axis distance from the axis, as shown in [Figure 6.14](#). The chirp of the attosecond pulse at a given point on the detector is weighted by the intensity. When the phase-matching conditions are fulfilled, the major contribution comes from the region near the z axis, where the laser intensity is the highest.

To generate the shortest attosecond pulse, one should choose filters with negative GDD over a broader photon energy range. Sn is one of such materials. When metal filters such as Sn are inserted in the beam, the high-power NIR laser is blocked, preventing it from reaching the detector, which is a bonus. In [Figure 6.15](#), the on-axis attosecond spectral phase is compared with the phase of a Sn filter with 1 μm thickness. For clearness, the sign of the Sn phase is reversed. In the region between $90\hbar\omega_1$ and $130\hbar\omega_1$ photon energy, the two phases are very close. Outer side of this range, the material phase deviates from the parabolic shape,

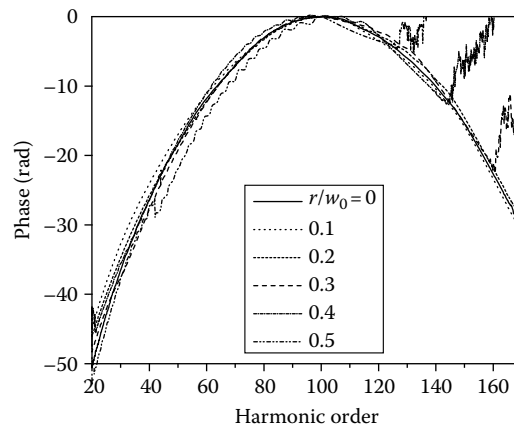


Figure 6.13 Phases of the high-harmonic field at different radial positions of the exit plane. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

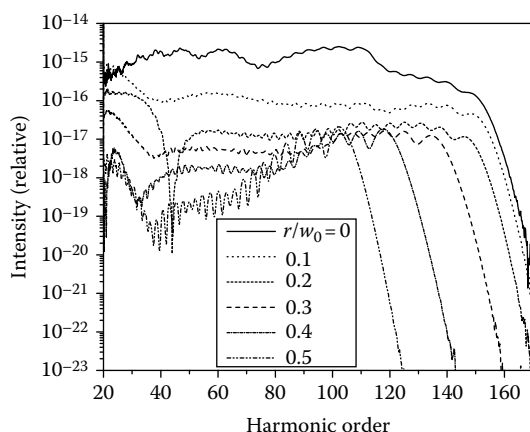


Figure 6.14 Intensities of the high-harmonic field at different radial positions of the exit plane. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

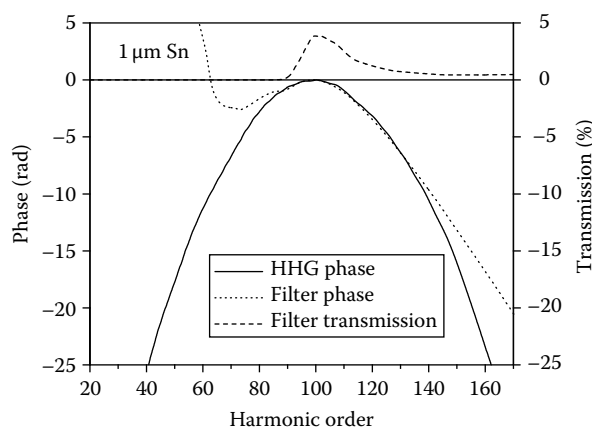


Figure 6.15 Comparison of the phases between the Sn filter and the high-harmonic field at the center of the exit plane. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

which puts a limit on the shortest attosecond that can be achieved with this filter. Finding better schemes that can compensate the chirp over a much large frequency range is an important subject in attosecond optics research.

On the lower frequency side (below $90\hbar\omega_1$), the mismatch between the two phases is not a problem since the XUV intensity there is suppressed by the filter absorption, as indicated by the filter transmission curve. The effects of mismatch on the higher frequency side (above $130\hbar\omega_1$) can be reduced by setting the cutoff frequency at the desired value. This is how the laser intensity, $1.4 \times 10^{15} \text{ W/cm}^2$, is chosen.

The attosecond XUV spectrum after a $0.8 \mu\text{m}$ thick Sn filter is shown in Figure 6.16. The FWHM is 30 eV. It covers the region where the chirp of the XUV pulse can be relatively well compensated by the filter. The

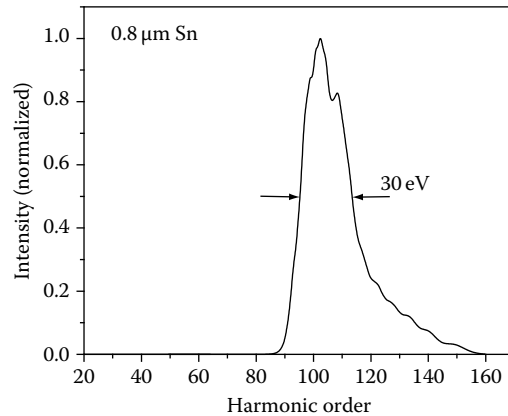


Figure 6.16 Intensity of the high-harmonic spectrum after the Sn filter. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

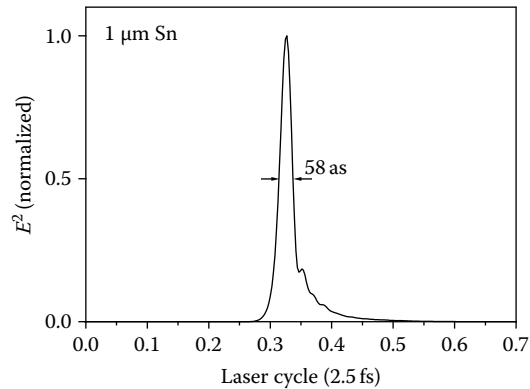


Figure 6.17 Shape of the attosecond pulse after the Sn filter. (From Z. Chang, *Phys. Rev. A*, 71, 023813, 2005. Copyright 2005 by the American Physical Society.)

isolated XUV pulse after the filter is shown in Figure 6.17 which is 58 as. The pulse has wings at the trailing edge, which is caused by mismatch of phases on the high photon energy side. The transmission of the filter is rather low (a few percent), which is a drawback of using Sn. Another issue with the Sn is that the intense spectral below $90\hbar\omega_1$ cannot be used. Nevertheless, it is interesting that the attosecond chirp can be removed by using such a simple method.

6.6 Phase Matching in Double-Optical Gating

The XUV photon flux of isolated attosecond pulses is determined by the pump laser energy and the conversion efficiency of the generation scheme. The pulse energy of the ~ 5 fs, carrier-envelope phase controlled Ti:Sapphire lasers for generating isolated attosecond pulses with polarization gating are limited to a few millijoule by the damage of the fiber and

ionization of the gas inside the fiber core, which makes it difficult to scale up the energy of the attosecond pulses. Lasers based on OPCPA (optical parametric chirped pulse amplification) may provide more powerful few-cycle pulses than hollow-core fibers, which is one approach to obtain high energy attosecond pulses. The main challenge of developing OPCPA lasers is the lack of pump lasers.

High-power (up to Petawatt) longer than 20 fs NIR pulses can be produced from chirped-pulse amplifiers. Here, we introduce another gating method, double-optical gating, for generating isolated attosecond pulse, which can be implemented with much longer laser pulses, i.e., 10–30 fs, although it also works with few-cycle lasers. With this gating scheme, single attosecond pulses can be generated directly from chirped pulse amplifiers. Consequently, the attosecond pulse flux can be scaled up with almost no limit.

6.6.1 Principle of Double-Optical Gating

Double-optical gating is a combination of polarization gating and two-color gating introduced in Chapter 4. As predicted by the three-step model, when atoms are driven by strong, linearly polarized, NIR laser pulses containing many optical cycles, a train of attosecond pulses is generated. The spacing between the two neighboring attosecond pulses is half of an optical cycle, as illustrated in the top graph of Figure 6.18. If we want to “switch out” a single pulse from the train, such as the one drawn in green color, the gate width of the fast switch should be close to the spacing, i.e., half of a laser period, which is 1.3 fs for Ti:Sapphire laser.

In the two-color gating, a weak SH field (10%–20% of the fundamental laser amplitude) is superimposed on the fundamental laser in the same polarization direction to break the symmetry of the driving field. The relative phase between the two fields is chosen to be either zero or π so that the ionization rate between the two adjacent field maxima differs by more than two orders of magnitude. In other words, electrons are essentially only ionized when the field is pointing in one direction. As a result, when the combined laser pulse is focused on a gas target, the spacing between the neighboring attosecond pulses is a full fundamental cycle, i.e., 2.6 fs

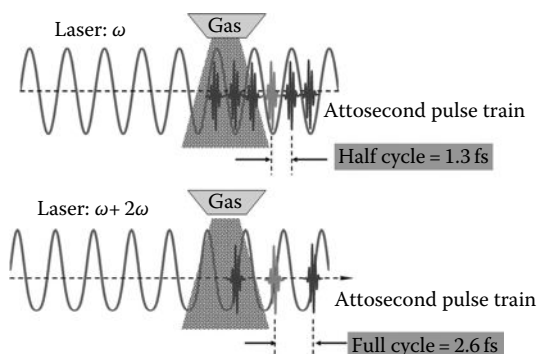


Figure 6.18 Generation of attosecond pulse train in two-color field. The spacing between two adjacent attosecond pulses is a full laser cycle.

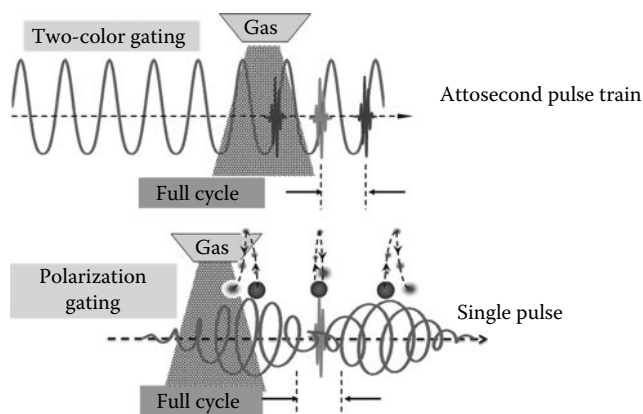


Figure 6.19 Generation of isolated attosecond pulse train by double optical gating. The gate width should be narrower than the spacing between two adjacent attosecond pulses.

for Ti:Sapphire lasers, as shown in Figure 6.18. This wide spacing makes the selection of the single pulse easier.

Polarization gating takes advantage of the findings that attosecond pulses generated by the recombination of the returning electrons are efficient when the driving laser is linearly polarized, which was first proposed by Corkum in 1994. The laser pulse used for polarization gating experiments is linearly polarized in the middle of the envelope and is elliptically polarized in the leading and trailing edges. When the width of the linearly polarized portion equals the spacing between neighboring attosecond pulses in the pulse train, only one attosecond pulse can be generated per laser shot, as shown in Figure 6.19. The width of the linear portion is defined as the width of the polarization gate.

The polarization gating pulse is from the fundamental wave, which can be resolved into two orthogonally polarized fields, where one serves as a driving field that generates the attosecond pulses while the other acts as a gating field that suppresses the attosecond emission. When a linearly polarized SH field is added to the driving field, the time interval between adjacent attosecond pulses becomes one full optical cycle of the fundamental wave. To allow one attosecond pulse emission, the width of the polarization gating should be close to one full cycle of the driving field, which is two times of what is required by the conventional polarization gating, as illustrated in Figure 6.19. The wider gate width allows the usage of longer lasers without increasing the ionization probability by the leading edge of the laser before the gate, which is the main advantage of the double-optical gating.

6.6.2 Major Factors

For given pump laser parameters and target gas pressures, the highest achievable photon flux of the attosecond pulses is determined by the balance between the coherence length, absorption length, and the media length. This has been studied extensively for high-harmonic generation (attosecond pulse train) with linearly polarized multicycle lasers. As an example, by loosely focusing the pump laser beam to a long gas cell, 25 nJ per harmonic

order at 13 nm from the neon gas were produced using 30 fs, 50 mJ lasers centered at 800 nm. For conventional high-harmonic or attosecond pulse-train generation, it is well known that phase matching is affected by the location of the gas target in the laser focus region and by the gas pressure.

There are three major differences in the phase-matching requirements between optimizing the signal of the high-harmonic signal and that of the isolated attosecond pulses.

First, for high-harmonic or attosecond pulse-train generation, since the laser intensity and the ionization of the target change from one half cycle to the next half cycle of the driving laser field, the phase-matching conditions change from one attosecond pulse to the next. It is difficult to reach perfect phase matching for all pulses in the attosecond train. A single isolated attosecond pulse is generated which half of laser cycle; therefore, one just needs to optimize phase matching in that time period.

Second, an isolated attosecond pulse is generated within one laser cycle. Thus the energy contained in other cycles of the NIR laser is wasted. Obviously, conversion efficiency is higher for shorter pulse lasers. Even for double-optical gating which works for both few-cycle and multi-cycle NIR lasers, one should use lasers with durations as short as possible if conversion efficiency is the key issue. That is one of the reasons that the experiments discussed in this section were done using 8 fs lasers.

Finally, single isolated attosecond pulses are generated when the carrier-envelope phase of the NIR laser field in the generated target is set correctly. Since the CE phase changes significantly in the Rayleigh range, the thickness of the target should be much smaller than the Rayleigh range. When the photon flux of attosecond pulses is optimized, the experimental conditions should keep the correct dependence of the harmonic spectra on the CE phase of the NIR lasers.

6.6.2.1 Intrinsic Phase of Isolated Attosecond Pulses

It is well known that the focusing geometry and the location of the generation target have a large influence on the phase matching of high-harmonic generation/attosecond pulse train through the Gouy phase and atomic dipole phase shifts. The phase matching of pulse train takes advantage of the fact that the dipole intrinsic phase, which is part of the spectral phase, depends on the driving laser intensity. Here we show that intrinsic phase of isolated attosecond pulse depends on the laser intensity in the same way as that of pulse train.

We assume that infinitely numbers of transform-limited attosecond pulses are generated by a CW driving laser. The temporal envelope $E(t)$ and phase $\phi(t)$ do not change from pulse to pulse. The electric field of the transform-limited pulse train at a given point can be expressed as

$$\varepsilon_{PT}(t) = \sum_{j=-\infty}^{+\infty} E(t - jT_{ir})e^{i[\phi(t - jT_{ir})]}, \quad (6.128)$$

where

j is the label of a particular pulse in the train

T_{ir} is spacing between two adjacent pulses in the train, which is half a driving laser period for one-color high-harmonic generation, or one fundamental laser period for two-color gating

In the frequency domain, the XUV field can be obtained by performing the Fourier transform

$$\tilde{E}_{PT}(\omega) = \int_{-\infty}^{\infty} \varepsilon_{PT}(t) e^{-i\omega t} dt, \quad (6.129)$$

which gives

$$\tilde{E}_{PT}(\omega) = \sum_{j=-\infty}^{+\infty} e^{-ij\omega T_{tr}} \int_{-\infty}^{\infty} E(t - jT_{tr}) e^{-i\omega(t - T_{tr})} d(t - T_{tr}). \quad (6.130)$$

The integral gives the electric field of one attosecond pulse in the train in the frequency domain, i.e.,

$$\tilde{E}(\omega) = \int_{-\infty}^{+\infty} E(t) e^{-i\omega t} dt. \quad (6.131)$$

Thus,

$$\tilde{E}_{PT}(\omega) = \tilde{E}(\omega) \sum_{j=-\infty}^{+\infty} e^{ij\omega T_{tr}}. \quad (6.132)$$

The sum is the Fourier series of the a Dirac comb, i.e.,

$$\tilde{E}_{PT}(\omega) = \tilde{E}(\omega) \sum_{q=-\infty}^{+\infty} \delta\left(\omega - q \frac{2\pi}{T_{tr}}\right). \quad (6.133)$$

Finally,

$$|\tilde{E}_{PT}(\omega)| e^{i\varphi(\omega)} = |\tilde{E}(\omega)| \sum_{q=-\infty}^{+\infty} \delta\left(\omega - q \frac{2\pi}{T_{tr}}\right) e^{i\varphi(\omega)}. \quad (6.134)$$

It is clear that two spectra have the same spectral phase $\varphi(\omega)$. Since the intrinsic phase is a portion of the spectral phase, we expect the intrinsic phase of one attosecond pulse to be the same as that of the pulse train at the given NIR intensity.

6.6.2.2 On-Axis Phase Matching

In isolated attosecond pulse generation, the ionization probability can be rather high ($\sim 50\%$). The dispersion of the remaining neutral gas has negligible effects on the phase matching. In this case, the spectral phase difference between the polarization wave and attosecond field is

$$\begin{aligned} \varphi_P(\omega, z) - \varphi(\omega, z) &= \frac{\alpha I_o}{1 + \left(\frac{z}{z_R}\right)^2} + \frac{\omega}{\omega_1} \tan^{-1} \left(\frac{z}{z_R}\right) \\ &+ \frac{\omega}{c} \frac{e^2}{2\epsilon_0 m_e \omega_1^2} N(z) p(z) z. \end{aligned} \quad (6.135)$$

For the attosecond emission from the short trajectory and for sufficient high frequency, the first term on the right-hand side can be dropped. Thus,

$$\varphi_P(\omega, z) - \varphi(\omega, z) \approx \frac{\omega}{\omega_1} \left[\tan^{-1} \left(\frac{z}{z_R} \right) + \frac{e^2}{2c\epsilon_0 m_e \omega_1} N(z) p(z) z \right]. \quad (6.136)$$

When

$$\frac{d}{dz} \left[\tan^{-1} \left(\frac{z}{z_R} \right) + \frac{e^2}{2c\epsilon_0 m_e \omega_1} N(z) p(z) z \right] = 0. \quad (6.137)$$

Phase matching, $\frac{d}{dz} [\varphi_P(\omega, z) - \varphi(\omega, z)] = 0$, can be achieved at any frequency ω , which is critical for generating the intense short attosecond pulse.

We can compare the effects of the plasma dispersion to that of the Gouy phase by taking the derivative of the Gouy phase at $z = z_R$:

$$\frac{d\psi}{dz} = \frac{1/z_R}{1 + \left(\frac{z}{z_R} \right)^2} = \frac{1}{2z_R}. \quad (6.138)$$

A typical value in the Rayleigh range in experiments is 2.5 mm, which yields $\frac{d\psi}{dz} \approx 0.2 \text{ mm}^{-1}$.

For $\lambda_1 = 800 \text{ nm}$, and for 1 torr uniform gas pressure, $e^2/2c\epsilon_0 m_e (N/\omega_1) = 0.074 \text{ mm}^{-1}$. Therefore, $d/dz([\Delta k(z)z]) = 0.074 P d/dz([p(z)]) \text{ mm}^{-1}$, where the unit of the pressure is in torr. At $z = z_R, d[p(z)z]/dz < 0$, then the plasma dispersion can effectively cancel the Gouy phase effects.

The quantity

$$z_R \frac{dp(z)}{dz} \Big|_{z=z_R} + p|_{p \approx 0.5} \frac{dz}{dz} \Big|_{z=z_R} = z_R \frac{dp(z)}{dI} \Big|_{p \approx 0.5} \frac{dI}{dz} \Big|_{z=z_R} + 0.5, \quad (6.139)$$

where

$$\frac{dI}{dz} \Big|_{z=z_R} = -\frac{I_0}{2} \frac{1}{z_R}. \quad (6.140)$$

It means that the laser power drops by a factor of two over a distance of z_R . So

$$z_R \frac{dp(z)}{dz} \Big|_{z=z_R} = -\frac{1}{2} \frac{dp(z)}{d(I/I_0)} \Big|_{p \approx 0.5}. \quad (6.141)$$

For Ne atom interacting with a 30 fs laser, calculations using ADK rate yield $\frac{dp(z)}{dI/I_0} \Big|_{p \approx 0.5} \approx 2 \cdot z_R \frac{dp(z)}{dz} \Big|_{z=z_R} + 0.5 \approx -0.5$. Setting

$$0.037P = \frac{1}{2z_R} = 0.2. \quad (6.142)$$

We have the phase-matching pressure $P \approx 5 \text{ torr}$. For loosely focused laser beam, the pressure is even lower.

For Ar atom in a 30 fs laser field,

$$\left. \frac{dp(z)}{d(I/I_0)} \right|_{p \approx 0.5} \approx 1.7, \quad z_R \left. \frac{dp(z)}{dz} \right|_{z=z_R} + 0.5 \approx -0.35.$$

Setting

$$0.013P = \frac{1}{2z_R} = 0.2 \text{ mm}^{-1}. \quad (6.143)$$

We have the phase-matching pressure $P \approx 15$ torr.

The length of the target should be smaller than the Rayleigh range to avoid large CE phase variation in the propagation direction introduced by the Gouy phase shift. When the target length $L = \frac{z_R}{2}$, the pressure–length product for near perfect phase matching in neon is

$$PL = 6.8 \text{ torr} \cdot \text{mm}. \quad (6.144)$$

For argon, $PL \approx 20 \text{ torr} \cdot \text{mm}$.

The phase-matching pressure–length product depends on the NIR laser pulse duration and the ionization probability inside the polarization gate.

6.6.2.3 Pressure Gradient

The existence of a maximum PL value is because the ionization probability drops too fast with z . The phase-matching pressure–length product can be increased by using a gas target where the pressure varies along the propagation direction so that

$$\begin{aligned} & \frac{d}{dz} \left[\tan^{-1} \left(\frac{z}{z_R} \right) + \frac{e^2}{2c\epsilon_0 m_e \omega_1} N(z)p(z)z \right] \\ &= \frac{\frac{1}{z_R}}{1 + \left(\frac{z}{z_R} \right)^2} + \frac{d}{dz} \left[\frac{0.074}{\text{torr} \cdot \text{mm}} P(z)p(z)z \right] = 0, \end{aligned} \quad (6.145)$$

where $N(z)$ and $P(z)$ are the distributions of the gas density and pressure, respectively. When $N(z)$ increases with z , it can effectively slow down the variation of $p(z)$.

For ionization probability close to 100%, the phase-matching conditions become

$$\frac{\frac{1}{z_R}}{1 + \left(\frac{z}{z_R} \right)^2} + \frac{0.074}{\text{torr} \cdot \text{mm}} \frac{d}{dz} [P(z)z] = 0. \quad (6.146)$$

We can introduce a function $f(z)$ to describe the normalized pressure distribution, so that

$$\frac{\frac{1}{z_R}}{1 + \left(\frac{z}{z_R} \right)^2} + \frac{0.074P_0}{\text{torr} \cdot \text{mm}} \frac{d}{dz} [f(z)z] = 0, \quad (6.147)$$

The phase-matching pressure for a target located at z is

$$P_0(\text{torr}) = -\frac{1}{z_R(\text{mm})} \frac{13.5}{1 + \left(\frac{z}{z_R}\right)^2} \frac{d}{dz} [f(z)z]. \quad (6.148)$$

Obviously, $\frac{d}{dz} [f(z)z]$ must be less than zero. By controlling the gas density distribution, phase-matching pressure can be achieved at any location and any pressure!

When the pressure is high and ionization probability is large, plasma defocus starts to change the laser beam propagation in the target. That effect must be taken into account when comparing with experimental results.

6.6.3 Experimental Results

There are many parameters for optimizing the XUV flux generated with the double-optical gating. The best target location and pressure were searched to optimize the photon flux of the XUV supercontinua. Argon and neon gases are also used in the experimental studies of phase matching of double-optical gating carried out in the author's lab.*

As compared to other gating schemes, double-optical gating works for a much broader range of NIR laser pulse durations. However, there is a tradeoff between NIR pulse duration and laser-to-XUV conversion efficiency. The shorter the pulse duration is, the higher the conversion efficiency due to the reduction of the depletion of the ground-state population. As a compromise between the XUV photon flux and the pump laser operation difficulties, 8 fs laser was used in this work. Experiments on double-optical gating with 25 fs lasers are discussed in Chapter 8.

6.3.3.1 Experimental Setup

The NIR laser pulses originate in a grating-based chirped pulse amplifier. The laser produces 30 fs, 2.5 mJ pulses centered at 800 nm at 1 kHz. The output power was stabilized during the experiments to avoid the effects of power fluctuation. The CE phase of the amplified pulse was also stabilized by feedback control of the grating separation in the chirped pulse amplifier. The output beam was sent to a hollow-core fiber filled with neon gas and chirped mirror to produce the 8 fs pulse. The linearly polarized NIR laser is centered at 790 nm with a pulse energy of 0.85 mJ. The top graph in Figure 6.20 shows a schematic of the experimental setup.

6.3.3.2 Gating Optics

Three optical components were used to transform the linearly polarized beam for the fiber to the required field for the double-optical gating, which

* The work is described in more detail in Mashiko, H., S. Gilbertson, C. Li, E. Moon, and Z. Chang, Optimizing the photon flux of double optical gated high-order harmonic spectra, *Phys. Rev. A* 77, 063423 (2008).

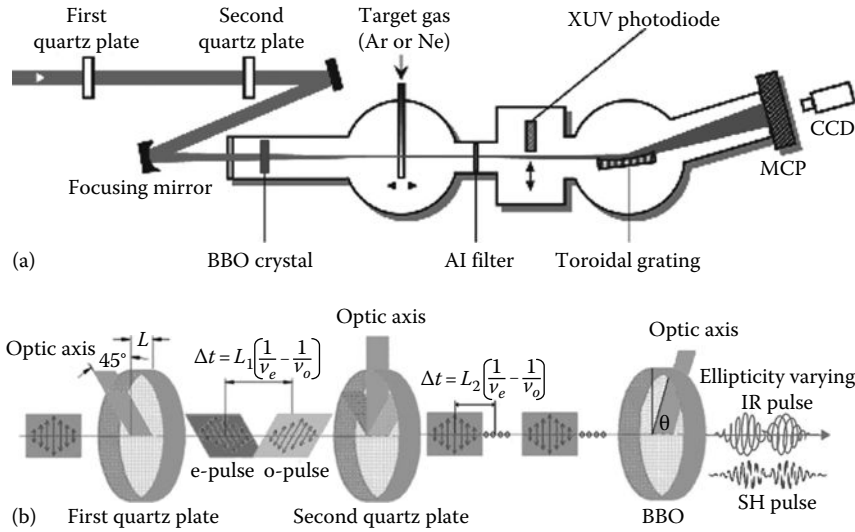


Figure 6.20 (a) Generation of isolated attosecond pulse train by double-optical gating. The gate width should be narrower than a full fundamental wave period. (b) Schematic of the double-optical gating setup for optimizing the XUV flux. (From H. Mashiko, S. Gibertson, C. Li, E. Moon, Z. Chang, *Phys. Rev. A*, 77, 063423, 2008. Copyright 2008 by the American Physical Society.)

includes two birefringent quartz plates and a barium borate (BBO) crystal. Their orientations are depicted in the bottom graph in Figure 6.20. The function of the first quartz plate is the same as in case of polarization gating. The second quartz plate and the BBO work together to serve as a quarter wave plate and convert the two orthogonally polarized pulses from the first quartz plate into a two counter rotating circularly polarized pulses for polarization gating. The 150 μm thick BBO crystal generates the SH field that is superimposed on the polarization gating field for double-optical gating. The first and second quartz plates have thicknesses of 270 and 440 μm , respectively. The first plate thickness sets the polarization gate width to 2.5 fs, i.e., one optical cycle.

A spherical mirror with focal length of 400 mm was used to focus the laser beam on the gas target. The focal spot sizes of the NIR and SH beams were 40 and 30 μm , yielding Rayleigh lengths of 6.3 and 7.2 mm, respectively. The intensity of the linearly polarized NIR beam at the focus can be calculated by using Equation 1.9:

$$I_0 = \frac{1.88}{\pi} \frac{5}{w^2 \tau} = \frac{1.88 \times 0.85 \times 10^{-3}}{\pi (40 \times 10^{-4})^2 \times 8 \times 10^{-15}} = 4 \times 10^{15} \frac{\text{W}}{\text{cm}^2}. \quad (6.149)$$

The peak intensities of the linear polarized portion of the NIR can be estimated using the intensity form of Equation 4.168,

$$I(t=0) = I_0 e^{-\ln 2 \left(\frac{t_d}{\tau}\right)^2} \approx 4 \times 10^{15} \times e^{-\ln 2 \left(\frac{8}{8}\right)^2} = 2 \times 10^{15} \frac{\text{W}}{\text{cm}^2}. \quad (6.150)$$

The peak intensity of the SH pulses at the focus was estimated to be $1.7 \times 10^{14} \text{ W/cm}^2$.

The gas cell length is 1.4 mm, which is much smaller than the Rayleigh range to minimize the CE phase change in the target. The intensity at the target position is lower than this value because it is placed after the laser focus. When the distance of the target to the focal point equals the Rayleigh range where the phase matching is close to the best, the intensity is half that at the focus, $\sim 1 \times 10^{15}$ W/cm². Such a high field will fully ionize argon atom, an iris in front of the focusing mirror is used to control intensity.

The generated XUV light were sent to an aluminum filter with thickness of 200 nm to block the laser light. The XUV photon flux was measured by a Si photodiode. The XUV spectra were measured by a grating spectrometer.

6.6.4 Gas-Target Location

For the given NIR laser energy and focal spot size, the dependence of the flux on the gas-target location is examined first to find the best location of the gas cell within the confocal range of the fundamental laser.

6.6.4.1 Argon Gas

Figure 6.21a shows the XUV spectra with the double-optical gating at different location. The gas cell is filled with 90 mb of argon gas. The location of the cell in front of the NIR focus corresponds to the negative sign. The XUV spectrum intensity is stronger when the target is located after the laser focus, as the theory predicted. It reaches the maximum at 2.5 mm after the laser focus.

Figure 6.21b shows the XUV photon flux measured by the XUV photodiode located after an aluminum filter. The maximum XUV pulse energy from argon is 6.5 nJ.

6.6.4.2 Neon Gas

Figure 6.22a shows the XUV spectra that vary with the location of the cell filled with neon gas (the backing pressure is ~ 40 mb). The brightest XUV is generated at 3 mm after the focus. Figure 6.22b is the photon flux before

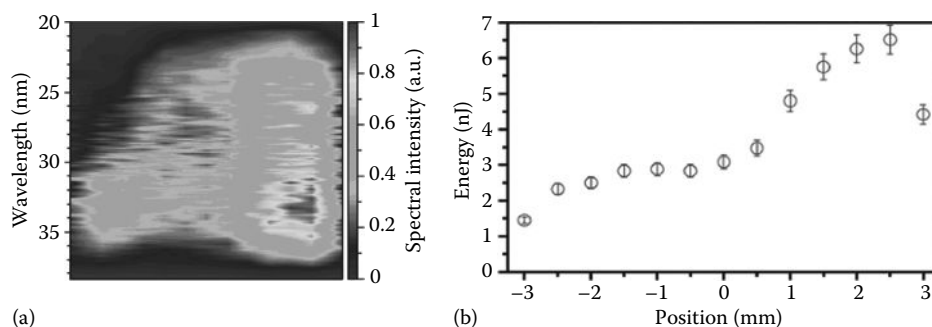


Figure 6.21 Comparison of double optical-gated high-harmonic spectra at several locations within laser focus with an argon target gas. (a) The XUV spectrum images. (b) The open circles are pulse energy before aluminum filter. (From H. Mashiko, S. Gibertson, C. Li, E. Moon, Z. Chang, *Phys. Rev. A*, 77, 063423, 2008. Copyright 2008 by the American Physical Society.)

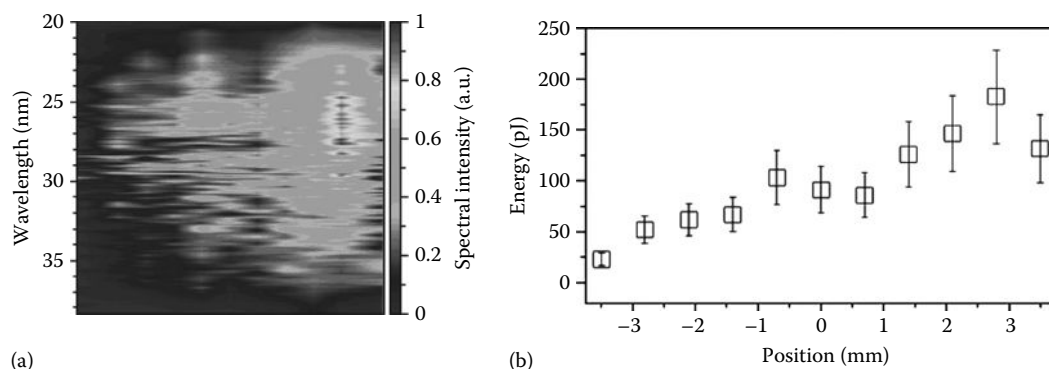


Figure 6.22 Comparison of double optical-gated harmonic spectra at several locations within laser focus with a neon target gas. (a) The harmonic spectrum images. (b) The open squares are pulse energy before aluminum filter. (From H. Mashiko, S. Gibertson, C. Li, E. Moon, Z. Chang, *Phys. Rev. A*, 77, 063423, 2008. Copyright 2008 by the American Physical Society.)

the filter as a function of the target location. Like in the case of argon, it is also higher when the target is placed after the NIR focus.

6.6.5 Gas Pressure

6.6.5.1 Argon Gas

The double-optically gated XUV spectra in the pressure range up to 135 mb are illustrated in Figure 6.23a. The gas cell was kept at a location 2.5 mm after the laser focus. The optimal signal strength occurs near 90 mb in backing pressure, or 126 mbar·mm of pressure–length product. The absorption length–pressure products for the 30 and 35 nm light is 70 and 20 mbar·mm, respectively. It can be seen that the optimum pressure is lower for the longer wavelength.

Figure 6.23b shows the results of the attosecond photon flux as a function of the gas pressure. Under the optimal condition, the pulse energy (open circles) was ~ 6.5 nJ before the aluminum filter as measured with the XUV photodiode. In the Si photodiode, one XUV photon produces several electron–hole pairs. The quantum efficiency is calibrated at synchrotron facilities. The total electric charges are measured with an oscilloscope, which gives the number of XUV photons, as discussed in Chapter 1.

The solid line shows the transmission of residual gas between the cell and aluminum filter (length is 241 mm) inside the chamber, which is higher than the transmission inside the cell (dashed line). The estimated coherence length was found to be 10 times longer than the absorption length, indicating the phase matching is nearly optimized for argon.

This maximum energy, 6.5 nJ, corresponds to an estimated photon number of 9×10^8 per pulse and a conversion efficiency of 6×10^{-6} . This is rather high considering that only one cycle of the NIR laser contributes to the generation of the XUV continuum. The conversion efficiency from the NIR energy in one cycle to the XUV is as high as 4×10^{-5} ! The residual gas in the chamber can be reduced further by using better vacuum pumps, which would allow further increase of the XUV flux.

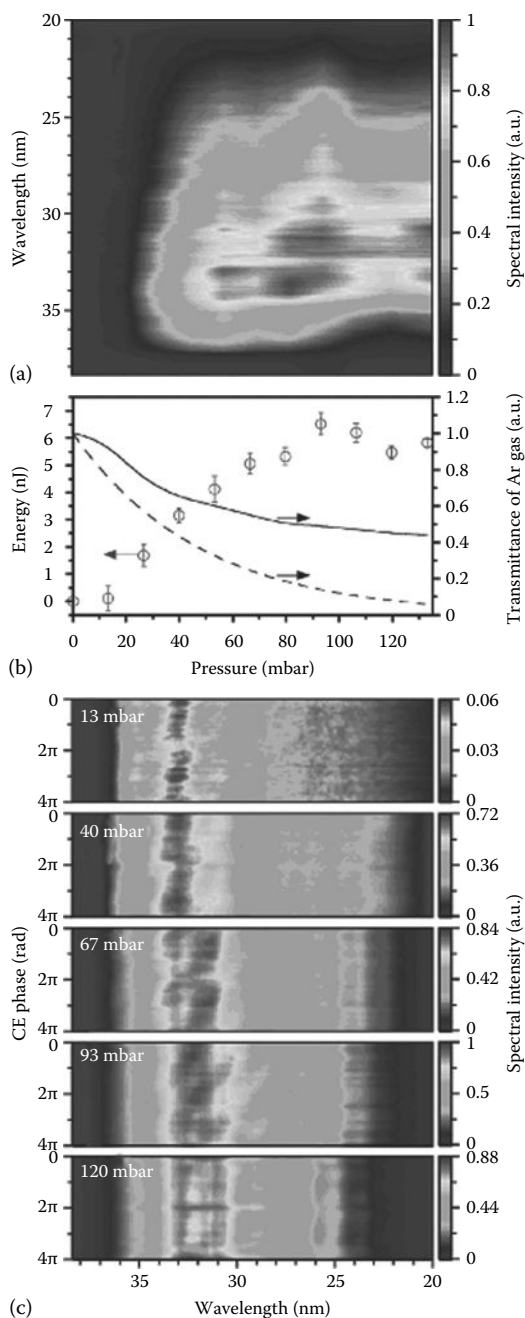


Figure 6.23 Comparison of double optical-gated harmonic spectra at several pressures of argon target gas. (a) The harmonic spectrum images. (b) The open circles are pulse energy before aluminum filter. The solid and dashed lines correspond to the absorption by residual gas and plasma in the chamber and the target cell, respectively. (c) The dependence of harmonic spectra with CE phase in pressure range of 13–120 mb of our backing pressure. (From H. Mashiko, S. Gibertson, C. Li, E. Moon, Z. Chang, *Phys. Rev. A*, 77, 063423, 2008. Copyright 2008 by the American Physical Society.)

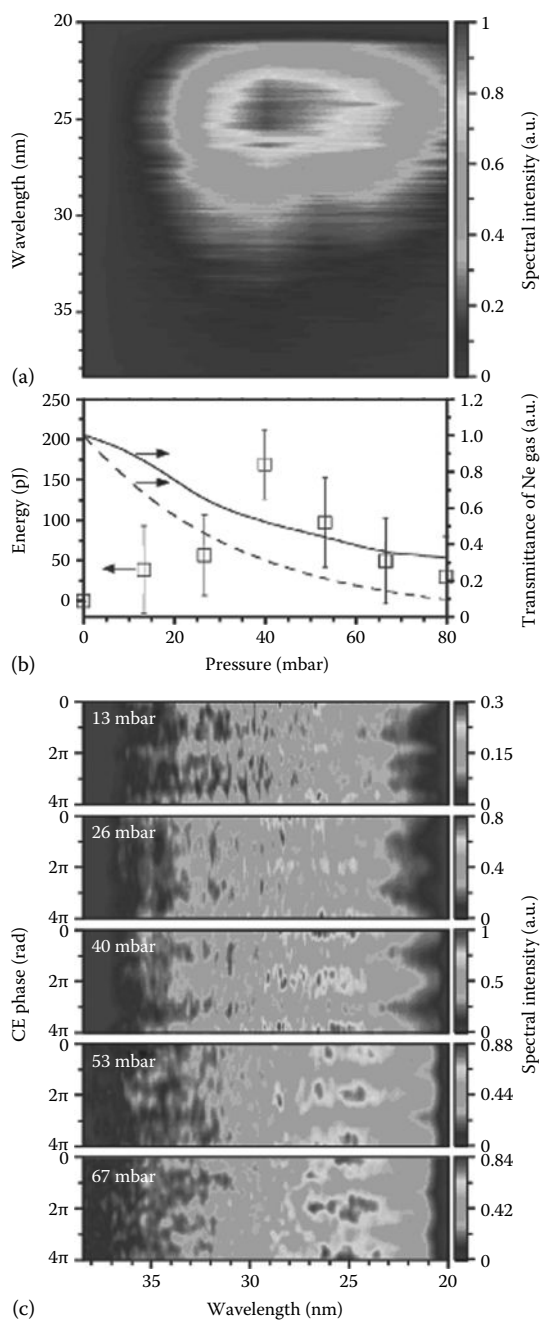


Figure 6.24 Comparison of double optical-gated harmonic spectra with several pressures of neon gas. (a) The harmonic spectrum images. (b) The open squares are pulse energy before aluminum filter. The solid and dashed lines correspond to the absorption by residual gas and plasma in the chamber and the target cell, respectively. (c) The dependence of harmonic spectra with CE phase in pressure range of 13–67 mb. (From H. Mashiko, S. Gibertson, C. Li, E. Moon, Z. Chang, *Phys. Rev. A*, 77, 063423, 2008. Copyright 2008 by the American Physical Society.)

The effect of the CE phase on the XUV spectra under different target gas pressures is shown in Figure 6.23c. The 2π periodicity provides evidence that the spectra are generated in a narrow gate close to one laser cycle by the double-optical gating, as discussed in Chapter 8. In the optimum pressure, the dependence of the spectrum on the CE phase is very strong, which indicates that isolated attosecond pulse can be generated with 6.5 nJ.

6.6.5.2 Neon Gas

The measured XUV spectra from the double-optical gating in the 0–80 mb are shown in Figure 6.24. The gas cell was located 3 mm after the laser focus. There are three important facts.

First, the spectrum is a smooth continuum in the whole pressure range, as shown in Figure 6.24a, indicating that the double-optical gating is robust against target pressure. The spectrum is broader than argon. The spectrum cutoff at the short wavelength side is due to the measurement range of the detector.

Second, there exists an optimal pressure, 40 mb, where the spectrum intensity is the highest. This is also seen in Figure 6.24b where the XUV pulse energy measured by using the x-ray photodiode is plotted. The maximum pulse energy achieved was 170 pJ before the aluminum filter, corresponding to 2×10^7 photons per pulse. The conversion efficiency is 2×10^{-7} , which is much lower than Ar.

Above the optimal pressure, the XUV pulse decreases with the pressure due the strong absorption by the neon gas. The absorption from the chamber and the gas cell is shown with the solid and dashed lines, respectively. The estimated coherence length is 1.5 times longer than the absorption length.

Finally, as Figure 6.24c shows, the XUV spectral intensity shows a 2π periodicity with the CE phase of the NIR laser at all five gas pressures tested, which is the another evidence that the double-optical gating is robust against gas pressure.

6.7 Summary

Phase matching in attosecond XUV pulse generation is very different from that of SH generation in nonlinear crystal. Instead of relying on birefringence effect, the so called intrinsic phase and the plasma density variation along the propagation direction play important roles to achieve attosecond phase matching. Unlike nonlinear crystals what are transparent in the applied wavelength range, absorption of gas is a factor that may limit the conversion efficiency in attosecond generation.

Problems

- 6.1 Consider the semiclassical model. Can one atom in the strong laser field emit two XUV photons?
- 6.2 Can Maxwell's equations be derived?

- 6.3** Explain the physical meaning of each of the Maxwell's equations.
- 6.4** Are the quantities in the Maxwell's equations real or imaginary?
- 6.5** How many unknown quantities are there in the Maxwell's equations in 3D space?
- 6.6** Give the explicit expression of the transverse part of the Laplace operator, ∇_T^2 , in the Cartesian coordinates.
- 6.7** Summarize all the approximations that lead to Equation 6.19.
- 6.8** Describe the differences between the quantities: $\vec{\mathcal{E}}(\vec{r}, t)$, $\vec{\mathcal{E}}_q(\vec{r}, t)$, $E_q(\vec{r})$, and $\tilde{A}_q(\vec{r})$.
- 6.9** Calculate the plasma frequency for the electron density $N_e = 10^{17}/\text{cm}^3$. Compare it with the frequency of the laser centered at 800 nm.
- 6.10** For the parameters of Problem 6.9, calculate the phase velocities of laser and its 30th harmonics.
- 6.11** Fill in the missing numbers in [Table 6.1](#).
- 6.12** For plane waves, show that the phase-matching condition is met when $n_R(\omega_0) = n_R(\omega_q)$.
- 6.13** Plot the absorption length–pressure product for (a) xenon and (b) helium gas in the 25–200 eV photon energy range.
- 6.14** Plot the normalized harmonic intensity (the Maker Fringe) as a function of $\frac{LP}{(LP)_c}$ using Equation 6.37. Do you see the fringe?
- 6.15** How is phase matching achieved in SH generation of NIR lasers in nonlinear optical crystals? Can we apply the same technique in phase matching of high-order harmonic generation?
- 6.16** What is the physical meaning of the retarded coordinate frame, $z' = z$, $t' = t - \frac{z}{c}$?
- 6.17** Examine whether $\mathcal{E}(z', t') = e^{-at'^2} e^{i\omega_0 t'}$ is a solution of Equation 6.85.
- 6.18** How would the flux of the attosecond pulse scale with the laser pulse energy if the phase-matching condition does not change?

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Attosecond Pulse Trains

Soon after high-order harmonic generation was discovered in late 1980s, an argument began that the harmonics in the plateau region could be an attosecond pulse train in the time domain. However, it was not until 2001 that the confirmation of this hypothesis was demonstrated. Although the generation of the attosecond pulse train is straightforward, the determination of the number of pulses, the pulse duration, and the phase is rather difficult. In this chapter, we focus on methods for characterizing attosecond pulse trains.

Before the discovery of high-harmonic generation in the XUV range, femtosecond metrology using FROG and SPIDER had already been developed for completely characterization of the temporal structure of ultrashort light pulses in the near infrared (NIR), visible and even ultraviolet spectral range. Such a complete characterization of ultrashort laser pulses with a slow detector requires the use of a time-nonstationary filter, e.g., an amplitude or phase gate in the time domain. The amplitude or phase modulation occurs in a fraction of the pulse to be characterized. In many schemes, the unknown pulse itself is turned into a time-nonstationary filter through the use of a nonlinear effect such as second harmonic generation.

Because attosecond light pulses are in the XUV range and have only reached moderate intensities so far, using nonlinear effects for their characterization is challenging. It is also difficult to rapidly modulate either the amplitude or the phase of the XUV pulse to serve as the nonstationary filter. Most attosecond measurement methods developed so far rely on a different approach. Attosecond XUV fields can efficiently ionize atoms by single-photon absorption. This ionization generates an attosecond electron pulse, which is a replica of the attosecond pulse: the phase and amplitude of the XUV field are transferred to the photoelectron wave packet. Direct information on the temporal structure of the XUV field can therefore be obtained by characterizing this electron wave packet. In other words, the electron wave packet serves as an intermediate tool. It turns out that NIR laser fields acting on attosecond electron pulses constitute ideal time-nonstationary filters. The acceleration and deceleration of the electron by an intense laser field essentially acts as an ultrafast phase modulator on the electron wave packet. Although the NIR pulse duration is much longer than the attosecond XUV pulse, the phase

modulation occurs in a small fraction of an NIR cycle, which leads to a significant change of phase of the electron wave during the time that attosecond pulse last.

This idea works for characterizing both attosecond pulse trains and single isolated attosecond pulses. The experimental setup is identical to the attosecond streak camera introduced in Chapter 1. We first discuss some unique beam properties of the NIR laser for the electron phase modulator.

7.1 Truncated Gaussian Beam

In the measurement of attosecond XUV pulses, the phase of the photoelectron wave packet produced by the XUV light is modulated by an NIR laser field. This requires the XUV and NIR beams to co-propagate and overlap on the detection gas target. The two beams must be collinear to avoid introducing a time delay in the transverse direction of the beam.

Due to the lack of a dichroic beam splitter to combine the NIR beam with the XUV beam, a mirror with a hole is commonly used as the beam combining optics. The XUV beam travels through the hole and NIR beam is reflected by the mirror, as shown in Figure 7.1. Both beams are focused to the detection gas target. At the focal spot of the NIR beam, the XUV is also focused. It is desirable to have the photoelectron generated by the XUV exposed to a uniform NIR field so that the phase modulation is the same for all photoelectrons. In reality, the NIR field at the focus changes in both the transverse and longitudinal directions. It is therefore necessary to find out how the field varies for precise measurement of the attosecond pulses.

7.1.1 Electric Field

The NIR laser reflected from the hole-drilled mirror is not a Gaussian beam anymore because the center portion is missing. Consequently, the equations given in Chapter 2 on describing the electric-field distribution in the focal region are not valid. In general, we can use an annular aperture to

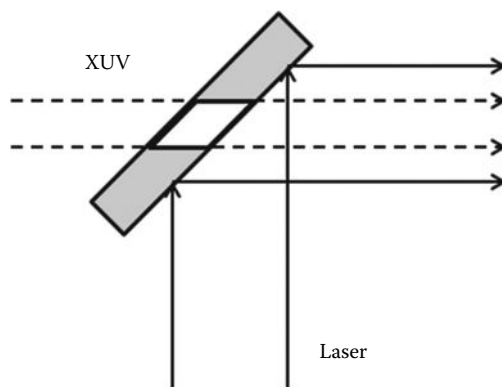


Figure 7.1 Combining NIR and XUV beam collinearly with a holed mirror.

study the truncated Gaussian beam. Suppose ρ_i and ρ_o are the inner and outer radius of an annular aperture, respectively. We can consider the case where a collimated Gaussian beam is focused by an ideal thin lens with a focal length of f , as shown in Figure 7.2. Experimentally, a toroidal mirror (the surface of mirror is torus which has two curvatures) or a spherical mirror can also be used to focus the NIR beam, but the conclusions obtained with the lens are valid for the mirrors.

For mathematic convenience, we place the lens before the annular aperture. In the cylindrical coordinate system shown in Figure 7.3, the transverse profile of the collimated laser field on the input surface of the lens can be written as

$$u_1(\rho) = u_0 e^{-\left(\frac{\rho}{w_G}\right)^2}, \quad (7.1)$$

where

- ρ is the radius in the aperture plane
- w_G is the radius of the beam
- u_0 is the on-axis field amplitude

Since the beam is collimated, the transverse phase is flat.

Right out of the lens, the field becomes

$$u_2(\rho) = u_1(\rho) e^{-ik\frac{\rho^2}{2f}}, \quad (7.2)$$

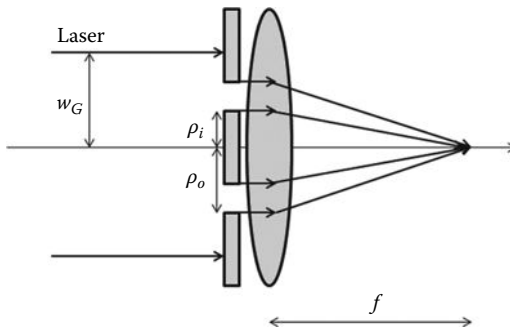


Figure 7.2 Focus of a truncated Gaussian beam.

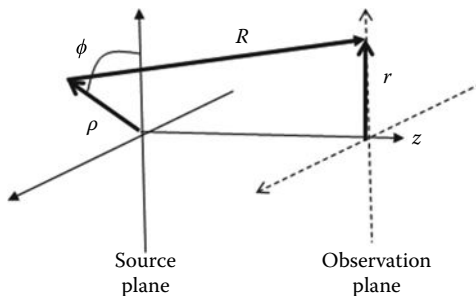


Figure 7.3 Coordinate system for the Fresnel integral.

where the propagation constant $k = 2\pi/\lambda$. λ is the wavelength of the monochromatic NIR beam. Equation 7.2 shows that a wavefront curvature with radius f is introduced by the lens.

After the aperture, the field can be calculated by the Fresnel diffraction integral

$$u_3(r, z) = \left(-\frac{i}{\lambda}\right) \int_{\rho_i}^{\rho_o} \int_0^{2\pi} \frac{u_2(\rho)}{R} e^{-ikR} \rho d\rho d\phi, \quad (7.3)$$

where

R is the distance between the source point (the annular aperture) and the observation point (the focal region)

ϕ is the azimuth angle at the source plane, as shown in Figure 7.3.

Under the Fresnel approximation, R in the denominator is replaced by z . The R in the exponent is approximated by

$$R = z - \frac{1}{2z}(r^2 + \rho^2 + 2r\rho \cos \phi). \quad (7.4)$$

Thus, the field at the observation point becomes

$$u_3(r, z) = \left(-\frac{i}{\lambda}\right) \int_{\rho_i}^{\rho_o} \int_0^{2\pi} \frac{u_2(\rho)}{z} e^{-ikz} e^{ik\frac{r^2}{2z}} e^{ik\frac{\rho^2}{2z}} e^{ik\frac{r\rho}{z} \cos \phi} \rho d\rho d\phi \quad (7.5)$$

or

$$u_3(r, z) = \left(-\frac{i}{\lambda}\right) e^{-ikz} e^{ik\frac{r^2}{2z}} \frac{1}{z} \int_{\rho_i}^{\rho_o} \int_0^{2\pi} u_2(\rho) e^{ik\frac{\rho^2}{2z}} e^{ik\frac{r\rho}{z} \cos \phi} \rho d\rho d\phi. \quad (7.6)$$

Next, we can use the definition of the Bessel function

$$J_0(v) = \frac{1}{2\pi} \int_0^{2\pi} e^{iv \cos(s)} ds. \quad (7.7)$$

and omit the fast variation phase term $e^{-ikz} e^{ik(r^2/2z)}$ to arrive at

$$u_3(r, z) = \left(-\frac{i}{\lambda}\right) \frac{2\pi}{z} \int_{\rho_i}^{\rho_o} u_2(\rho) e^{ik\frac{\rho^2}{2z}} J_0\left(\frac{k\rho r}{z}\right) \rho d\rho. \quad (7.8)$$

For the Gaussian input, this becomes

$$u_3(r, z) = \left(-\frac{i}{\lambda}\right) u_0 \frac{2\pi}{z} \int_{\rho_i}^{\rho_o} e^{-\left(\frac{\rho}{w_G}\right)^2} e^{-ik\frac{\rho^2}{2z}} e^{ik\frac{\rho^2}{2z}} J_0\left(\frac{k\rho r}{z}\right) \rho d\rho. \quad (7.9)$$

7.1.1.1 Bessel Functions

There are similarities between Bessel functions and sinusoidal functions, as shown in Figures 7.4 and 7.5. $J_0(v)$ is the zeroth-order Bessel function

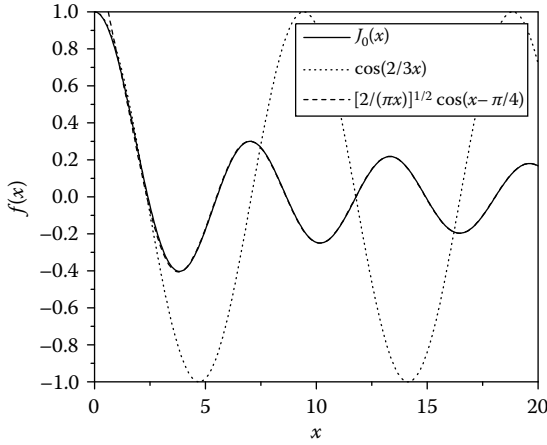


Figure 7.4 Comparison between $J_0(x)$ functions and cosine functions.

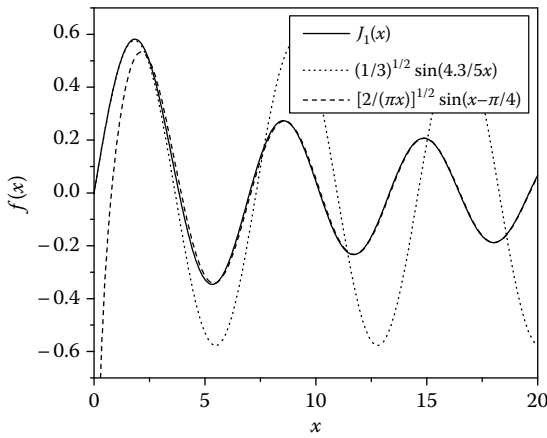


Figure 7.5 Comparison between $J_1(x)$ functions and sine functions.

of the first kind, which is similar to an amplitude damped cosine function. In fact, it can be approximated by a cosine function as

$$J_0(v) \approx \cos\left(\frac{2}{3}v\right), \quad v < 2 \quad (7.10)$$

and

$$J_0(v) \approx \sqrt{\frac{2}{\pi v}} \cos\left(v - \frac{\pi}{4}\right), \quad v > 2. \quad (7.11)$$

Similarly, the first-order Bessel function of the first kind can be approximated by a sine function

$$J_1(v) \approx \sqrt{\frac{1}{3}} \sin\left(\frac{4.3}{5}v\right), \quad v < 3 \quad (7.12)$$

and

$$J_1(v) \approx \sqrt{\frac{2}{\pi v}} \sin\left(v - \frac{\pi}{4}\right), \quad v > 3. \quad (7.13)$$

7.1.1.2 Narrow Annular Aperture

The field can also be expressed as

$$u_3(r, z) = \left(-\frac{i}{\lambda}\right) u_0 \frac{2\pi}{z} \int_{\rho_i}^{\rho_o} d\rho \rho J_0\left(k\rho \frac{r}{z}\right) e^{-ik\frac{\rho^2}{2q(z)}}, \quad (7.14)$$

where

$$\frac{1}{\tilde{q}(z)} \equiv \frac{1}{\frac{z}{1 - z/f}} - i \frac{\lambda}{\pi w_G^2}. \quad (7.15)$$

If $\rho_o - \rho_i \ll w_G$ is satisfied (the conditions for a narrow annular aperture), then

$$u_3(r, z) = \left(-\frac{i}{\lambda}\right) u_0 \frac{2\pi}{z} e^{-i\frac{k}{2q(z)}\left(\frac{\rho_o + \rho_i}{2}\right)^2} \int_{\rho_i}^{\rho_o} \rho J_0\left(k\rho \frac{r}{z}\right) d\rho. \quad (7.16)$$

From the Bessel function's recurrence relation we know

$$\frac{d}{dv} [vJ_1(v)] = vJ_0(v), \quad (7.17)$$

which is similar to the derivative property of the sinusoidal function $d/dv [\sin(v)] = \cos(v)$. From Equation 7.17, we have

$$\int_0^v v' J_0(v') dv' = vJ_1(v). \quad (7.18)$$

Thus, the integral

$$\begin{aligned} \int_{\rho_i}^{\rho_o} \rho J_0\left(k\rho \frac{r}{z}\right) d\rho &= \left(\frac{z}{kr}\right)^2 \int_{\rho_i}^{\rho_o} \left(k\frac{r}{z}\rho\right) J_0\left(k\frac{r}{z}\rho\right) d\left(k\frac{r}{z}\rho\right) \\ &= \frac{z}{kr} \left[\rho_o J_1\left(\frac{kr\rho_o}{z}\right) - \rho_i J_1\left(\frac{kr\rho_i}{z}\right) \right]. \end{aligned} \quad (7.19)$$

Finally, we can substitute these results into Equation 7.16 and we arrive at

$$\begin{aligned} u_3(r, z) &\approx \left(-\frac{i}{2\pi}\right) u_0 e^{-i\frac{k}{2q(z)}\left(\frac{\rho_o + \rho_i}{2}\right)^2} \frac{k}{2z} \\ &\times \left[\rho_o^2 \frac{2J_1\left(\frac{kr\rho_o}{z}\right)}{\frac{kr\rho_o}{z}} - \rho_i^2 \frac{2J_1\left(\frac{kr\rho_i}{z}\right)}{\frac{kr\rho_i}{z}} \right]. \end{aligned} \quad (7.20)$$

The functions $[2J_1(k\rho_0 r/z)]/[k\rho_0 r/z]$ and $[2J_1(k\rho_i r/z)]/[k\rho_i r/z]$ express the airy disks corresponding to a circular aperture with radii ρ_0 and ρ_i , respectively. Thus, the beam can be considered as the superposition of two airy disks.

7.1.1.3 On Axis

For $r = 0$, $J_0(k\rho(r/z)) = 1$, we have

$$\begin{aligned} u_3(0, z) &= u_0 \left(-i \frac{1}{\lambda z} \right) \int_{\rho_i}^{\rho_o} d\rho \rho e^{-ik \frac{\rho^2}{2q(z)}} \\ &= u_0 \left(-i \frac{1}{\lambda z} \right) \frac{2\tilde{q}(z)}{k} \left(e^{-ik \frac{\rho_o^2}{2q(z)}} - e^{-ik \frac{\rho_i^2}{2q(z)}} \right). \end{aligned} \quad (7.21)$$

In general, it turns out that an approximation of the field expression near the focus is

$$\begin{aligned} u_3(r, z) &\approx -i \frac{u_0}{2\pi} \frac{\tilde{q}(z)}{z} \left[e^{-ik \frac{\rho_o^2}{2q(z)}} - e^{-ik \frac{\rho_i^2}{2q(z)}} \right] \\ &\quad \times \left[\rho_o^2 \frac{2J_1\left(\frac{k\rho_o r}{z}\right)}{\frac{k\rho_o r}{z}} - \rho_i^2 \frac{2J_1\left(\frac{k\rho_i r}{z}\right)}{\frac{k\rho_i r}{z}} \right]. \end{aligned} \quad (7.22)$$

7.1.2 Transverse Variation

At the geometric focus toward $z=f$, the transverse field distributions becomes

$$u_3(r, z) = u_0 \frac{2\pi}{f} \int_{\rho_i}^{\rho_o} e^{-\left(\frac{\rho}{w_G}\right)^2} J_0\left(\frac{k\rho r}{z}\right) \rho d\rho. \quad (7.23)$$

The electric field expressed by Equation 7.23 is similar to a Bessel beam. As an example, a laser beam with a wavelength of $\lambda = 0.8 \mu\text{m}$ and a beam radius of $w_G = 8 \text{ mm}$ is focused by a lens with a focal length of $f = 250 \text{ mm}$. The inner and out radii of the annular aperture are $\rho_i = 9.35 \text{ mm}$ and $\rho_o = 25 \text{ mm}$, respectively. The electric field at the geometric focus is shown in Figure 7.6. The radius of the center spot is only $\sim 6 \mu\text{m}$. It is worthwhile to point out that the direction of the electric in the first ring of the Bessel beam is opposite to that of the center spot, which is shown by the sign change in Figure 7.6. For attosecond streaking, it is crucial to confine the XUV spot within the center NIR focal spot, which means that the XUV spot should be smaller than $6 \mu\text{m}$ for the given configuration. If a portion of the XUV beam is in the first Bessel ring, the photoelectron produced by the XUV light will be streaked to opposite direction, which affects the precision of the pulse characterization. The experimental observation of the reversal of the streaking direction is illustrated in Chapter 9.

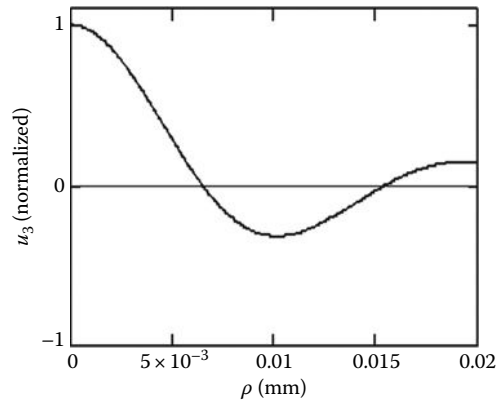


Figure 7.6 Electric field at the focus of the truncated Gaussian beam.

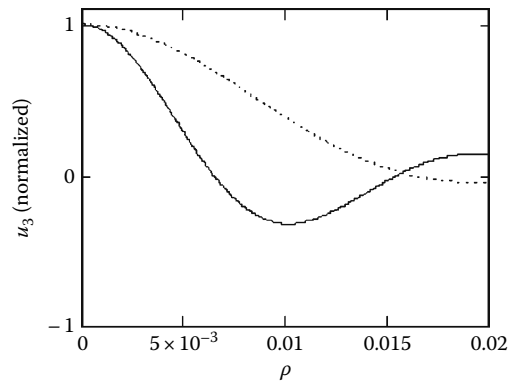


Figure 7.7 Electric-field comparisons in the transverse direction. Dotted line: circular aperture. Solid line: annular aperture.

Figure 7.7 shows the comparison of the electric fields from a circular aperture with a radius ρ_i (dotted line) and the annular aperture (solid line). The focal spot of the former is much larger.

7.1.3 Field Distribution in the Propagation Direction

For an on-axis point, since $J_0(0) = 1$,

$$u_3(0, z) \approx -i \frac{u_0}{2\pi} \frac{\tilde{q}(z)}{z} \left[e^{-ik \frac{\rho_o^2}{2q(z)}} - e^{-ik \frac{\rho_i^2}{2q(z)}} \right]. \quad (7.24)$$

Using the parameters from the previous example, the calculated field distribution in the propagation direction for the annular aperture is shown in Figure 7.8. The field strength changes significantly in 1 mm. Thus, the gas target should be much shorter than 1 mm so that the phase modulation does not change with for photoelectrons originated from different longitudinal positions.

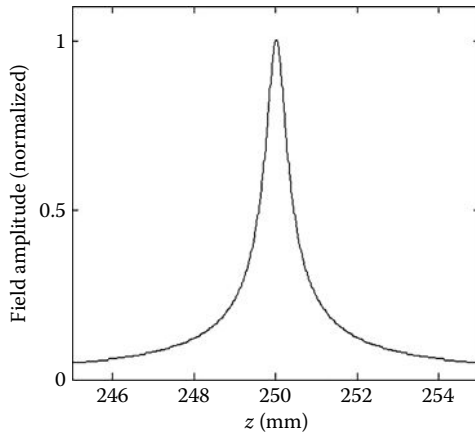


Figure 7.8 Electric-field comparisons in the propagation direction.

7.1.3.1 Gouy Phase

Like in the case of Gaussian beam propagation, the phase variation in the propagation direction in the focal region is called the Gouy phase. For attosecond streaking, the detection gas target should be confined to a region where the phase variation in the propagation direction is much less than π rad, otherwise the direction of the electron streaking on one side of focus is opposite to that of the other side. The phase can be calculated by

$$\Phi(z) = a \tan \left[\frac{\text{Im}(u_3(0,z))}{\text{Re}(u_3(0,z))} \right], \quad (7.25)$$

where the functions $\text{Im}(u)$ and $\text{Re}(u)$ yield the imaginary and real part of u .

Using the parameters in the previous example, the calculated Gouy phase shift is shown in Figure 7.9. It changes from zero to π rad in 0.4 mm. Thus, the detection gas target must be much smaller than this value.

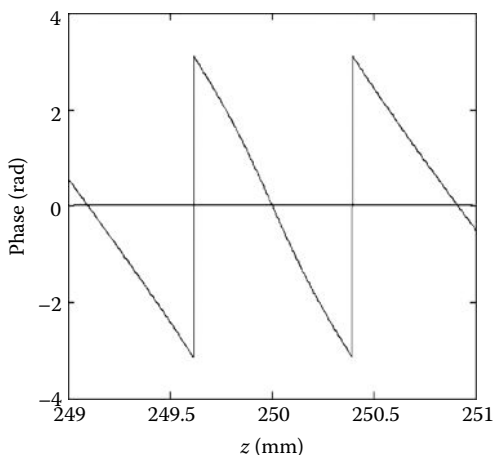


Figure 7.9 Phase variation in the propagation direction near the focus.

7.2 Detection Gas

The electron replica of the attosecond XUV pulses is created in the detection gas through photoionization. Due to the low XUV flux, the detection gas should have large absorption cross section in the XUV wavelength range to produce as many photoelectrons as possible to ease the electron detection. The ionization potential should be large enough so that the number of electrons generated by the NIR laser field through above-threshold ionization is much lower than the photoelectrons produced by the XUV light. As such, noble gases are commonly used as targets. Figure 7.10 shows the scattering factor f_2 for five types of atom, which is proportional to the photoionization cross section by

$$\sigma(\lambda_x) = 2r_e\lambda_x f_2, \quad (7.26)$$

where

r_e is the classical electron radius

λ_x is the wavelength of the XUV light

The cross sections shown here do not include the contributions of auto-ionization. The study of autoionization by using attosecond pulses is described in Chapter 9.

The absorption probability can be calculated by $1 - e^{-N\sigma(\lambda_x)L}$. Here, N is the gas density and L is the length of the target, which should be comparable to the NIR laser spot size that is on the order of 20 μm .

7.2.1 Effects of Spin–Orbit Coupling and Inner Shells

Except for helium, the outmost shell has two sub-shells due to the spin–orbit coupling. Their ionization potentials are 12.129 eV ($J=3/2$) and 13.436 eV ($J=1/2$), respectively, for xenon. Thus, photoelectrons with two different energy are separated by 1.3 eV, which set the energy resolution of the spectrum measurement.

The energy differences for Kr and Ar are 0.7 and 0.18 eV, respectively. The bind energy of the third level (N_1 or M_1) from the top is 25 eV. They are not suitable for characterizing attosecond pulses with bandwidth larger than 10 eV centered above 25 eV.

Atomic neon is a good candidate because of the relative flat response over a broad XUV photon energy range. The spin–orbit split is ~ 0.1 eV. The cross section is also large in comparison to helium. The bind energy of the third level (L_1) from the top is 48 eV, thus, XUV photons with energy higher than 48 eV produce electrons with two groups of electrons separated by 26.4 eV. If the bandwidth of the attosecond pulses covers more than 26.4 eV, the photoelectron spectra from them will overlap, which cause difficulties for retrieving the attosecond pulses from the measured spectrogram.

Helium has the simplest level structure. There is no possible confusion caused by the overlapping of spectra from different levels.

7.2.2 Maximum Pressure

The highest gas density is set by the electron mean free path in the gas. When the attosecond pulse is centered at 40 eV, the kinetic energy of the

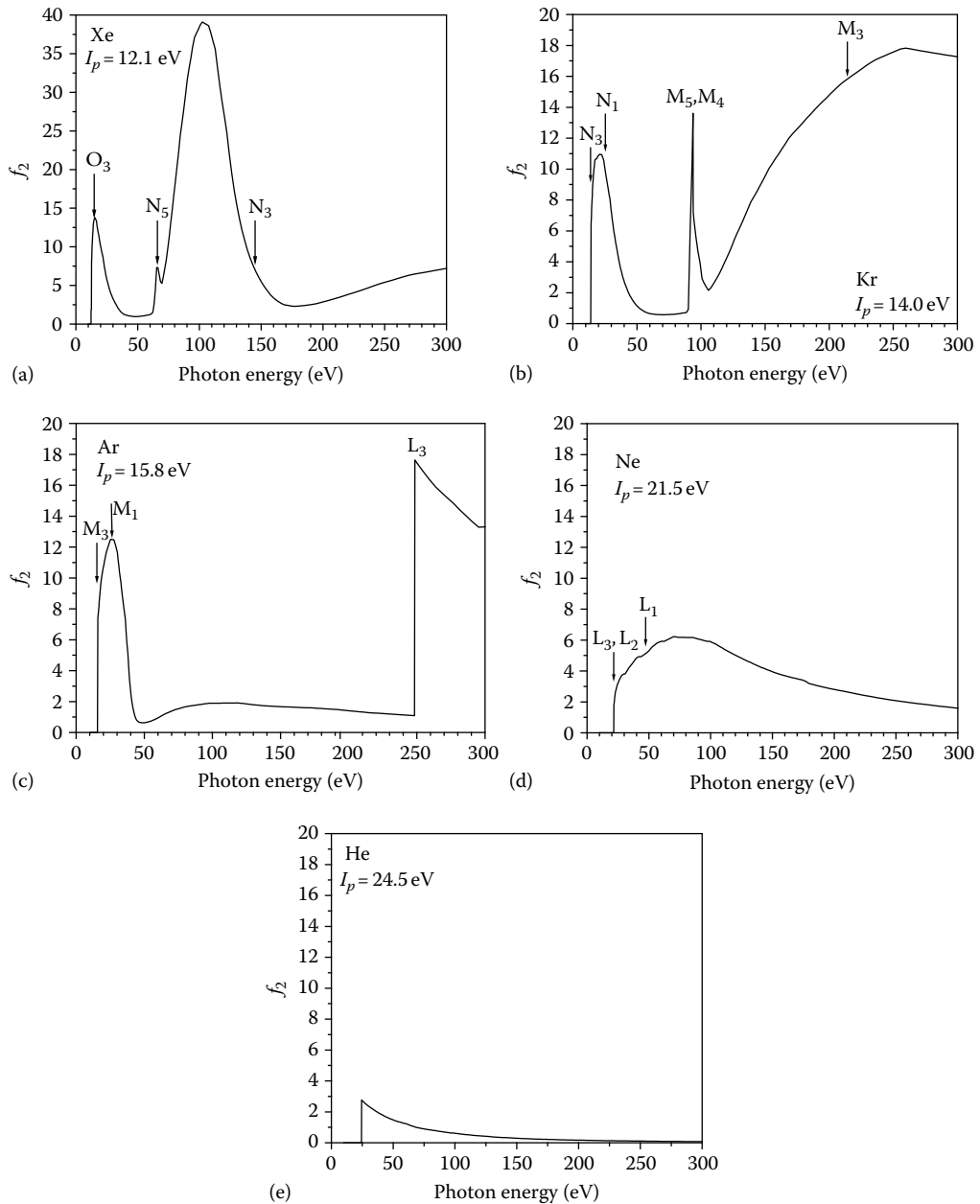


Figure 7.10 Scatter factors of five noble gases. The binding energy of the outer shells are marked.

electrons is about 20 eV. When N_{e0} electrons with same kinetic energy travel through a distance L , the number of unscattered electrons is

$$N_e(L) = N_{e0} e^{-\sigma_e(K)NL}, \quad (7.27)$$

where

$\sigma_e(K)$ is the scattering cross section for electrons with kinetic energy K
 N is the gas density

Based on the Born–Bethe approximation, the total electron scattering cross section is

$$\sigma_e(K) = a_0^2 \left[A_{tot} \left(\frac{1}{K/R} \right) + B_{tot} \left(\frac{1}{K/R} \right) \ln \left(\frac{K}{R} \right) + C_{tot} \left(\frac{1}{K/R} \right)^2 \right], \quad (7.28)$$

where

$a_0 = 5.3 \times 10^{-11}$ m is the Bohr radius

$R = 13.6$ eV is the Rydberg constant

For argon gas, $A_{tot} = 800$, $B_{tot} = 63.9$, $C_{tot} = -1018$. The unit of K is eV. The scattering cross section for argon is shown in Figure 7.11.

With the increase of the detection gas density, the XUV detection efficiency increases, but the probability of electron scattering is also increased. To achieve the maximum number of unscattered electrons generated from XUV photon absorption, the optimum gas pressure is needed.

The yield of detection is defined as

$$\frac{N_e(L)}{N_x} = (1 - e^{-N\sigma(\lambda_x)L}) e^{-\sigma_e(K)NL}, \quad (7.29)$$

where N_x is the number of XUV photons. As an example, for $L = 50$ μm , $\sigma(\lambda_x) = 3 \times 10^{-22}$ m^2 , and $\sigma_e(K) = 6 \times 10^{-20}$ m^2 , the yields as a function of gas density are shown in Figure 7.12.

Assuming a 50 μm diameter detection gas target is used, an argon target, yield peaks at $5 \times 10^{23}/\text{m}^3$ (14 torr) and has a maximum yield of 3.5×10^{-3} . As a comparison, for neon under the same conditions, the yield peaks at $1 \times 10^{24}/\text{m}^3$ (28 torr) with a maximum yield at 1.8%. Electrons are commonly detected by microchannel plate (MCP) detectors, which have to operate under vacuum better than 1×10^{-5} torr. Thus, it is very

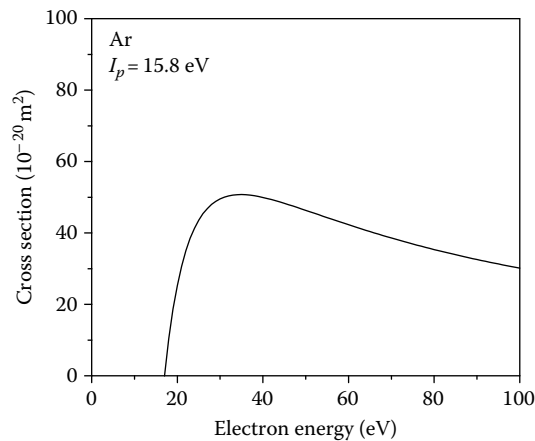


Figure 7.11 Electron scattering cross section of Ar.

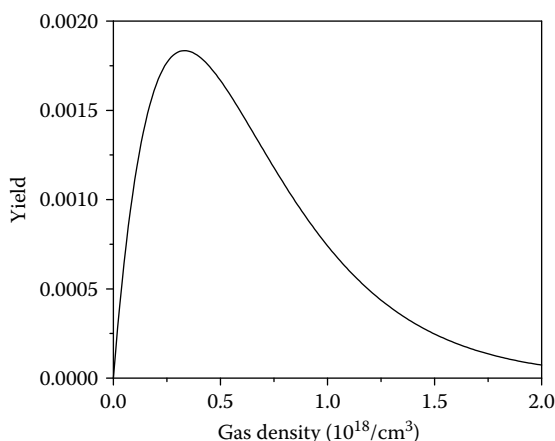


Figure 7.12 Relation of unscattered electron detection yield to gas density.

important to come up with a differential pumping design that can achieve pressure of approximately 10 torr in the detection region while maintaining a high vacuum in the MCP area.

7.3 Electron Time-of-Flight Spectrometer

So far, all demonstrated methods for measuring the duration of attosecond pulses are based on the measurements of the photoelectron spectra produced by the XUV attosecond pulses. A time-of-flight (TOF) spectrometer is the principal electron spectrometer. It draws on the principle that electrons of different momentum pointing to a detector have a different flight time to the detector, which is placed at a certain distance away from their generation point. The implementation of the concept calls for a time-resolved detection of the arriving electrons with descent resolution. The femtosecond laser pulse that generates the attosecond pulse serves at the start signal, whereas the output of the electron detector gives the stop signal to calculate the flight time.

The three key specifications of TOF are

1. *The range of the electron energy that can be measured.* The required range is determined by the attosecond pulse duration, τ_X , to be measured. As is discussed in Chapter 1, a 25 as pulse corresponds to a spectrum with a 75 eV FWHM and extends over a 150 eV range. The photoelectron replica covers the same energy range.
2. *The energy resolution.* The required resolution, ΔK , depends on the scale of the time structure, Δt , that needs to be resolved, i.e., $\Delta K \cdot \Delta t \approx h$. For FROG-Complete Reconstruction of Attosecond Bursts (CRAB) measurements of an attosecond pulse train, which is discussed in later sections, the resolution (the width of each harmonic peak) determines the duration of the overall duration pulse train. For example, if the envelope that contains

all the pulses is 25 fs, the resolution needed is 75 meV. As is mentioned in Chapter 8, for CRAB measurement of single isolated pulses that contain satellite pulses with one cycle (2.5 fs) spacing to the main pulse, a resolution better than 0.5 eV is required to reconstruct these pre- and post-pulses.

3. *The acceptance angle.* The conversion efficiency of generating attosecond pulse is low (on the order of 10^{-6}) and considering the loss of XUV optics and the low photon detection yield discussed in [Section 7.2](#), the number of electrons leaving the detection gas target is small (<1000 per laser shot). Therefore, it is desirable to collect as many electrons as possible by the TOF.

The major advantage of the TOF over other types of electron spectrometer is that the whole spectrum is measured at the same time, which could reduce the data taking time significantly. At least four types of TOF have been used in attosecond pulse measurements, each with their own advantages and weaknesses.

7.3.1 Field-Free TOF

The simplest TOF consists of a drift tube and an electron detector. There is neither an electric nor a magnetic field in the drift tube, as shown in Figure 7.13. The tube is wrapped with a μ -metal (a material with a very large magnetic permeability) sheet to shield the magnetic field of the earth. The detector consists of two to three MCPs stacked together and an anode. An electron is multiplied to $>10^6$ electrons by the MCPs to generate a large sub-nanosecond current pulse in the anode for single event detection. The diameter of the MCP is 10–100 mm. The current pulse picked up by the anode is sent to a low noise amplifier and then to a constant fraction discriminator (CFD).

Using the laser pulse as the start signal and the signal from the CFD as the stop signal, the flight time of the electron in the drift tube is converted to digital numbers in a time-to-digital converter (TDC). The time resolution of the detector and the electronics is on the order of 100 ps to 1 ns. The CFD is needed because the amplitude of the MCP output has large fluctuations for the same input (one electron). The waveform and amplitude of the CFD output does not change much. The peak of its waveform

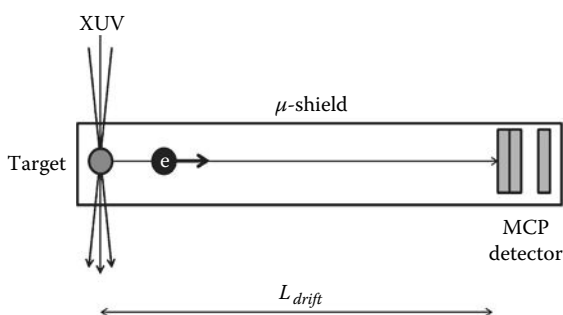


Figure 7.13 Field-free electron TOF spectrometer.

is locked to the peak time of the detector output pulse even when the amplitude of the latter fluctuates.

7.3.1.1 Energy Resolution

For an electron moving along the axis of the TOF with a velocity v , the time it takes to drift through a tube with length L_{drift} is

$$T_{drift} = \frac{L_{drift}}{v} = \frac{L_{drift}}{\sqrt{2K/m_e}}, \quad (7.30)$$

where

m_e is the mass of the electron

K is the kinetic energy of the electron

For two electrons with a slight energy difference, ΔK , their drift time difference is

$$\Delta T_{drift} = -\frac{\sqrt{m_e}}{2\sqrt{2}} L_{drift} \frac{\Delta K}{K^{\frac{3}{2}}}. \quad (7.31)$$

The ratio then becomes

$$\frac{\Delta T_{drift}}{T_{drift}} = \frac{1}{2} \frac{\Delta K}{K}. \quad (7.32)$$

If the minimal time interval that detection electronics can resolve is Δt , then the energy resolution ΔK is given by

$$\Delta K = \frac{\sqrt{8}}{\sqrt{m_e}} \frac{\Delta t}{L_{drift}} K^{\frac{3}{2}}. \quad (7.33)$$

The relative energy resolution

$$\frac{\Delta K}{K} = \frac{\sqrt{8}}{\sqrt{m_e}} \frac{\Delta t}{L_{drift}} \sqrt{K}. \quad (7.34)$$

It is clear that the resolution decreases with the electron energy. For $K = 300$ eV, $\Delta t = 0.25$ ns, to reach $\Delta K/K = 0.2\%$, or $\Delta K = 0.6$ eV, the drift tube length has to be $L_{drift} = 2.6$ m. For a 50 mm diameter detector, the acceptance angle is $\sim 0.57^\circ$, or 7.8×10^{-5} sterad. If the angular distribution of the photoelectron is isotropic, then only $< 0.01\%$ of the electrons are detected. The low detection efficiency is the major drawback of the field-free TOF.

Equation 7.34 reveals that the energy resolution can be improved by three approaches. The first one is increasing the length of the drift tube. This is limited by the space of the laboratory and by the minimum acceptance angle that one can tolerate. The typical length is 0.5–5 m.

7.3.1.2 Retarding Potential

The second method for improving the energy resolution is to slow down the electron before it enters the drift tube by applying a bias voltage between the gas target region and the drift tube, as shown in [Figure 7.14](#). The drawback is that electrons with initial energy less than the bias

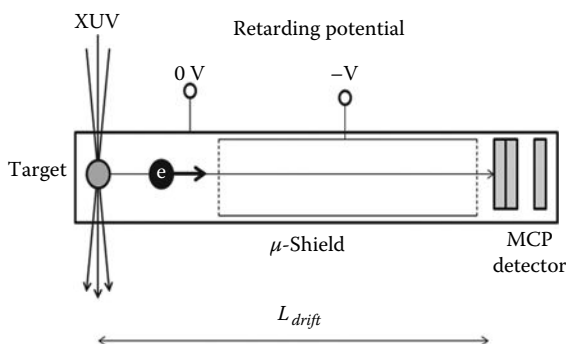


Figure 7.14 Electron TOF spectrometer with retarding potential.

voltage cannot enter the drift tube. The whole spectrum can be obtained by taking many measurements at different bias voltages and then piecing them back together. The acceptance angle is further reduced as the divergence angle of the electron beam is increased when the electrons pass the first mesh.

For example, for studying the $2s2p$ autoionization of helium at 60.1 eV discussed in Chapter 9, the kinetic energy of the photoelectron is 35 eV and the width of the resonance peak is ~ 40 meV. If a 35 eV electron is slowed down to 5 eV, then the energy resolution can be improved by

$$\frac{\Delta K}{\Delta K'} = \left(\frac{35}{5}\right)^{3/2} = 18.5. \quad (7.35)$$

Here, ΔK and $\Delta K'$ are energy resolution for the 35 and 5 eV electrons, respectively. If the resolution is 740 meV without the retarding potential, then slowing the electron from 35 to 5 eV would reach the required 40 meV, comparable to the autoionization line width.

Finally, by choosing MCP detectors with fast response and associated electronics with high time resolution. Specially designed MCP detectors with sub-nanosecond time response have been used in TOF spectrometers.

7.3.1.3 Time-Resolution Measurement

In most attosecond experiments, the start signal is produced by shining femtosecond lasers onto fast photodiode with a response time better than 1 ns. Due to the good stability of the laser pulse, the voltage pulses from the photodiode do not change much from one laser shot to the next, yielding very good time accuracy for the TDC.

The time resolution is limited by the stop signal from the MCP detector. Due to the statistics in the electron multiplication in the MCP, the low-amplitude voltage (a few mV) pulse from the anode fluctuates significantly from shot to shot. Combined this with CFDs, the timing error is on the order of 50–200 ps. Finally, the TDCs bin the electron signal in time intervals of 25 ps or more. Overall, it is difficult to achieve time resolution better than 100 ps.

Experimentally, the time resolution of the MCP detector can be measured by XUV photons arriving on the MCP from gas target by Rayleigh scattering. Or by shining the MCP with the third harmonic of the Ti:Sapphire laser, which is a UV pulse at 267 nm. Since the MCP is sensitive to UV and XUV light, and the duration of the UV/XUV pulse is <1 ps, which can be considered as a delta function for the detector, the time resolution can be measured precisely.

7.3.2 Magnetic Bottle

For characterizing attosecond pulses, it is extremely important to collect all the electrons, while maintaining high-energy resolution over a wide energy range. This is difficult to achieve with field-free designs because the energy resolution deteriorates rapidly as the acceptance angle is increased. For a given MCP detector diameter, the drift tube length has to be reduced to increase the acceptance angle.

These problems can be overcome in the “magnetic bottle” spectrometer. It combines a high collecting efficiency (2π sterad) with good energy resolution. In this instrument, electrons initially emitted in all directions from the region of the detection gas target are formed into a nearly parallel beam in an inhomogeneous magnetic field, which serves as a collimating magnetic lens, as shown in Figure 7.15.

7.3.2.1 Parallelization of the Trajectories

The detector gas target is located between the pole pieces of an electromagnet or a permanent magnet producing a 1 T magnetic field. The Lorentz force causes each emitted electron to spiral around a magnetic field line, as depicted in Figure 7.16.

An electron initially emitted at an angle Θ_i to the z direction (the spectrometer axis), and with an energy K and velocity v , undergoes helical motion in the field B_i . The angular frequency (cyclotron frequency) of the motion is

$$\omega_i = \frac{e}{m_e} B_i, \quad (7.36)$$

where e is the charge of the electron.

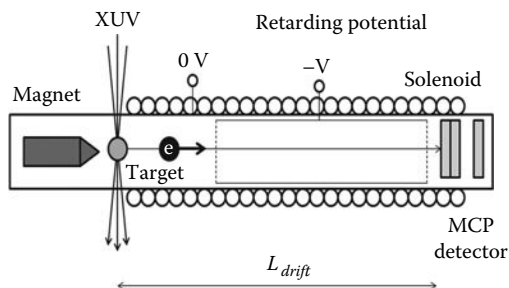


Figure 7.15 Magnetic bottle electron TOF spectrometer.

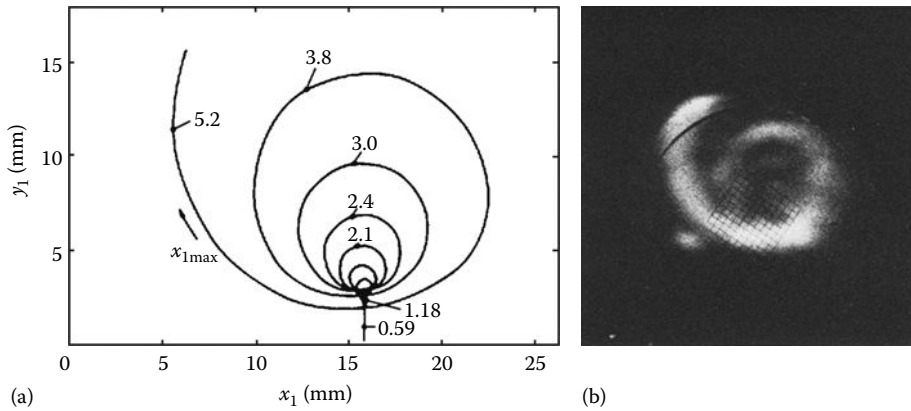


Figure 7.16 Helical motion of an electron moving in a magnetic field that changes gradually from a strong field B_i to a weaker uniform field B_f . The right picture shows the image on the MCP detector with and a fluorescent screen. (Reprinted from P. Kruit and F.H. Read, Magnetic field paralleliser for 2π electron-spectrometer and electron-image, *J. Phys. E*, 16, 313, 1983.)

The radius of the orbit (cyclotron radius) of an electron with initial speed v is

$$r_i = \frac{v \sin \Theta_i}{\omega_i}. \quad (7.37)$$

The moment of inertia of the electron is $m_e r_i^2$. Thus, the angular momentum of the circular motion in the strong field B_i is

$$l_i = (m_e r_i^2) \omega_i = \frac{m_e^2 v^2 \sin^2 \Theta_i}{e B_i} \quad (7.38)$$

Then, the electron enters the region of the weak uniform magnetic field, B_f , of 1 mT (10 gauss). If the variation of the magnetic field with z is adiabatic, by which we mean that if the field experienced by an electron changes negligibly in the course of one revolution of the helical motion, then the angular momentum is a conserved quantity, i.e., $m_e^2 v^2 \sin^2 \Theta_i / e B_i = m_e^2 v^2 \sin^2 \Theta_f / e B_f$.

The angle of the helical motion in the region of low field is given by

$$\frac{\sin^2 \Theta_f}{\sin^2 \Theta_i} = \frac{B_f}{B_i} \approx 10^{-3}. \quad (7.39)$$

The transverse component of the velocity is thus reduced. Since the total velocity is unchanged the longitudinal component increases from v to

$$v_{zf} = v \cos \Theta_f = v \sqrt{1 - \frac{B_f}{B_i} \sin^2 \Theta_i}, \quad (7.40)$$

which is much larger than the transverse velocity component $v_{rf} = v \sin \Theta_f$. This means that the electron trajectories are parallelized.

7.3.2.2 Acceptance Angle

Even electrons that are initially emitted nearly perpendicularly to the field lines in the high-field region are nearly parallel in the low-field region, having the angle

$$\Theta_{f,max} = \sin^{-1} \left(\sqrt{\frac{B_f}{B_i}} \right) \approx 1.8^\circ. \quad (7.41)$$

Thus, the acceptance angle can reach 2π sterad. If the field strengths in the two regions are sufficiently different, after the electron beam has been parallelized, the electrons travel in a uniform magnetic field down to a flight tube before being detected.

7.3.2.3 Energy Resolution

The TOF in the drift tube is

$$T_{drift} = \frac{L_{drift}}{v_{zf}} \approx \frac{L_{drift}}{v} \left[1 + \frac{1}{2} \frac{B_f}{B_i} \sin^2(\Theta_i) \right], \quad (7.42)$$

which implies that it depends on the electron energy, but because of the parallelization of the trajectories it is almost independent of the initial direction of emission.

The maximum difference of the drift time caused by the emission angle is

$$\frac{\Delta T_{drift}}{T_{drift}} = \frac{1}{2} \frac{B_f}{B_i} \approx 10^{-3}. \quad (7.43)$$

The effect is comparable to the difference of the drift time caused by initial energy difference

$$\frac{\Delta T_{drift}}{T_{drift}} = \frac{1}{2} \frac{\Delta K}{K} \quad (7.44)$$

when $\Delta K/K \approx 10^{-3}$.

7.3.2.4 Adiabaticity Parameter

To judge whether the motion of an electron in a magnetic field is adiabatic, we introduce an adiabaticity parameter χ_1 . For an electron orbiting around the z axis at an angle Θ to the axis, the magnitude of the pitch (i.e., the longitudinal distance that the electron moves during one orbit) when the electron is near the position z is

$$\Delta z_{pitch} = \frac{2\pi}{\omega} v_z = 2\pi \frac{m_e}{e} \frac{v \cos \Theta}{B_z}, \quad (7.45)$$

and the change in B_z , over this distance, is

$$\Delta B_z = \frac{dB_z}{dz} \Delta z_{pitch}. \quad (7.46)$$

Therefore, the adiabaticity parameter is defined by

$$\chi_1 = \left. \frac{\Delta B_z}{B_z} \right|_{\Theta \rightarrow 0} = 2\pi \frac{m_e}{e} \frac{v}{B_z^2} \left| \frac{dB_z}{dz} \right|, \quad (7.47)$$

which is the fractional change in the field that the electron experiences in one cyclotron orbit. χ_1 should be less than 1 to assure that the electron motion is adiabatic.

7.3.2.5 Transition Region

In the region between the gas target and the drift tube, the flight time is different for electrons with different emission angle Θ_i . For a given magnetic distribution, the flight time difference can be calculated numerically.

For example, Kruit and Read found that electrons with energies of 1 eV giving the ratio $\Delta T_{tr}/T_{tr}$ for isotropic electron distribution are 0.56%, where T_{tr} and ΔT_{tr} are the average flight time and the flight time spread (FWHM), respectively. This value is larger than that in the drift region as expressed by Equation 7.43. They found that the major contribution to $\Delta T_{tr}/T_{tr}$ comes from the region of the strong magnetic field and can be decreased if the length of strong-field region is decreased.

For the best energy resolution, it is important for the changing-field region to be as short as possible (implying $|dB_z/dz|$ as large as possible) while remaining in the adiabatic limit. These two requirements are contradictory which means a compromise must be found. The magnetic field distribution in the changing-field region can be measured or calculated to determine the adiabaticity parameter. Typically, the length of transition region is $\sim 3\text{--}10$ cm.

Since the flight time T is proportional to $\sqrt{K}$, the energy resolution is related to the time resolution as

$$\frac{\Delta K}{K} = 2 \frac{\Delta T_{tr}}{T_{tr}}. \quad (7.48)$$

Therefore, the minimal theoretical resolution of $\Delta K/K$ expected from the spectrometer would be 0.5%.

For the electrons with kinetic energies of 1 eV the minimal energy resolution, ΔK , is 5 meV. For 200 eV electrons, the corresponding resolution is 1 eV. Thus, the spectrometer offers the possibility of extremely good photoelectron kinetic energy resolution.

7.3.2.6 Transverse Magnification

The magnetic field in the transition region can be considered as a short magnetic lens, like what is used in the electron microscope. The transverse magnification is $M = B_i/B_f = 1000$. The value is used for choosing a detector size. For a gas target diameter of 50 μm , the image diameter on the MCP detector is 50 mm. Figure 7.16 shows the image on the detector for electrons with different energies.

7.3.2.7 Overall Considerations

In order to obtain good energy resolution, four conditions must be satisfied:

1. The magnetic field B_i at the source must be much stronger than the field B_f in the drift tube, to give a high degree of parallelization of the trajectories.

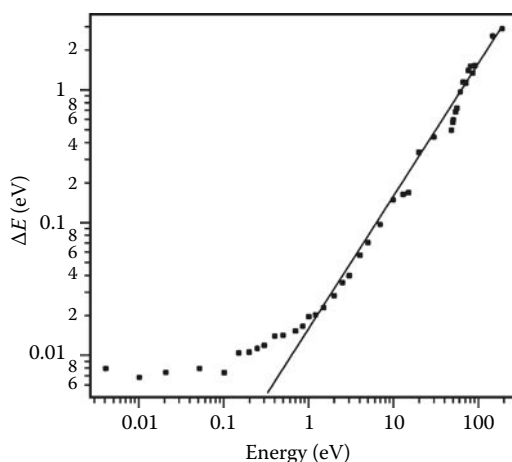


Figure 7.17 Resolution of the magnetic bottle TOF spectrometer as a function of electron energy. The line $\Delta K/K = 1.6\%$ is a fit through the data points. (Reprinted from P. Lablanquie, L. Andric, J. Palaudoux, U. Becker, M. Braune, J. Viehhaus, J.H.D. Eland, and F. Penent, Multielectron spectroscopy: Auger decays of the argon 2p hole, *J. Electron Spectros. Relat. Phenom.* 156–158, 51–57, Copyright 2007, with permission from Elsevier.)

2. The reduction in field from B_i to B_f must take place in a short distance compared to the length of the drift tube to ensure a small spread in the flight times.
3. The adiabaticity parameter χ_1 must be kept sufficiently small, to avoid the harmful effects of strongly nonadiabatic behavior.
4. The field in the drift tube must be at least a few mT to mask the effects of any stray electric or magnetic fields. The Earth's magnetic field should be shielded with μ -metal.

The resolution of the magnetic bottle is best at low photoelectron energies (Figure 7.17). At intermediate photoelectron energies, the resolution is limited by the magnetic bottle. At very high photoelectron energies, the data collection process determines the limit of resolution, since the TOF signal is recorded in finite time bins.

7.3.2.8 Construction of the Magnetic Bottle

The spectrometer for attosecond pulse detection consists of a gas target, a strong magnetic field around the target, a drift tube located in a uniform weak magnetic field and an MCP detector. The strong magnetic field (about 1 T) in the gas target region can be created from either an electromagnet or a permanent magnet. The weak field is generated by coils wrapped around the drift tube. The coils are then wrapped with a μ -metal shielding to minimize the earth magnetic field in the tube. Electrodes can be added to the entrance of the drift tube to provide retarding electric field. All of the surfaces in the detection region and TOF tube should be coated with conductive carbon powder to eliminate any charge buildup effects. All parts should be constructed with aluminum or nonmagnetic stainless steel.

7.3.2.9 Experimental Energy Resolution

For a very narrow bandwidth electron pulse, the observed temporal widths of the pulse can be much wider than the calculated value. The main contributions to the broadening are the following:

1. The time width of a pulse coming from the MCP detector corresponding to a single electron can be a few nanoseconds long.
2. The time resolution of the electronic measuring system is a limited value, typically <100 ps.

Kruit found the minimal kinetic energy resolution, ΔK , to be approximately 15 meV for a 0.62 eV electron energy. In fact, the energy resolution is dependent on the energy of photoelectrons, as shown in [Figure 7.17](#).

7.3.2.10 Retarding Potential

Like the field-free TOF, a way of improving the energy resolution is to retard the electrons before they enter the flight tube, just after they have passed through the diverging part of the magnetic field. To achieve the best resolution for particular photoelectron energy, the flight tube itself can be electrically isolated from the rest of the interaction chamber and can be held at a voltage that is adjustable. In this way, the flight-tube voltage can be adjusted to bring the desired part of the spectrum into the region of best resolution.

7.3.3 Position-Sensitive Detector

When the detector of the TOF spectrometer has spatial resolution, one can measure the angular distribution for each electron energy value. In other words, the momentum distribution of the photoelectrons can be determined. Such a TOF spectrometer has been used in the cold target recoil ion momentum spectroscopy (COLTRIMS) setup where the momenta of both electrons and ions are measured at the same time for coincidence experiments.

A half CONTRIMS that detect electrons only has been built in the author's laboratory for an attosecond streak camera, which is schematically shown in [Figure 7.18](#).^{*} The position-sensitive detector of the TOF spectrometer consisting of two stacked MCPs in a chevron configuration and a delay-line anode with a two-layer of wires for independent detection of electrons along two transverse directions (x and y). The diameter of the MCP is 120 mm. The drift tube of the TOF is 290 mm long and a uniform magnetic field (~ 1.6 gauss) is applied along the TOF axis between the gas jet and MCPs (the z direction).

There are several advantages by using a large-area position-sensitive detector, 120 mm, and the uniform magnetic field along the TOF axis as compared to some other types of spectrometers.

^{*} The setup is described in more detail in Feng, X., S. Gilbertson, S.D. Khan, M. Chini, Y. Wu, K. Carnes, and Z. Chang, Calibration of electron spectrometer resolution in attosecond streak camera, *Opt. Express* 18, 1316 (2010).

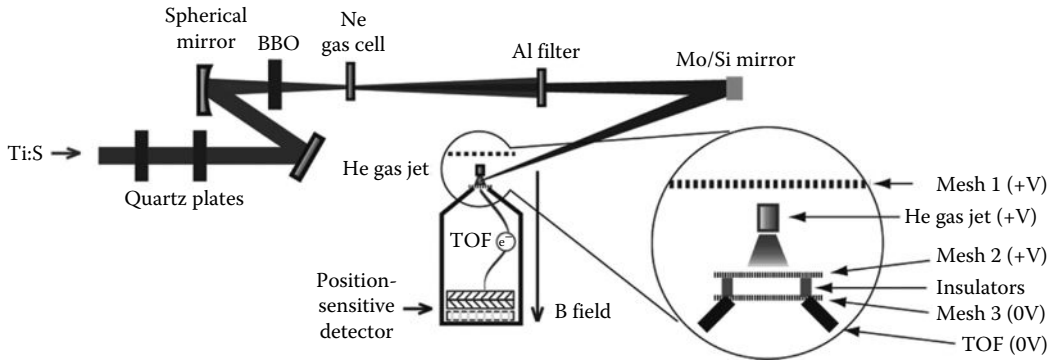


Figure 7.18 TOF with a position sensitive detector in a setup for calibrating its energy resolution. (Reprinted from X. Feng, S. Gilbertson, S.D. Khan, M. Chini, Y. Wu, K. Carnes, and Z. Chang, Calibration of electron spectrometer resolution in attosecond streak camera, *Opt. Express*, 18, 1316, 2010. With permission of Optical Society of America.)

First, the acceptance angle is large because of the large detector size, which is important for increase the count rate. The acceptance angle is $\pm 11.7^\circ$ even without the magnetic field. The angle is further increased by the magnetic field. In the magnetic field, the acceptance angle changes with electron energy.

Second, compared to a single large anode, a delay-line detector can improve the energy resolution when the acceptance angle is large by taking advantage of the additional position information. This is because the flight time changes with the electron emission angle. In a field-free TOF, the flight time is determined by the velocity component in the z direction, $v_z = \sqrt{2K/m_e} \cos \Theta$. For the given setup, $\cos \Theta = \cos(11.7^\circ) = 0.98$. This introduces a 2% flight time error, which limits the energy resolution to $\Delta K/K = 4\%$, according to Equation 7.48. For $K = 50$ eV, the resolution is 2 eV.

Finally, as compared to velocity map imaging (VMI) discussed in the next section, which also has angular resolutions, it is easier to implement differential pump, i.e., to add a cone with a small opening (~ 1 mm diameter) between the gas jet and the MCPs so that high gas density can be delivered to the detection region without affecting the MCP operation.

The disadvantage of applying the magnetic field B along the TOF axis is the blind spots at certain energy values caused. When an electron flies to the detector with energy, K , the radial position versus the arrival time, t , can be written as

$$r = \frac{2\sqrt{2m_e K - (m_e L_{drift}/t)^2}}{eB} \left| \sin\left(\frac{1}{2}\omega t\right) \right|, \quad (7.49)$$

where the cyclotron frequency $\omega = (e/m_e)B$. Figure 7.19 presents some trajectories of electrons with different energies. The unit of the energy labeling the trajectories is electron volt. The horizontal dotted line shows the radial boundary of the detector ($r = 60$ mm), beyond which the electrons cannot be detected. All the electrons, independent of their initial kinetic energies, hit the center of the detector ($r = 0$) at 320 and 650 ns,

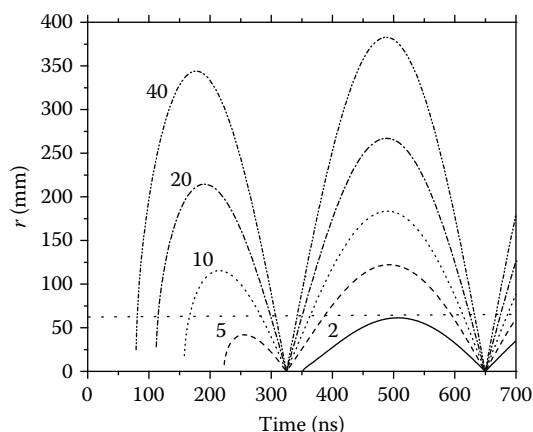


Figure 7.19 Calculated radial position on the detector as a function of the TOF for electrons of different energies when they arrive at the detector. The number beside each curve shows the kinetic energy (in eV) of the electron corresponding to that curve. The horizontal dotted line represents the radius of the detector (60 mm). (Reprinted from X. Feng, S. Gilbertson, S.D. Khan, M. Chini, Y. Wu, K. Carnes, and Z. Chang, Calibration of electron spectrometer resolution in attosecond streak camera, *Opt. Express*, 18, 1316, 2010. With permission of Optical Society of America.)

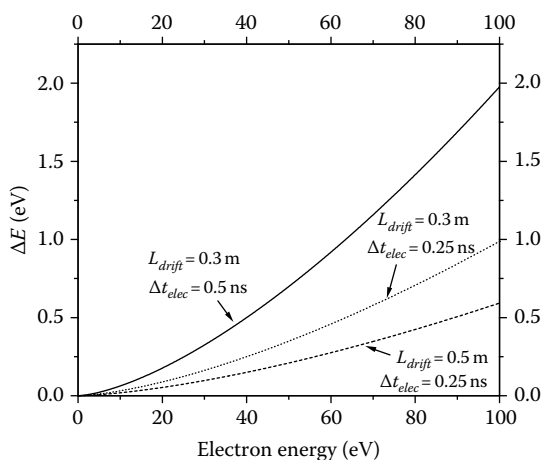


Figure 7.20 Calculated energy resolution of an electric field-free TOF with different drift tube length and time resolution.

where the momentum information cannot be determined. When the setup shown in Figure 7.18 is used to measure attosecond pulses, the flight-time range from 0 to 320 ns is used to avoid the nodes.

The energy resolution can be calculated in the same way as the field free TOF. The calculated results for three combinations of drift tube length and detection time resolution are shown in Figure 7.20. A 0.5 eV resolution in the 100 eV range is achievable for a 0.5 m drift tube and detector with 0.25 ns resolution.

7.3.3.1 Experimental Determination of the Energy Resolution

When electrons with large emission angles are collected with magnetic or electrostatic lens, the energy resolution of the TOF spectrometer is affected by many factors, such as the lens field distribution, the settings of the CFD etc. For such TOF, it is hard to estimate the resolution accurately by calculations. It is therefore necessary to determine the energy resolution experimentally. It would be even better if the test can be done in the attosecond second pump-probe or streak camera setup so that it can be checked when needed.

The main difficulty of in situ calibration of the TOF electron spectrometers in the attosecond streak cameras is due to the fact that it is hard to find XUV light source with a known linewidth comparable to the resolution of the spectrometer. High-order harmonics are used sometimes, but the width of the harmonic peaks can be comparable or even broader than the spectral resolution.

7.3.3.2 Setup

It is demonstrated that the Fano resonance peaks due to the autoionization of atoms are good calibration sources. The origin of the $2s2p$ Fano peak of helium at 60.1 eV is discussed in detail in Chapter 9, which is a pronounced photoabsorption absorption peak with a linewidth of 38 meV. This calibration method requires the generation of a pulse XUV continuum around the Fano peak, which is 60.1 eV for the $2s2p$ of helium. Such a source can naturally exist in an attosecond streak camera because the spectrum of an isolated attosecond pulse is a XUV continuum. Consequently, this approach allows in situ calibration of the TOF resolution.

Figure 7.18 shows a subset of the attosecond streak camera in the author's lab for testing the TOF energy resolution. First, single isolated attosecond XUV pulses corresponding to a smooth continuous XUV spectrum around 60 eV are generated in the gas cell filled with neon. The attosecond XUV continuum is generated by the double optical gating (DOG), which is a combination of two-color gating and polarization gating, as introduced in Chapter 6. With the DOG, XUV supercontinua covering the 30–600 eV have been generated.

The XUV beam is focused onto a helium gas jet by a broadband-multilayer Mo/Si mirror which has 6% reflectance around 60 eV. When the XUV light is absorbed by the helium gas in front of the TOF, a photoelectron pulse is produced. Due to the Fano resonance at 60.1 eV photon energy, the spectrum of the electron pulse consists of a sharp peak with 38 meV at 35 eV on a continuum. The freed electrons were detected by the TOF electron spectrometer with a position-sensitive detector. The resolution of the electron spectrometer at 35 eV can be considered to be equal to the detected width of the measured Fano peak.

A start signal for the TOF measurement is taken from a fast photodiode triggered by the femtosecond laser for generating the XUV pulse. The stop signal is from the MCP output. The signals from the photodiode, the MCP and the delay-line anode are sent to CFDs. The timings are measured with a digital converter (TDC) that has a 25 ps resolution.

7.3.3.3 Energy Resolution Calibration

To characterize the energy resolution over a range of electron energy, a retarding potential is applied to vary the kinetic energy of electron from the $2s2p$ resonance, which is depicted in Figure 7.18. Three fine meshes are used to slow down the electrons. Meshes 1 and 2 are applied with the same positive potentials (+V) as the helium gas jet to provide field free region around the jet. Mesh 3 is conductively connected to the cone of the detector chamber and is grounded so that there is no electric field in the drift tube. The electron decelerates only in a uniform axial field between meshes 2 and 3. Such a configuration assumes that all electrons are slowed down by the same amount regardless of their emission angle and initial energy.

The photoelectron spectra from helium measured with different retarding potentials are presented in Figure 7.21. They are shifted vertically for a clear presentation. The $2s2p$ autoionization peak, sitting on a continuous photoelectron background, shifts from 35.5 eV when the retarding potential is 0, to 10.9 eV when the retarding potential is -30 V. The resulted effective retarding potential is around 80% of the actual one. The big hump near the high-energy cutoff of the spectra results from the high reflectance of the multilayer Mo/Si mirror around 65 eV photon energy. The TOF spectrometer energy resolution for electrons with different kinetic energies obtained from the widths of the $2s2p$ peak is plotted in Figure 7.22. The resolution changes from 0.3 to 0.6 eV when the electron energy increases from 10 to 35 eV.

The significant improvement of energy resolution of the TOF spectrometer by adding the retarding potential is the evidence that the time resolution of the MCP detector is a major limiting factor. The energy resolution as a function of the electron kinetic energy calculated with Equation 7.34 is

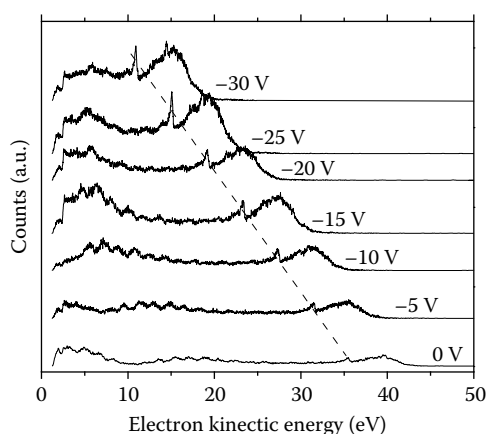


Figure 7.21 Photoelectron energy spectra produced by absorbing isolated XUV attosecond pulses. The number beside each of the spectra shows the retarding potential added in collecting the corresponding spectrum. The dashed line connects all the autoionization peaks for guiding the eye. (Reprinted from X. Feng, S. Gilbertson, S.D. Khan, M. Chini, Y. Wu, K. Carnes, and Z. Chang, Calibration of electron spectrometer resolution in attosecond streak camera, *Opt. Express*, 18, 1316, 2010. With permission of Optical Society of America.)

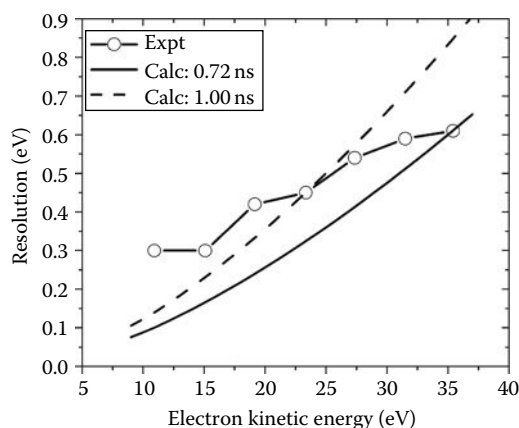


Figure 7.22 Comparison of the spectrometer resolution as a function of the electron kinetic energy between the experimentally measured and the calculated values. (Reprinted from X. Feng, S. Gilbertson, S.D. Khan, M. Chini, Y. Wu, K. Carnes, and Z. Chang, Calibration of electron spectrometer resolution in attosecond streak camera, *Opt. Express*, 18, 1316, 2010. With permission of Optical Society of America.)

plotted in Figure 7.22 for two different time resolutions: 0.72 and 1 ns. The deviation of the measured curve from the $K^{3/2}$ indicates that other factors such as the earth's magnetic field also contribute to the energy resolution. This is not a surprise because the applied magnetic field (~ 1.6 gauss) is comparable to the magnitude of the earth field (~ 0.5 gauss).

7.3.4 Velocity Map Imaging

An example of VMI in an attosecond streak camera is illustrated in Figure 7.23. Neutral gas atoms are ionized by the XUV attosecond pulses and projected by electron optics onto an imaging MCP detector. A CCD camera captures the two-dimensional (2D) image and established techniques to allow for the reconstruction of the full three-dimensional velocity distribution of the electrons after they left the target region. The use

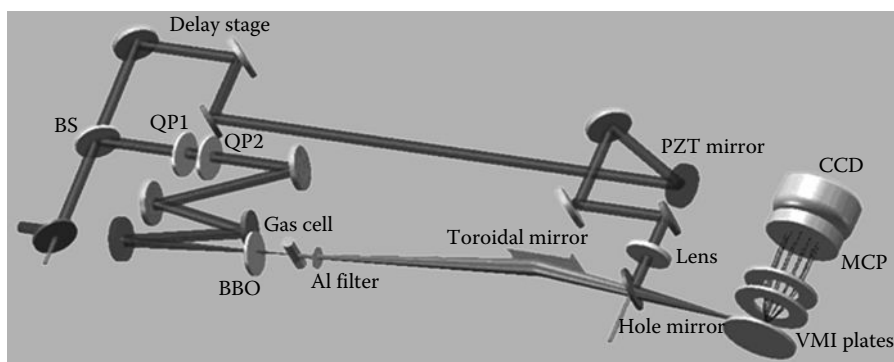


Figure 7.23 Attosecond pump-probe setup with a VMI spectrometer.

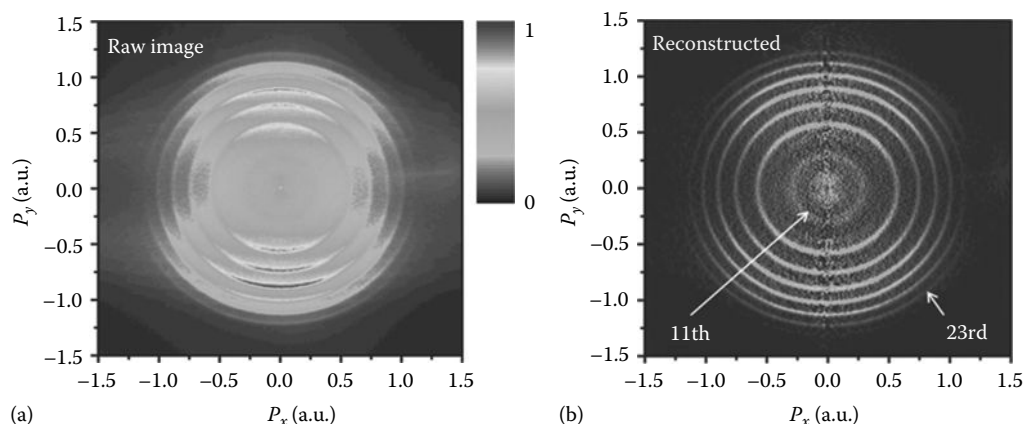


Figure 7.24 Angular distribution of photoelectrons emitted from argon gas measured with a homemade VMI spectrometer. (a) Raw image. (b) Reconstructed momentum distribution. The 11th and the 23rd order harmonics are labeled.

of electron lenses eliminates the spatial blurring of the image, allowing velocity resolution on the order of 1% or better.

The combination of electron optics lens and 2D detection makes VMI possible because particles sharing the same initial velocity are mapped onto the same spot of a position-sensitive detector. Abel inversion, onion peeling, or 2D inversion algorithms, permits the velocity map of the photoelectrons to be reconstructed from a single, raw, experimental image. An example of the image captured by the CCD is shown in Figure 7.24. In this case, high-order harmonics are used to generate photoelectrons from argon gas.

The acceptance angle of VMI is 4π sterad. However, it is difficult to implement differential pumping since the gas target and the MCP are located in the same chamber. High local density gas can be released from a small opening in the repeller plate to improve XUV detection efficiency.

7.4 Measurement of Temporal Width of a Single Harmonic Pulse

When laser pulses containing many optical cycle are used to generate high-order harmonics, each harmonic peak is the result of the interference of many, m , attosecond pulses. The spacing between the two neighboring attosecond pulses is half of the optical cycle, T_0 . When one of the harmonic peaks is filtered out, its temporal duration is on the order of $m \times (T_0/2)$, that is comparable to the pump laser pulses duration, i.e., a few to tens of femtoseconds. Such XUV pulses can be measured by using a cross-correlation method base on the laser assisted photoelectric effect (LAPE). The mechanism of the LADE is shown in the left graph of Figure 7.25.

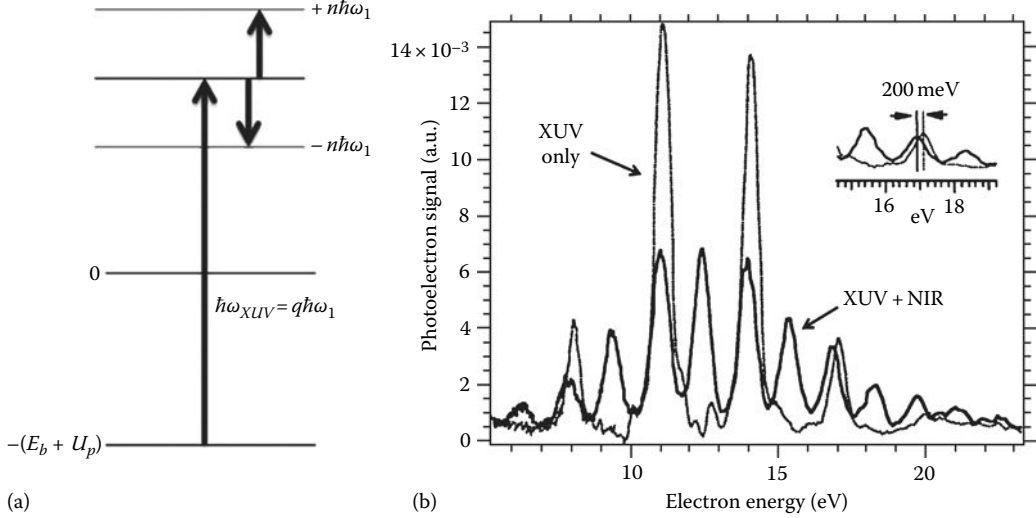


Figure 7.25 (a) The principle of LAPE. (b) Measured photoelectron spectrum. The thin line is the spectrum with harmonic field only. The thick line is the spectrum with both the XUV and the NIR laser field. (Reprinted with permission from T.E. Glover, R.W. Schoenlein, A.H. Chin, and C.V. Shank, *Phys. Rev. Lett.* 76, 2468, 1996. Copyright 1996 by the American Physical Society.)

An XUV beam contains one harmonic lifts the electron of the detection atom from the ground state into the continuum. An intense NIR laser beam that spatially and temporally overlaps with the XUV induces free-free transitions involving the absorption or emission of n photons after the initial ionization. In the electron energy spectrum such free-free transitions result in sidebands on both sides of each ionization peak, separated by the laser photon energy. When a laser beam with carrier frequency ω_1 and the q th harmonic beam with carrier frequency ω_X interacts with an atom simultaneously, the kinetic energy of the electrons exposed to both frequencies is given by

$$K_{q,n} = (\hbar\omega_X + n\hbar\omega_1) - (I_p + U_p), \quad (7.50)$$

where

I_p is the binding energy of the target atom

U_p is the ponderomotive energy of the electron in the NIR field

For a given peak laser intensity, I_0 , the corresponding maximum ponderomotive potential is

$$U_{p0} = 9.3 \times 10^{-14} I_0 \lambda_0^2. \quad (7.51)$$

The unit of U_{p0} is eV, the unit of I_0 is W/cm^2 , and the unit of the wavelength of the laser is in micrometers. For a Ti:Sapphire laser, $\lambda_0 = 0.8 \mu\text{m}$. As an example, if the intensity of a Ti:Sapphire laser is $7.4 \times 10^{11} \text{ W}/\text{cm}^2$, the ponderomotive potential is 44 meV.

When more than one harmonic peaks are filtered out, the harmonic peaks are separated by two photon energies. In addition, photoelectron energy peaks show up between the two adjacent harmonic peaks due to the $n = 1$ and $n = -1$ process, as shown in the right graph of Figure 7.25.

The height of this sideband peak depends on both the intensity of the harmonic pulse and the laser pulse that assist the photoelectric effect. This effect can be used to build a cross correlator to measure the XUV pulse duration if the width of the laser pulse is known.

Supposing the sideband intensity is linearly proportional to the assisting laser intensity, when the delay between the XUV and the laser, τ_d , is changed, the sideband peak can be expressed as

$$S(\tau_d) \propto \int_{-\infty}^{+\infty} I_X(t - \tau) I_{IR}(t) dt. \quad (7.52)$$

For a known laser-pulse profile $I_{IR}(t)$, the width of the $I_X(t)$ can be obtained by deconvolution of $S(\tau_d)$ for some assumed pulse shape.

7.4.1 Sidebands

The required laser intensity for achieving a sideband with comparable height of the harmonic peaks can be estimated by the Keldysh–Faisal–Reiss (KFR) theory, which neglects the influence of the Coulomb field of the detection atom on the free-electron wave function. The electron wave function in the continuum is described by Volkov states introduced in Chapter 4.

Based on the KFR theory, the intensity dependent probability that the photoelectric effect is accomplished by absorption ($n > 0$) or emission ($n < 0$) of n laser photons is

$$p_n(U_p) = \frac{3}{2} \left[\frac{p(U_p)}{p_0} \right]^3 \left[\frac{1 + \left(\frac{a_0}{\hbar} \right)^2 p_0^2}{1 + \left(\frac{a_0}{\hbar} \right)^2 p^2(U_p)} \right]^4 \int_0^\pi \sin(\Theta) \cos^2(\Theta) \times J_n^2[\alpha(U_p), \beta(U_p)] d\Theta. \quad (7.53)$$

Here, a_0 is the Bohr radius divided by the nuclear charge, $p_0 = \sqrt{2m_e(\hbar\omega_X - I_p)}$ is the momentum the photoelectron in the absence of the laser field, and $p(U_p) = \sqrt{2m_e[(\hbar\omega_X + n\hbar\omega_1) - (I_p + U_p)]}$ is the momentum the photoelectron in the presence of the laser field.

Helium is chosen as a target gas because its ground state has no fine structure that would complicate the interpretation of the energy spectra. We consider the photoionization of the helium atom by the 23rd harmonic of Ti:Sapphire laser as an example. In this case, $I_p = 24.587$ eV and $\hbar\omega_X = 35.65$ eV. For $n = 1$ and $I_0 = 7.4 \times 10^{11}$ W/cm², we have $p_0 = 1.018$ a.u. and $p(U_p) = 1.071$ a.u. Thus,

$$\left[\frac{p(U_p)}{p_0} \right]^3 = 1.211 \quad (7.54)$$

and

$$\left[\frac{1 + \left(\frac{a_0}{\hbar} \right)^2 p_0^2}{1 + \left(\frac{a_0}{\hbar} \right)^2 p^2(U_p)} \right]^4 = 0.913. \quad (7.55)$$

In the integral, $J_n(\alpha, \beta) = 1/2\pi \int_{-\pi}^{+\pi} e^{i(\alpha \sin \theta + \beta \sin 2\theta - n\theta)} d\theta$ is the n th-order generalized Bessel function. In Gaussian units, $\alpha = [eA_0 p(U_p)/m_e \hbar \omega_0] \cos(\Theta)$ and $\beta = -U_p/2\hbar\omega_0$. The vector potential is given by $A_0 = E_0 c/\omega_1$, where E_0 the electric field amplitude of the laser. For $I_0 \leq 1 \times 10^{12}$, $\beta \leq 0.02$, and $\alpha \lesssim 1.7$, we have $J_n(\alpha, \beta) \approx J_n(\alpha) + (1/2)\beta[J_{n-2}(\alpha) - J_{n+2}(\alpha)]$, where $J_n(\alpha)$ is the n th-order Bessel function of the first kind. At the given intensity

$$\int_0^\pi \sin(\Theta) \cos^2(\Theta) J_n^2[\alpha(U_p), \beta(U_p)] d\Theta = 0.135. \quad (7.56)$$

Of the three terms expressed in 7.54, 7.55, and 7.56, this one depends strongly on the laser intensity. For the parameters chosen in our example, we can fit the intensity dependence for $n = 1$ by a simple function

$$p_{1,q=23}(U_p) = 7.2 U_p e^{-7.75 U_p}. \quad (7.57)$$

In the XUV region, neon gas is another option of target because of the larger absorption cross section. As another example, we consider $q = 23$ and $n = 1$ for this gas. In this case, the probability can be approximated by

$$p_{1,q=23}(U_p) = 9.3 U_p e^{-9.65 U_p}, \quad (7.58)$$

or

$$p_{1,q=23}(I_0) = 5.54 \times 10^{-13} I_0 e^{-5.74 \times 10^{-13} I_0}. \quad (7.59)$$

Typical intensities needed to saturate the $n = \pm 1$ continuum–continuum transitions are of the order of 10^{12} W/cm² and are easily achieved with pulse energies of a few microjoules. The probability depends almost linearly on the laser intensity, as shown in Figure 7.26. For intensities much less than the saturation intensity, $p_{1,q=23}(I_{IR}) \propto I_{IR}$.

Using the probability derived from the KFR theory, the sideband peak signal as a function of the delay between the XUV and NIR pulse can be expressed as

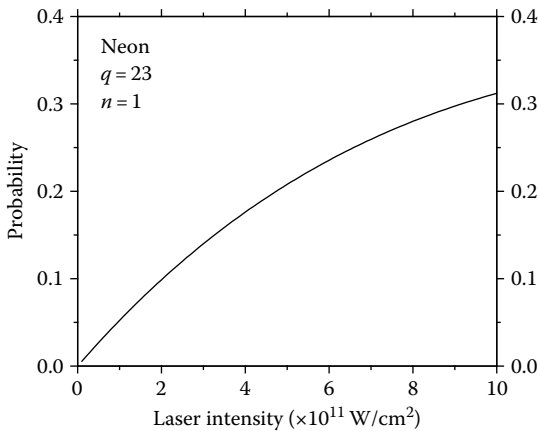


Figure 7.26 Transition probability to the first sideband.

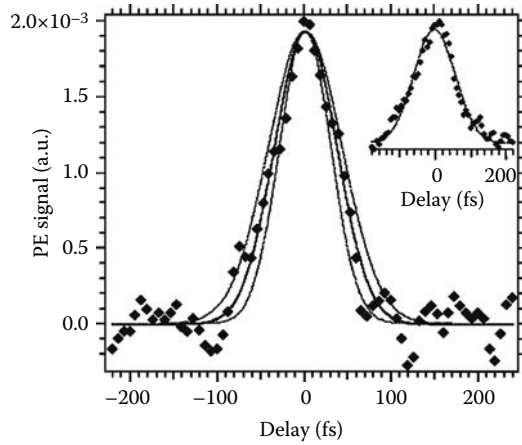


Figure 7.27 Cross-correlation trace of an XUV pulse and an NIR laser pulse. (Reprinted with permission from T.E. Glover, R.W. Schoenlein, A.H. Chin, and C.V. Shank, *Phys. Rev. Lett.* 76, 2468, 1996. Copyright 1996 by the American Physical Society.)

$$\begin{aligned}
 S_{q=24}(\tau_d) &\propto \int_{-\infty}^{+\infty} I_X(t - \tau_d) \{p_{23,1}[I_{IR}(t)] + p_{25,-1}[I_{IR}(t)]\} dt \\
 &\propto \int_{-\infty}^{+\infty} I_{IR}(t - \tau) I_X(t) dt.
 \end{aligned} \tag{7.60}$$

Here, we ignore the effects of the phase difference between the two adjacent harmonics on the height of the sideband peak. In other words, interference effects are neglected. A measured cross-correlation trace is shown in Figure 7.27. In this case, the XUV pulse duration is 50 fs, obtained from the deconvolution, which is comparable to that of the laser pulse, 70 fs, that generated the high harmonics.

7.5 Reconstruction of Attosecond Beating by Interference of Two-Photon Transition

7.5.1 Reconstruction of Attosecond Beating by Interference of Two-Photon Transition Experiments

Reconstruction of attosecond beating by interference of two-photon transition (RABITT) was developed for measuring the average duration of the attosecond pulses in a train. In the spectrum domain, these pulses correspond to a finite number of high-harmonic peaks. The power spectrum of high-order harmonics can be easily measured with either an XUV grating spectrometer, or an electron TOF spectrometer. In the later case, the XUV photons are converted to photoelectrons through photoelectric effect, as

described in Section 7.4. Here, we assume that each harmonic is a monochromatic wave, which means that their temporal width is infinite, contrast to what is stated in Section 7.4. This is, of course, an approximation. If the relative phases between all the harmonics are known, then the temporal structure of the corresponding attosecond pulse train can be determined.

The temporal profile $I_X(t)$ of the intensity of the attosecond pulse train can be expressed as

$$I_X(t) = \left| \sum_N A_q e^{i[\omega_q t + \varphi_q]} \right|^2, \quad (7.61)$$

where A_q and phases φ_q are the spectral amplitude and phase of the q th harmonic, respectively. Here, we assume that there are N harmonics contributing to the pulse train. The spectral phase φ_q , which is sampled only at the harmonic frequencies, reflects their synchronization during the emission process. RABITT is a technique that measures the spectral phase of each harmonic.

The setup is similar to that used in the LAPE experiments. Unlike the LAPE experiments, here the relative phase between two adjacent harmonics is not neglected. In fact, it is the main quantity to be determined. An example of the RABITT setup is illustrated in Figure 7.28. The time delay between the XUV pulse and the NIR laser field has to be stabilized and controlled to a fraction of laser cycle to observe the interference due to the phase difference, which is not required by the LAPE measurements.

The NIR laser beam from a CPA is split into two by a mirror with a hole. The XUV attosecond pulse train is generated in the first gas jet by the laser beam reflected from the area outside the hole. Interestingly, the on-axis intensity of the XUV beam is the highest, unlike the driving laser beam, which is a hollow due to the hole. The NIR beam passing through the hole serves as the dressing laser. Both the XUV and the NIR dressing

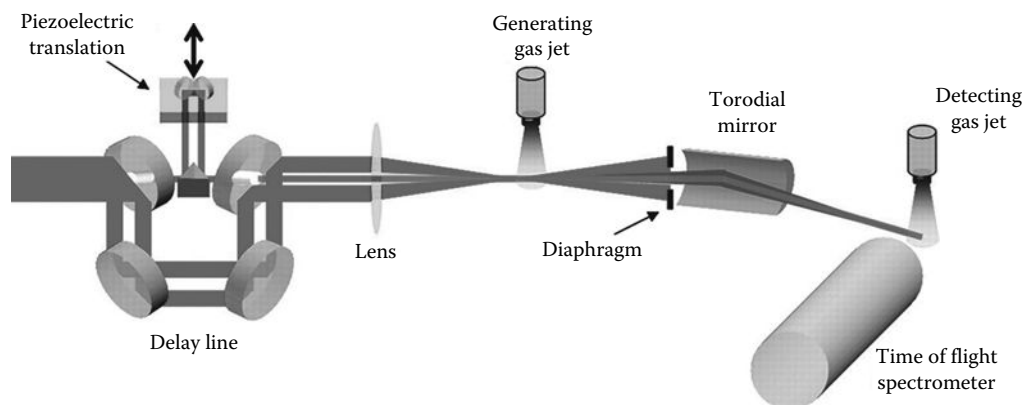


Figure 7.28 RABITT setup. (Reprinted from Y. Mairesse, A. de Bohan, L. J. Frasinski, H. Merdji, L.C. Dinu, P. Monchicourt, P. Breger, M. Kovacev, R. Tai'eb, B. Carré, H.G. Muller, P. Agostini, and P. Salieres, Attosecond synchronization of high-harmonic soft x-rays, *Science*, 302, 1540, 2003. With permission of AAAS.)

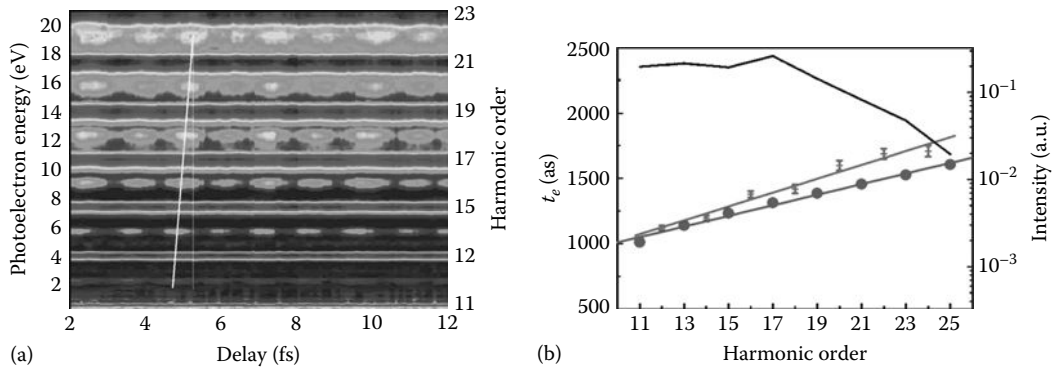


Figure 7.29 RABITT trace. (Reprinted from Y. Mairesse, A. de Bohan, L. J. Frasinski, H. Merdji, L.C. Dinu, P. Monchicourt, P. Breger, M. Kovacev, R. Tai'eb, B. Carré, H.G. Muller, P. Agostini, and P. Salieres, Attosecond synchronization of high-harmonic soft x-rays, *Science*, 302, 1540, 2003. With permission of AAAS.)

beams are focused by a toroidal mirror onto the second gas jet, where the photoelectrons are generated in the two-color (XUV plus NIR) field. The electron spectrum is measured by a TOF.

The measured photoelectron spectra as function of the delay between the XUV pulse and the NIR field are shown in Figure 7.29. The spectral phase of each harmonic can be reconstructed from the position of the sideband peaks as a function of the delay. Combined with the high harmonic intensity information, we can figure out the pulse duration, assuming that the width of all the pulses in the train is the same and the attosecond pulses is an infinitely long train.

The first spectral phase measurements of five consecutive harmonics generated using Ti:Sapphire laser in argon revealed that the phase is almost linearly proportional to the photon energy, corresponding to a train of 250 as pulses.

7.5.1.1 Spectral Phase and Harmonic Emission Time

It is known in Fourier optics that a delay in the time domain corresponds to a phase shift in the frequency domain. As an example, when a delta function is displaced by t_0 , the Fourier transform of it is

$$\mathcal{F}[\delta(t - t_0)] = e^{i\omega t_0}. \quad (7.62)$$

The phase of two harmonics is different because they are emitted at different time, t_{eq} . By defining the phase of a harmonic q_r emitted at $t_e = 0$ as the common reference, then the phase of q th harmonic is

$$\varphi_q = q\omega_1 t_{eq}. \quad (7.63)$$

If all the harmonics are emitted at the same time, then is a constant, then t_e the width of pulse in the train is transform-limited. In that case, the phase deference between two adjacent harmonics is

$$\Delta\varphi = (q + 2)\omega_1 t_e - q\omega_1 t_e = 2\omega_1 t_e. \quad (7.64)$$

Because of the uncertainty principle, one cannot define an instant associated with a single energy: The emission time of harmonic q applies to a group of harmonics centered on $q\omega_1$.

To understand the mechanism of RABITT, we discuss the physics of photoemission in one-color (XUV only) and two-color field (XUV and NIR fields).

7.5.2 Transition-Matrix Element in XUV Field

Consider the photoionization of an electron from an atom. The vector potential of the electromagnetic field is $\vec{A}(t)$, whereas the scalar potential $\phi = 0$. For the 20–500 eV photon energy range, the corresponding wavelength is 2.5–62 nm.

The field-free Hamiltonian of the atom is

$$H_0 = \frac{p^2}{2m_e} + V(\vec{r}), \quad (7.65)$$

where the momentum operator is

$$\vec{p} = i\hbar\nabla. \quad (7.66)$$

$V(\vec{r})$ is the potential energy of the electron in the atom. When the XUV field is weak, it adds a perturbation H' to the Hamiltonian. The perturbation can be expressed as

$$H'(t) = \frac{e}{2m_e c} (\vec{A}(t) \cdot \vec{p} + \vec{p} \cdot \vec{A}(t)) + \frac{e^2}{2m_e c^2} A^2(t). \quad (7.67)$$

7.5.2.1 Fermi's Golden Rule

Fermi's golden rule is a way to calculate the transition rate (probability of transition per unit time) from one energy eigenstate of a quantum system into a continuum of energy eigenstates, due to a perturbation. It is valid when the initial state has not been significantly depleted by scattering into the final states. We consider an atom to begin in an eigenstate ψ_i of a given Hamiltonian H_0 . For the RABITT experiments, it is the ground state.

7.5.2.2 First-Order Approximation

The transition rate from an initial state Ψ_i to a final state Ψ_f is given, to first order approximation in the perturbation, by

$$w \propto \frac{2\pi}{\hbar} |\langle \Psi_f | H' | \Psi_i \rangle|^2. \quad (7.68)$$

$\langle \Psi_f | H' | \Psi_i \rangle = M_{fi}$ is the transition matrix element of the perturbation H' between the final and initial states.

Because the XUV field is weak, we neglect the two-photon process contribution, i.e., we can set

$$A^2(t) = 0. \quad (7.69)$$

This is different from the case in Chapter 4 when the Volkov states are introduced.

In the weak field, the transition matrix element becomes

$$M_{f,i} = \frac{e}{2m_e c} \langle \Psi_f | \vec{A}(t) \cdot \vec{p} + \vec{p} \cdot \vec{A}(t) | \Psi_i \rangle. \quad (7.70)$$

The commutation can be expressed as

$$\vec{A}(t) \cdot \vec{p} + \vec{p} \cdot \vec{A}(t) = 2\vec{A}(t) \cdot \vec{p} + i\hbar \nabla \cdot \vec{A}(t). \quad (7.71)$$

It should be noted that we use the Coulomb gauge. By definition

$$\nabla \cdot \vec{A}(t) = 0, \quad (7.72)$$

then

$$M_{f,i} = \frac{e}{m_e c} \langle \Psi_f | \vec{A}(t) \cdot \vec{p} | \Psi_i \rangle. \quad (7.73)$$

It is called the “velocity” form because it contains p .

7.5.2.3 Dipole Approximation

From the commutation relations, we have

$$\langle \Psi_f | \vec{A}(t) \cdot \vec{p} | \Psi_i \rangle \propto \langle \Psi_f | \vec{A}(t) \cdot \nabla V(\vec{r}) | \Psi_i \rangle \propto \langle \Psi_f | \vec{A}(t) \cdot \vec{r} | \Psi_i \rangle. \quad (7.74)$$

Next, we can make the dipole approximation. Since the XUV light wavelength is much larger than the size of the atom (the range of r where the two wave functions are not vanished), we can assume $\vec{A}(t)$ is a constant in the integral. Finally, we have

$$M_{f,i} \propto \langle \Psi_f | \vec{r} | \Psi_i \rangle. \quad (7.75)$$

It is called the “length” form as it contains r .

7.5.2.4 Absorption Cross Section

The absorption cross section is defined as the ratio of the intensity absorbed by the atom to the incident intensity in the electromagnetic field

$$\sigma = \frac{4\pi^2}{3} \alpha a_0^2 \hbar \omega_X |M_{f,i}|^2. \quad (7.76)$$

Here, α is the fine-structure constant. $\hbar \omega_X$ is in Rydberg unit.

7.5.2.5 Neon Atom

The photoelectric emission in neon can be expressed by

$$Ne(1s^2 2s^2 2p^6; ^1S) \rightarrow Ne^+ \left(1s^2 2s^2 2p^5; ^2P_{\frac{1}{2}, \frac{3}{2}} \right) + e. \quad (7.77)$$

For unpolarized light, the partial cross section under the dipole approximation is

$$\frac{d\sigma(\omega_X, \theta)}{d\Omega} = \frac{\sigma(\omega_X)}{4\pi} [1 + \beta(\omega_X) P_2(\cos \theta)]. \quad (7.78)$$

Here, Ω is the solid angle, θ is the angle between the incoming photon and the emitted electron momenta, $\beta(\omega_X)$ is the asymmetry factor, and P_2 is

the second-order Legendre polynomial. For perfectly linearly polarized light, $P_2 = 0$ at $\theta = 54.7^\circ$, which is called the “magic angle.”

7.5.3 Transitions in XUV and IR Fields

7.5.3.1 Attosecond Pulse Train Generated with One-Color Driving Field

When the attosecond pulse train is generated with a multicycle laser pulses at the fundamental frequency, the two adjacent attosecond pulses in the train are separated by half a laser cycle. Consequently, the spacing between the two adjacent harmonics corresponding to the pulse train is two-laser photon energy, which is about 3 eV for Ti:Sapphire laser.

In the RABITT experiment, the field that causes the transition is a combination of the weak XUV field and the strong NIR laser field. The upper first-order sideband of the harmonic $q - 1$ overlaps and interferes with the lower first-order sideband of the harmonic $q + 1$, as shown in Figure 7.30. The intensity of the side band q signal can be calculated using the second-order perturbation theory.

For simplicity, we assume that the fields of the two contributing harmonics and the dressing laser are all monochromatic, i.e.,

$$\varepsilon_{q-1}(t) = E_{q-1}e^{i(\omega_{q-1}t + \phi_{q-1})}, \quad (7.79)$$

$$\varepsilon_{q+1}(t) = E_{q+1}e^{i(\omega_{q+1}t + \phi_{q+1})}, \quad (7.80)$$

$$\varepsilon_1(t) = E_1e^{i\omega_1 t} \text{ for absorption,} \quad (7.81)$$

$$\varepsilon_1(t) = E_1e^{-i\omega_1 t} \text{ for emission.} \quad (7.82)$$

According to Fermi’s golden rule with the second order approximation, the amplitude of the electron wave at the sideband energy $E_q = -E_b + q\hbar\omega_{IR}$ is proportional to

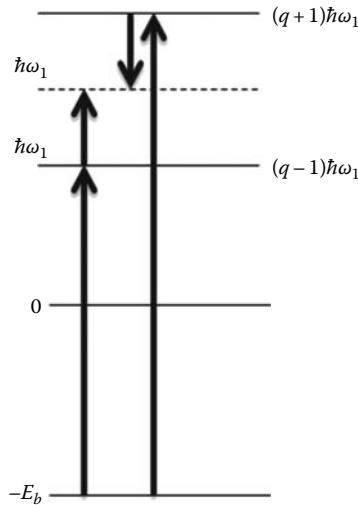


Figure 7.30 Interference at the inner sideband.

$$b_q(\omega, \tau_d) = \int_{-\infty}^{\infty} dt \left[M_{q,q-1} M_{q-1,g} \mathcal{E}_{q-1}(t) E_1 e^{i\omega_1(t-\tau_d)} + M_{q,q+1} M_{q+1,g} \mathcal{E}_{q+1}(t) E_1 e^{-i\omega_1(t-\tau_d)} \right] e^{-i\omega t}, \quad (7.83)$$

where τ_d is the time delay between the harmonic field and the laser field.

To simplify the analysis, we assume

$$M_{q,q-1} M_{q-1,g} = M_{q,q+1} M_{q+1,g} = M_1 M, \quad (7.84)$$

and

$$E_{q-1} = E_{q+1} = E_X, \quad (7.85)$$

which means that all the harmonic peaks have the same intensity. Then,

$$b_q(\omega, \tau_d) = M_1 M E_X E_1 \int_{-\infty}^{\infty} dt \times \left[e^{i(\omega_{q-1}t + \phi_{q-1})} e^{i\omega_1(t-\tau_d)} + e^{i(\omega_{q+1}t + \phi_{q+1})} e^{-i\omega_1(t-\tau_d)} \right] e^{-i\omega t} \quad (7.86)$$

or

$$b_q(\omega, \tau_d) = M_1 M E_X E_1 \int_{-\infty}^{\infty} dt \left[e^{i\phi_{q-1}} e^{-i\omega_1\tau_d} + e^{i\phi_{q+1}} e^{+i\omega_1\tau_d} \right] e^{i\omega_q t} e^{-i\omega t}. \quad (7.87)$$

Since $\int_{-\infty}^{\infty} dt e^{i\omega_q t} e^{-i\omega t} = \delta(\omega - \omega_q)$, we have

$$b_q(\omega_q, \tau_d) = M_1 M E_X E_1 \left[e^{i\phi_{q-1}} e^{-i\omega_1\tau_d} + e^{i\phi_{q+1}} e^{+i\omega_1\tau_d} \right]. \quad (7.88)$$

The intensity of the sideband

$$I_q(\tau_d) \propto |b_q(\omega_q, \tau_d)|^2 = 2(M_1 M)^2 I_X I_1 \times [1 + \cos(2\omega_1\tau_d + \phi_{q+1} - \phi_{q-1})]. \quad (7.89)$$

7.5.3.2 Sideband Intensity Oscillation

As the delay is scanned, the interference between the two transitions leads to an oscillation of the “inner” sideband. By experimentally recording the intensity of the sideband peak I_q as a function of τ_d , and fitting a cosine to this, we determine $\phi_{q+1} - \phi_{q-1}$. The phase differences between other harmonic peaks are obtained from other sidebands. Once the relative phase between all harmonics is known, one can retrieve trains of identical attosecond bursts. Since the width of each harmonic peak is not measured as they are assumed to be infinitely thin, this method does not require high spectral resolution. The intensity of the NIR laser must be sufficiently low so that a single NIR photon contributing to the final state, which allows only the phase difference of the neighboring harmonics to be measured. A rule of thumb is that the strengths of the sidebands should not exceed 50% of that of the $q-1$ and $q+1$ peaks. The intensity of laser cannot be too low otherwise $I_q(\tau_d)$ signal could be buried in the background noise.

7.5.3.3 Two-Color Driving Field

When a weak second harmonic field is added to the fundamental driving field, the symmetry breaking of the driving laser field leads to the generation of both odd and even harmonics. In other words, the harmonic peaks are separated by one photon energy, which is about 1.5 eV for Ti:Sapphire laser. In this case, one cannot use the inner sidebands between two harmonics for RABITT anymore. The interference of the transitions from the $q-1$ and $q+1$ peaks still occurs at q when the dressing laser at the fundamental wavelength is applied. Assuming $E_{q-1} = E_{q+1} = E_q = E_X$, the amplitude of the continuum is

$$b_q(\omega, \tau_d) = \int_{-\infty}^{\infty} dt [M_{q,q-1} M_{q-1,g} \mathcal{E}_{q-1}(t) E_1 e^{i\omega_1(t-\tau_d)} + M_{q,g} \mathcal{E}_q(t) + M_{q,q+1} M_{q+1,g} \mathcal{E}_{q+1}(t) E_1 e^{-i\omega_1(t-\tau_d)}] e^{-i\omega t}, \quad (7.90)$$

which leads to

$$b_q(\omega_q, \tau_d) = M E_q [M_{q,q-1} E_1 e^{-i\omega_1 \tau_d} e^{i\phi_{q-1}} + e^{i\phi_q} + M_{q,q+1} E_1 e^{+i\omega_1 \tau_d} e^{i\phi_{q+1}}]. \quad (7.91)$$

The parity of $M E_q e^{i\phi_q}$ is different from the other two states in Equation 7.91. Since most electron detectors have limited acceptance angle, the measured intensity may contain oscillation terms from the interference of these orthogonal states

$$I_q(\tau_d) = M^2 I_X [M_{q,q-1} E_1 (e^{-i\omega_1 \tau_d} e^{i\phi_{q-1}} e^{-i\phi_q} + e^{i\omega_1 \tau_d} e^{-i\phi_{q-1}} e^{i\phi_q}) + M_{q,q+1} E_1 (e^{+i\omega_1 \tau_d} e^{i\phi_{q+1}} e^{-i\phi_q} + e^{-i\omega_1 \tau_d} e^{-i\phi_{q+1}} e^{i\phi_q}) + M_{q,q-1} M_{q,q+1} E_1^2 (e^{+i2\omega_1 \tau_d} e^{i\phi_{q+1}} e^{-i\phi_{q-1}} + e^{-i2\omega_1 \tau_d} e^{-i\phi_{q+1}} e^{i\phi_{q-1}})], \quad (7.92)$$

i.e.,

$$I_q(\tau_d) = M^2 I_X [M_{q,q-1} \sqrt{I_1} \cos(\omega_1 \tau_d + \phi_q - \phi_{q-1}) + M_{q,q+1} \sqrt{I_1} \cos(\omega_1 \tau_d + \phi_{q+1} - \phi_q) + M_{q,q-1} M_{q,q+1} I_1 \cos(2\omega_1 \tau_d + \phi_{q+1} - \phi_{q-1})]. \quad (7.93)$$

Assuming $M_{q,q-1} = M_{q,q+1} = M_1$, then

$$I_q(\tau_d) = M_1 (M)^2 I_X \left[\sqrt{I_1} \cos\left(\frac{\phi_{q+1} + \phi_{q-1}}{2} - \phi_q\right) \times \cos\left(\omega_1 \tau_d + \frac{\phi_{q+1} - \phi_{q-1}}{2}\right) + M_1 I_1 \cos(2\omega_1 \tau_d + \phi_{q+1} - \phi_{q-1}) \right], \quad (7.94)$$

which oscillates with the laser frequency and is affected by the phase difference of adjacent harmonics. The superposition of the two oscillations produces periodic appearance of the maximum. However, the weak

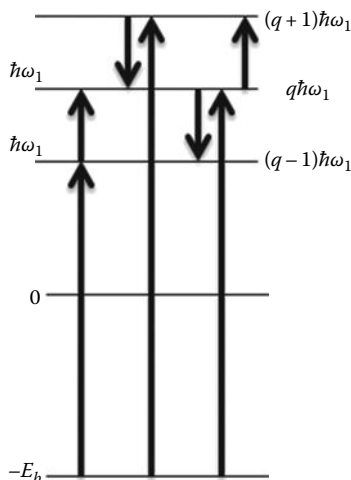


Figure 7.31 Interference signal overlaps with a harmonic peak.

interference signal is superimposed on the much stronger q th harmonic signal, as depicted in Figure 7.31.

This situation is very similar to the extraction of carrier-envelope phase from the single shot f -to- $2f$ traces. To extract the interference signal that oscillates with ω_1 or $2\omega_1$ frequency, one can perform a Fourier transfer of the signal of q harmonic. By applying a narrow window around the peak at ω_1 or $2\omega_1$, followed by an inverse Fourier transfer, one can obtain the $\cos[\omega_1\tau_d + (\phi_{q+1} - \phi_{q-1})/2]$, or, $\cos(2\omega_1\tau_d + \phi_{q+1} - \phi_{q-1})$ and thus $\phi_{q+1} - \phi_{q-1}$. The requirement on the dressing laser intensity is the same as the one-color driving laser case.

The required laser intensity is not related to the attosecond pulse duration to be measured. The delay step size is determined by sampling the ω_1 or $2\omega_1$ oscillation, which is independent of attosecond pulse duration.

7.6 Complete Reconstruction of Attosecond Bursts

As is mentioned in Section 7.5, the RABITT method assumes that the duration of the attosecond train does not change from pulse to pulse. In reality, this may not be true because the attosecond pulses in a train have different spectrum width as they are generated with different driving laser field strength. Another drawback of RABITT is that it cannot determine the number of attosecond pulses in a train because it assumes that each harmonic peak is a delta function. In other words, it assumes that there are infinite numbers of attosecond pulses in a train even when the duration of the driving laser is finite, which is obviously wrong.

The characterization of the XUV attosecond pulses can be performed using an extension of the FROG technique developed for characterizing femtosecond laser pulses. This procedure, called CRAB, has many advantages over RABITT. CRAB is based on the photoionization of atoms by

the XUV field, in the presence of a dressing (or streaking) NIR laser field, just like in the RABITT measurements. The XUV attosecond pulse ionizes a gas by single-photon absorption, thus, generating an attosecond electron pulse, which, far from any resonance, is a replica of the optical pulse. The NIR dressed photoelectron spectrum is measured as the time delay between the XUV pulse and the NIR field is changed. This yields a 2D set of data, called a spectrogram or CRAB trace. The XUV pulse duration can be retrieved by using various iterative reconstruction algorithms, which have been developed for FROG.

In the time domain, an isolated attosecond XUV pulse or a train of attosecond pulses can be described as

$$\varepsilon_X(t) = E_X(t)e^{i[\omega_X t + \phi_X(t)]}, \quad (7.95)$$

where the pulse envelope $E_X(t)$ and the temporal phase $\phi_X(t)$ need to be determined by the CRAB.

In the frequency domain, the XUV field can be described by the Fourier transform

$$\tilde{E}_X(\omega) = \int_{-\infty}^{+\infty} \varepsilon_X(t)e^{-i\omega t} dt. \quad (7.96)$$

The power spectrum of an XUV pulse can be measured with a spectrometer and is

$$I_X(\omega) = |\tilde{E}_X(\omega)|^2 = \left| \int_{-\infty}^{+\infty} \varepsilon_X(t)e^{-i\omega t} dt \right|^2. \quad (7.97)$$

The Fourier transform of $\sqrt{I_X(\omega)}$ gives the shortest pulses that can be supported by the measured spectrum. The pulse duration determined by the CRAB should be equal to or larger than the transform-limited value. In essence, CRAB measures the phase of $\tilde{E}_X(\omega)$. The major difference between CRAB and RABITT is the way that the spectral phase information is extracted from the measured trace. Unlike RABITT, CRAB can be used to characterize both pulse trains and single isolated attosecond pulses.

7.6.1 CRAB Trace

We consider an atom with ionization potential I_p , photoionized by a high-frequency XUV electric field $\varepsilon_X(t)$, in the presence of a low-frequency NIR laser field $\varepsilon_L(t) = -\partial \mathcal{A}_L / \partial t$. $\mathcal{A}_L$ is the vector potential of this laser field. In experiments, these fields are focused to the gas target. An example of the experimental setup is almost the same as that for the RABITT as shown in [Figure 7.28](#).

When there is a time shift, τ_d , between the XUV and the NIR pulses, the total field is

$$\vec{\varepsilon}(t, \tau_d) = \vec{\varepsilon}_X(t - \tau_d) + \vec{\varepsilon}_L(t). \quad (7.98)$$

From Chapter 5, we know that the complex amplitude of the electron wave packet in the continuum with momentum $\vec{v}$ is

$$b(\vec{v}, \tau_d) = -i \int_{-\infty}^{\infty} \vec{E}_X(t - \tau_d) \cdot \vec{d} \times [\vec{v} + \vec{A}_L(t)] e^{i\Phi_G(t)} e^{i[\omega_X(t - \tau_d) + \phi_X(t - \tau_d)]} e^{-i\left(\frac{v^2}{2} + I_p\right)t} dt, \quad (7.99)$$

$$\Phi_G(t) = - \int_t^{\infty} \left[\vec{v} \cdot \vec{A}_L(t') + \frac{1}{2} \vec{A}_L^2(t') \right] dt'. \quad (7.100)$$

$\Phi_G(t)$ is the quantum phase acquired by the electron due to its interaction with the laser field. The phase it accumulates along its trajectory is thus temporally modulated by the dressing field.

For the hydrogen atom

$$\vec{d}[\vec{v} + \vec{A}_L(t)] \propto \frac{\vec{v} + \vec{A}_L(t)}{\left((\vec{v} + \vec{A}_L(t))^2 + 2I_p \right)^{3/2}}. \quad (7.101)$$

The transition amplitude cannot be measured directly. The photoelectron spectrum is the power spectrum, not the wave amplitude, which can be represented by the expression

$$S_e(K, \tau_d) = \left| \int_{-\infty}^{\infty} \vec{E}_X(t - \tau_d) \cdot \vec{d}[\vec{v} + \vec{A}_L(t)] e^{i\Phi_G(\vec{v}, t)} e^{-i(K + I_p)t} dt \right|^2, \quad (7.102)$$

where K is the kinetic energy of the electron. The linear temporal phase $iI_p t$ corresponds to a frequency offset in the frequency domain, which can be dropped. Thus,

$$S_e(K, \tau_d) = \left| \int_{-\infty}^{\infty} \vec{E}_X(t - \tau_d) \cdot \vec{d}[\vec{v} + \vec{A}_L(t)] e^{i\Phi_G(\vec{v}, t)} e^{-iKt} dt \right|^2 \quad (7.103)$$

and

$$\Phi_G(\vec{v}, t) = - \int_t^{\infty} \left[\vec{v} \cdot \vec{A}_L(t') + \frac{1}{2} \vec{A}_L^2(t') \right] dt'. \quad (7.104)$$

Because of the scalar product $\vec{v} \cdot \vec{A}_L(t)$, the photoelectrons have to be observed in a given direction for the phase modulation to be well defined. $S_e(K, \tau_d)$ is the FROG-CRAB trace that can be measured experimentally.

In conventional femtosecond pulse measurements, the FROG trace is expressed as

$$S(\omega, \tau_d) = \left| \int_{-\infty}^{+\infty} \mathcal{E}_L(t - \tau_d) G(t) e^{-i\omega t} dt \right|^2. \quad (7.105)$$

Comparing Equations 7.104 and 7.102, we can see that $d[\vec{v} + \vec{A}_L(t)] e^{i\Phi_G(\vec{v}, t)}$ can be considered as a temporal gate, $G(t)$, in FROG. However, $G(t)$ does not depend on the frequency ω , whereas $d[\vec{v} - \vec{A}_L(t)]$ and $\varphi(\vec{v}, t)$ depend both on momentum and time, which makes it difficult to extract the phase of $\varepsilon_X(t)$ from the CRAB trace.

7.6.1.1 Temporal-Phase Gate

We remove the momentum dependence of these two terms by making the substitution $\vec{v} \rightarrow \vec{v}_0$, where $\vec{v}_0$ is the central momentum of the unstreaked electrons. The temporal phase can be rewritten as

$$\Phi_G(\vec{v}, t) \approx \Phi_G(\vec{v}_0, t) = - \int_t^\infty \left[\vec{v}_0 \cdot \vec{A}(t') + \frac{1}{2} A^2(t') \right] dt'. \quad (7.106)$$

$e^{i\Phi_G(t)}$ can be considered as a temporal-phase gate.

When $\varepsilon_X(t)$ represents an attosecond XUV pulse train or a single XUV pulse longer than the NIR laser cycle, the phase modulation on the photoelectron wave packet leads to the appearance of sidebands in the photoelectron energy spectrum, as in the case of RABITT. However, CRAB use the whole 2D trace, not just the sideband oscillation to extract the phase of $\tilde{E}_X(\omega)$, it is more accurate.

In the other limit, when $\varepsilon_X(t)$ represents an isolated attosecond XUV pulse significantly shorter than the dressing field optical period, the CRAB trace is the streaking camera trace for all τ_d , which covers many NIR laser cycles. Although the duration of the attosecond pulses can be obtained by comparing the distorted photoelectron spectrum measured at only one τ_d where the phase modulation is quadratic in time with the spectrum without streaking, the CRAB measurement which uses all the traces is more robust.

7.6.1.2 Reconstruction Algorithm

If we ignore the amplitude and phase variation of the dipole transition matrix element, i.e., assuming $d[\vec{v} + \vec{A}_L(t)] = 1$, then the CRAB trace can also be written as

$$S_e(\vec{v}, \tau_d) = \left| \int_{-\infty}^{\infty} \varepsilon_X(t - \tau_d) G(t) e^{-i\left(\frac{v^2}{2} + I_p\right)t} dt \right|^2, \quad (7.107)$$

where the phase gate

$$G(t) = e^{i\Phi_G(t)}. \quad (7.108)$$

Equation 7.107 reveals that such a CRAB trace can be interpreted as a spectrogram similar to a FROG trace. Such a spectrogram can be processed using a FROG retrieval algorithm to fully characterize the XUV pulse as well as the electric field of the NIR pulse. The full characterization of these wave packets provides all the information on the temporal structure of the attosecond XUV fields regardless whether it is pulse train or an isolated pulse.

Various iterative algorithms, such as the very efficient principal component generalized projections algorithm (PCGPA), can be used to extract both $\varepsilon_X(t)$ and $G(t)$ from $S_e(\vec{v}, \tau_d)$. The algorithm is based on generalized projections, which means that it adopts the approach of applying alternating constraints between the time and frequency domains. The frequency domain constraint consists of replacing the calculated spectrogram's intensity with the measured one, whereas the time domain constraint involves an optimization step to find the pulse and gate pair that can best reproduce the signal matrix, i.e., the one resulting from the frequency domain constraint.

Due to the high redundancy of information in the CRAB trace, it is very robust against noise, which is discussed in detail in Chapter 8. The identical pulse duration assumption for RABITT is not required by the FROG-CRAB so that it gives both the number of pulses and duration of each pulse. For this method, a single isolated attosecond pulse is just a special case of a pulse train, thus, can also be measured.

7.6.2 Linearly Polarized Dressing Laser Field

We consider the particular case of a linearly polarized dressing laser field

$$\varepsilon_L(t) = E_0(t) \cos(\omega_0 t). \quad (7.109)$$

A simple expression of phase gate is then obtained if the envelope $E_0(t)$ of the laser field is long enough to apply the slowly-varying envelope approximation, so that the vector potential is

$$\mathcal{A}_L(t) = - \int \varepsilon_L(t) dt = - \int E_0(t) \cos(\omega_0 t) dt \approx - \frac{E_0(t)}{\omega_0} \sin(\omega_0 t). \quad (7.110)$$

The phase gate

$$\Phi_G(\vec{v}, t) = - \int_t^\infty \left[\vec{v}_0 \cdot \vec{\mathcal{A}}_L(t') + \frac{1}{2} \vec{\mathcal{A}}_L^2(t') \right] dt' \quad (7.111)$$

becomes

$$\begin{aligned} \Phi_G(t) = & - \int_t^\infty \left[-v_0 \cos \theta \frac{E_0(t)}{\omega_0} \sin(\omega_0 t') \right] dt' \\ & - \int_t^\infty \frac{1}{2} \left[-\frac{E_0(t)}{\omega_0} \sin(\omega_0 t') \right]^2 dt'. \end{aligned} \quad (7.112)$$

The observation angle θ is the angle between $\vec{v}_0$ which is the direction of the electron after leaving the NIR field and the laser polarization direction. The phase can be resolved into three components

$$\Phi_G(t) = \Phi_1(t) + \Phi_2(t) + \Phi_3(t), \quad (7.113)$$

where

$$\Phi_1(t) = - \int_t^\infty \vec{v}_0 \cdot \vec{\mathcal{A}}_L(t') dt' = v_0 \cos \theta \frac{E_0(t)}{\omega_0^2} \cos(\omega_0 t). \quad (7.114)$$

In terms of energy

$$\Phi_1(t) = \cos \theta \frac{\sqrt{8K_0 U_p(t)}}{\omega_0} \cos(\omega_0 t). \quad (7.115)$$

Here, the kinetic energy $K_0 = (1/2)v_0^2$ and the ponderomotive energy $U_p(t) = E_0^2(t)/4\omega_0^2$.

We emphasize that around $\theta = 0$, the amplitude $\Phi_1(t)$ of the phase modulation reaches large values ($\Phi_1 \sim 2\pi$) even at moderate laser intensities due to the $\sqrt{8K_0}$ factor. At $\theta = 90^\circ$, the contribution from this term is zero.

7.6.2.1 Energy Shift

The kinetic energy shift at $\theta = 0$ due to $\Phi_1(t)$ is

$$\Delta K(t) = \Delta \omega_1(t) = \frac{d\Phi_1(t)}{dt} = -\sqrt{8K_0 U_p(t)} \sin(\omega_0 t), \quad (7.116)$$

which is the foundation of the attosecond streaking.

7.6.2.2 Phase and Laser Field

In the situation where $\Phi_1(t)$ is the dominating term in $\Phi_G(t)$ and can be obtained from the CRAB reconstruction, then the laser field can be deduced from the measured phase:

$$\varepsilon_L(t) = \frac{\omega_0^2}{v_0 \cos \theta} \Phi_1(t). \quad (7.117)$$

This relationship is used to directly map out the laser field, as discussed in Chapter 9.

7.6.2.3 Ponderomotive Shift

Using the identity

$$\sin^2(\omega_0 t') = \frac{1}{2} [1 - \cos(2\omega_0 t')], \quad (7.118)$$

we have

$$\begin{aligned} & \int_t^\infty \frac{1}{2} \left[-\frac{E_0(t)}{\omega_0} \sin(\omega_0 t') \right]^2 dt' \\ & \approx \int_t^\infty \frac{E_0^2(t)}{4\omega_0^2} dt' - \int_t^\infty \frac{E_0^2(t)}{4\omega_0^2} \cos(2\omega_0 t') dt'. \end{aligned} \quad (7.119)$$

The other two phase components are defined as

$$\Phi_2(t) = \int_t^{+\infty} \frac{E_0^2(t)}{4\omega_0^2} \cos(2\omega_0 t') dt' = \frac{U_p(t)}{2\omega_0} \sin(2\omega_0 t) \quad (7.120)$$

and

$$\Phi_3(t) = - \int_t^{\infty} \frac{E_0^2(t')}{4\omega_1^2} dt' = - \int_t^{+\infty} U_p(t') dt'. \quad (7.121)$$

$\Phi_3(t)$ is the origin of the ponderomotive shift. The energy shift due to this term is found from

$$\Delta\omega_3(t) = \frac{d\Phi_3(t)}{dt} = - \int_t^{\infty} \frac{d}{dt'} U_p(t') dt' = -U_p(\infty) + U_p(t) = U_p(t). \quad (7.122)$$

The shift follows the intensity profile of the dressing laser pulse, $I_L(t)$, since $U_p(t) \propto I_L(t)$. At $3.3 \times 10^{13} \text{ W/cm}^2$, the maximum shift at the Ti:Sapphire wavelength is 200 meV. The experimental evidence of the shift is shown in the insert of Figure 7.25b.

The energy shift caused by the second phase term is

$$\Delta\omega_2(t) = \frac{d\Phi_2(t)}{dt} = U_p(t) \cos(2\omega_0 t), \quad (7.123)$$

$\Phi_1(t)$ and $\Phi_2(t)$ oscillate, respectively, at the laser-field frequency and its second harmonic, so do their corresponding energy shift. At 10^{12} W/cm^2 , $K_0 \gg U_p$, $\sqrt{8KU_p} \gg U_p$, thus, $\Phi_1(t)$ is the dominating term at $\theta = 0$.

7.6.2.4 NIR Laser Intensity

In the attosecond streak camera measurement, the photoelectron is measured at the delay τ_d where the laser vector potential crosses zero. The photoelectron spectrum is broadened due to the fact that electrons in the leading edge of the electron pulse are energy shifted by a different amount as compared to that in the trailing edge. The amount of broadening must be comparable to the spectrum width of the attosecond pulse to reliably deduce the pulse duration. In the language of phase gate, this means that the bandwidth of the temporal phase $\Phi_1(t)$ should be a significant fraction of that of the attosecond field to be characterized, which will typically range from a few electron volts to tens of electron volts. Thus, the intensity of the streaking NIR laser must be sufficiently high.

The bandwidth of this filter can be defined as the maximum value in terms of $\partial\Phi_1(t)/\partial t$. Following the semiclassical model, this corresponds to the maximum energy shift of the photoelectron spectrum that is induced by the laser field. For $K_0 = 100 \text{ eV}$, $\theta = 0$, $I_{L0} = 10^{13} \text{ W/cm}^2$, $\lambda_0 = 800 \text{ nm}$, this electron-phase modulator has a bandwidth of

$$|\Delta\omega_1|_{\max} = \left| \frac{\partial\Phi_G}{\partial t} \right|_{\max} = \left| \frac{\partial\Phi_1}{\partial t} \right|_{\max} = \sqrt{8K_0 U_p} = 5 \text{ rad/fs} = 20 \text{ eV}, \quad (7.124)$$

which is the FWHM of transform-limited 94 as Gaussian pulse.

The bandwidth as a function of laser intensity and center energy is shown in Figure 7.32. It obviously increases with the laser intensity. At

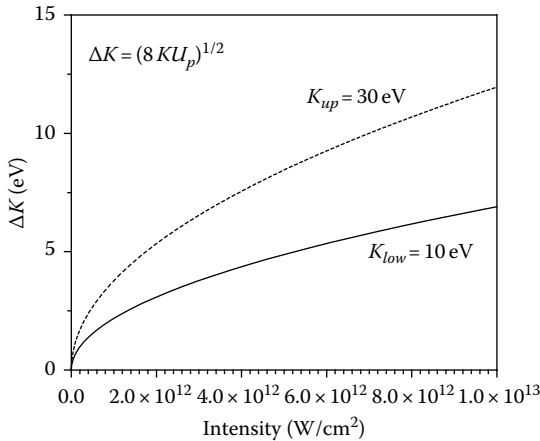


Figure 7.32 Phase-modulation bandwidth as a function of laser intensity.

the same streaking intensity, the bandwidth is proportional to the wavelength of the streaking field. Using difference frequency generation in nonlinear crystal, it is possible to obtain streaking field with twice the wavelength of the laser for generating the attosecond pulses.

The temporal resolution that can be achieved for a given bandwidth of the modulator depends on the specific measurement technique that is implemented. For RABITT measurement, the required NIR intensity is on the order of $10^{11} - 10^{12} \text{ W}/\text{cm}^2$. In the first RABITT experiments by Paul et al., the laser intensity is $< 10^{12} \text{ W}/\text{cm}^2$ and the energy of the electron is in the 2–16 eV range. The bandwidth of the phase filter is $\sim 2 \text{ eV}$ for $K_0 = 10 \text{ eV}$, which is smaller than the 7.5 eV FWHM spectral width of the measured 250 as pulses. The requirement of the laser intensity for CRAB is discussed in Chapter 8.

7.6.2.5 Observation Angle

Measurements around $\theta = 0$ can be performed with a large angular acceptance. However, the $\Phi_1(t)$ term which dominates in this angular range depends on the electron energy. The phase modulation experienced by an electron wave packet is therefore not homogenous across its spectrum. When the central momentum approximation is used, this dependence will thus generally introduce systematic errors in the retrieved attosecond pulse, if it has a large bandwidth ($\Delta K_0 \approx K_0$).

Such systematic errors do not occur at $\theta = \pi/2$, where $\Phi_2(t)$ and $\Phi_3(t)$ dominate, but measurements then have to be carried out with a much smaller collection angle (typically only a few degrees), which significantly reduces the electron counts. The experiment also needs much higher laser intensity to achieve the same modulation bandwidth. In this case, one does not need to make the Central Momentum Approximation since these two phase terms do not depend on the momentum of the electron. Thus it may be useful for characterizing extremely short attosecond pulses.

7.6.3 Attosecond Pulse Train

CRAB can be used for the complete characterization of trains of attosecond pulses. The spacing between two adjacent attosecond pulses in the train, T_{tr} , is half of optical cycle T_0 of the fundamental NIR laser field that generate the attosecond pulse train. When the driving field is the combined fundamental laser field and its second harmonic, the spacing between the adjacent attosecond pulses is one laser cycle. Both types of attosecond train can be characterized by the CRAB method using the same NIR laser for modulating the phase of the photoelectrons. In other words, the period of the phase modulator, T_m , is the same as T_0 , although the corresponding CRAB traces are somewhat different. Actually, CRAB requires no specific relationship between, T_{tr} and T_m .

When the dressing intensity is high and two or more NIR photons are involved in each transition, then smaller delay step size is needed to sample the high-frequency oscillations as a result of the interference between high-order transition channels.

7.6.3.1 $T_m = 2T_{tr}$

Only the fundamental laser field is used in generating the attosecond pulse train. The information on the attosecond pulses temporal structure is encoded in the sideband interference pattern. By scanning the delay until the two fields no longer overlap, trains of nonidentical pulses can be retrieved. To obtain the reconstruction, the full photoelectron spectrum is injected in the retrieval algorithm. However, a much higher spectral resolution than RABITT is required because the femtosecond envelope of the attosecond train needs to be measured. It is determined numerically that for an accurate retrieval, a resolution of 100 meV sets an upper limit of about 8 fs on the duration of the overall envelope of the attosecond train.

7.6.3.2 Attosecond Pulses near the Cutoff Region

[Figure 7.33](#) shows the CRAB trace near the cutoff region of the high harmonic spectrum obtained when an 800 nm, 7 fs chirped laser pulse. As is discussed in Chapter 8, the cutoff region of the high-harmonic spectrum merged to a continuum when the driving laser approaches a single-laser cycle, which is named amplitude gating. When the carrier-envelope phase is chosen properly, a single isolated attosecond pulse can be extracted from the continuous spectrum. The plateau region still has discrete harmonics.

Both attosecond streaking of an isolated pulse in the upper part and the oscillating sidebands in the discrete lower part are observed in the CRAB trace. A gradual transition occurs between these two regimes. The intensity $5 \times 10^{10} \text{ W/cm}^2$ are used as a phase gate. This shows that CRAB is a powerful method that allows us to study the transition regime between attosecond trains and single attosecond pulses.

7.6.4 Perturbative Regime of CRAB

When the energy shift of the streaking is much less than one NIR photon energy, i.e., $\sqrt{8K_0U_p} \ll \omega_1$,

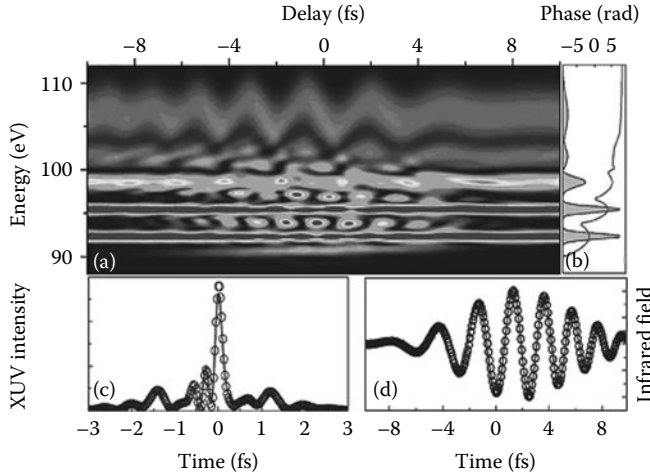


Figure 7.33 (a) CRAB trace that shows the transition from attosecond pulse train to single isolated pulses. (b) Retrieved power spectrum and phase. (c) Reconstructed XUV pulse. (d) The streaking IR field. (Reprinted with permission from Y. Mairesse and F. Quéré, *Phys. Rev. A*, 71, 011401(R), 2005. Copyright 2005 by the American Physical Society.)

$$e^{i\Phi_G(t)} = \cos[\Phi_G(t)] + i \sin[\Phi_G(t)] \approx 1 - \frac{\Phi_G^2(t)}{2} + i\Phi_G(t) \\ \approx 1 + i\Phi_1(t) - \frac{\Phi_1^2(t)}{2}. \quad (7.125)$$

For simplicity, we assume that the dipole moment element is a constant, $d(\vec{v}) = d$, then

$$b(\vec{v}, \tau_d) \approx d \int_{-\infty}^{\infty} \varepsilon_X(t - \tau_d) \left[1 + i\Phi_1(t) - \frac{\Phi_1^2(t)}{2} \right] e^{-i\left(\frac{v^2}{2} + I_p\right)t} dt, \quad (7.126)$$

The two phase terms can be rewritten as

$$\Phi_1(t) = v \frac{E_1}{\omega_1^2} \cos(\omega_1 t) = \frac{vE_1}{2\omega_1^2} (e^{i\omega_1 t} + e^{-i\omega_1 t}), \quad (7.127)$$

$$\Phi_1^2(t) = \left(\frac{vE_1}{2\omega_1^2} \right)^2 (2 + e^{i2\omega_1 t} + e^{-i2\omega_1 t}). \quad (7.128)$$

7.6.4.1 Attosecond Pulse Train Generated with One-Color Lasers

We consider the case that there are only two harmonics

$$\varepsilon_X(t) = E_X e^{i\omega_{q-1}t} e^{i\phi_{q-1}} + E_X e^{i\omega_{q+1}t} e^{i\phi_{q+1}} \quad (7.129)$$

or

$$\varepsilon_X(t) = E_X e^{i\omega_q t} (e^{-i\omega_1 t} e^{i\phi_{q-1}} + e^{i\omega_1 t} e^{i\phi_{q+1}}). \quad (7.130)$$

By just keeping the first order of the phase modulation, $\Phi_1(t)$, the continuum amplitude can be expressed as

$$b(v, \tau_d) \approx E_X e^{-i\omega_q \tau_d} d \int_{-\infty}^{\infty} \left[e^{-i\omega_1(t-\tau_d)} e^{i\phi_{q-1}} + e^{i\omega_1(t-\tau_d)} e^{i\phi_{q+1}} \right] \times \left[1 + i \frac{v E_1}{2\omega_1^2} (e^{i\omega_1 t} + e^{-i\omega_1 t}) \right] e^{i(\omega_q - \frac{v^2}{2} - I_p)t} dt. \quad (7.131)$$

Using the property of the δ function

$$\int_{-\infty}^{\infty} e^{i\omega t} dt = \delta(\omega), \quad (7.132)$$

we obtain the amplitude of the q th peak from the term

$$b(v_q, \tau_d) \approx E_X E_1 d \frac{v_q}{2\omega_1^2} e^{-i\omega_q \tau_d} \int_{-\infty}^{\infty} dt \times (e^{i\omega_1 \tau_d} e^{i\phi_{q-1}} + e^{-i\omega_1 \tau_d} e^{i\phi_{q+1}}) e^{i(\omega_q - \frac{v_q^2}{2} - I_p)t} dt. \quad (7.133)$$

where $v_q = \sqrt{2(\omega_q - I_p)}$ is the momentum of the electron ionized by the q th harmonic alone.

Finally,

$$b(v_q, \tau_d) \approx E_X E_1 d \frac{v_q}{2\omega_1^2} e^{-i\omega_q \tau_d} (e^{i\omega_1 \tau_d} e^{i\phi_{q-1}} + e^{-i\omega_1 \tau_d} e^{i\phi_{q+1}}), \quad (7.134)$$

which is essentially the same as the expression for the sidebands of RABITT. The intensity oscillation is expressed by the $\cos(2\omega_q \tau_d)$ interference term.

7.6.4.2 Attosecond Pulse Train Generated with Two-Color Lasers

Both odd and even high harmonics can be generated simultaneously with the two-color field which is combined fundamental laser field and its second harmonic. In the time domain, the spacing between the adjacent attosecond pulses is one laser cycle. The XUV field

$$\mathcal{E}_X(t) = \sum_{q=q_{min}}^{q_{max}} e^{i\omega_q t} e^{i\phi_q}. \quad (7.135)$$

As an example, we consider the case where there are only three harmonics

$$\mathcal{E}_X(t) = E_{q-1} e^{i\omega_{q-1} t} e^{i\phi_{q-1}} + E_q e^{i\omega_q t} e^{i\phi_q} + E_{q+1} e^{i\omega_{q+1} t} e^{i\phi_{q+1}}. \quad (7.136)$$

For simplicity, we assume that the amplitude of the three fields components are the same, i.e.,

$$\mathcal{E}_X(t) = E_X e^{i\omega_q t} (e^{-i\omega_1 t} e^{i\phi_{q-1}} + e^{i\phi_q} + e^{i\omega_1 t} e^{i\phi_{q+1}}). \quad (7.137)$$

The continuum amplitude

$$b(v, \tau_d) \approx E_X e^{-i\omega_q \tau_d} d \int_{-\infty}^{\infty} [e^{-i\omega_1(t-\tau_d)} e^{i\phi_{q-1}} + e^{i\phi_q} + e^{i\omega_1(t-\tau_d)} e^{i\phi_{q+1}}] \times \left[1 + i \frac{v E_1}{2\omega_1^2} (e^{i\omega_1 t} + e^{-i\omega_1 t}) \right] e^{i(\omega_q - \frac{v^2}{2} - I_p)t} dt. \quad (7.138)$$

Like in the one-color case, we obtain the peak with momentum v_q

$$b(v_q, \tau_d) \approx E_X e^{-i\omega_q \tau_d} d \times \left[i \frac{v_q E_1}{2\omega_1^2} (e^{i\omega_1 \tau_d} e^{i\phi_{q-1}} + e^{-i\omega_1 \tau_d} e^{i\phi_{q+1}}) + e^{i\phi_q} \right], \quad (7.139)$$

which is similar to RABBIT. The last term is the contribution from the ionization by the q th harmonic alone. Unlike the one-color case, now the interference signal $(v_q E_1)/(2\omega_1^2) (e^{i\omega_1 \tau_d} e^{i\phi_{q-1}} + e^{-i\omega_1 \tau_d} e^{i\phi_{q+1}})$ is superimposed on a strong background $e^{i\phi_q}$. The contrast is determined by $(v_q E_1)/(2\omega_1^2)$, which is higher for electrons with large momentum and for higher addressing laser intensity. The absolute value of the interference term is the same as that of the one-color case.

The intensity

$$|b(K_q, \tau_d)|^2 = E_X^2 d^2 \left\{ 1 + \frac{4\sqrt{8K_q U_p}}{\omega_1} \sin\left(\phi_q - \frac{\phi_{q-1} + \phi_{q+1}}{2}\right) \times \cos\left(\omega_1 \tau_d + \frac{\phi_{q-1} + \phi_{q+1}}{2}\right) + \frac{16K_q U_p}{\omega_1^2} \times [1 + \cos(2\omega_1 \tau_d + \phi_{q-1} - \phi_{q+1})] \right\}. \quad (7.140)$$

The NIR field is fixed while the XUV field is delayed by τ_d . This definition is different from what is used in the RABITT discussion. The intensity contains a strong $\omega_1 \tau_d$ and a weaker $2\omega_1 \tau_d$ oscillation components, both contains the phase difference $\phi_{q+1} - \phi_{q-1}$ information.

As an example, we consider an XUV pulse is a superposition of seven harmonics of Ti:Sapphire laser centered at 800 nm, $q = 17, 18, 19, 20, 21, 22, 23$. Their amplitudes are the same. Their phase

$$\phi_q = -0.5(q - 20)^2, \quad (7.141)$$

which is shown in [Figure 7.34](#). Such a parabolic phase corresponds to a train of positively chirped pulses.

The XUV pulse is converted to an electron pulse in neon gas, which has an ionization potential of 21.56 eV. The photon, electron energies, and phases of the seven harmonics are listed in [Table 7.1](#).

When the intensity of the 800 nm dressing laser is 1×10^{10} W/cm², the ratio $\sqrt{8K_0 U_p}/\omega_1 = 0.137$ for the $q = 20$, which satisfies the perturbative condition. The calculated intensities of the three dressed electron peaks corresponding to $q = 19, 20, 21$ using Equation 7.139 are shown in [Figure 7.35](#).

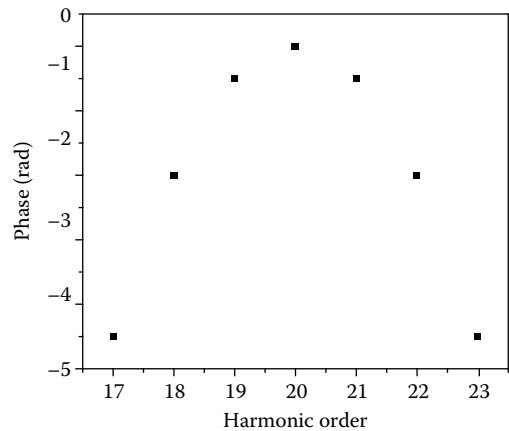


Figure 7.34 Phases of seven harmonics.

TABLE 7.1 Photon, Electron Energies, and Phases of the Seven Harmonics

<i>Q</i>	17	18	19	20	21	22	23
Photon (eV)	26.35	27.9	29.45	31	32.55	34.1	35.65
Electron (eV)	4.79	6.34	6.34	9.44	10.99	12.54	14.09
Phase (rad)	−4.5	−2	−0.5	0	−0.5	−2	−4.5

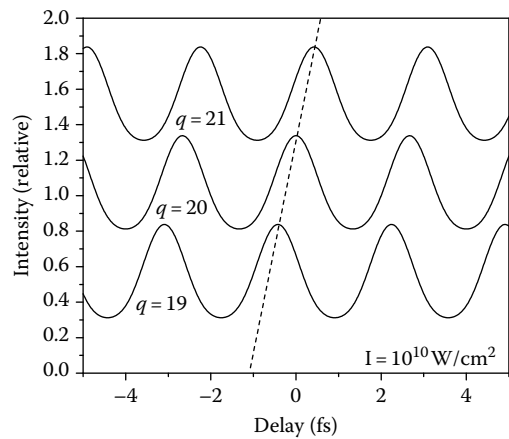


Figure 7.35 Laser-dressed electron intensities as a function of delay.

At such a low dressing-laser intensity, the $\cos(\omega_1\tau_d + (\phi_{q-1} - \phi_{q+1})/2)$ dominates the electron intensity variation. The horizontal shift of the three curves is due to the difference caused by the phase $(\phi_{q-1} - \phi_{q+1})/2$.

The CRAB can retrieve the phase information by fitting these oscillations which exists at dressing laser intensities on the same order of magnitude of RABBIT. In other words, even when the pulse train is

generated with two-color lasers, the CRAB should be able to retrieve the attosecond pulse at much lower intensity than what is required by the attosecond streak camera.

7.7 Summary

Attosecond pulse trains are easy to generate. The measurement of the XUV pulse duration can be accomplished by using either RABITT or CRAB. As compared to CRAB, RABITT does not require high spectral resolution, and can be performed with much low NIR laser intensity, thus is commonly used for pulse trains. On the contrary, isolated pulses are frequently characterized by CRAB.

Problems

- 7.1 A truncated Gaussian beam centered at $0.8\ \mu\text{m}$ is focused by a mirror with a focal length of $f=250\text{ mm}$. The $1/e^2$ radius of the input beam is $w=8\text{ mm}$. The outside radius of the annular aperture is $b=25\text{ mm}$. Compare the transverse electric field distribution between $a=5\text{ mm}$ and $a=10\text{ mm}$.
- 7.2 A truncated Gaussian beam centered at $0.8\ \mu\text{m}$ is focused by a mirror with a focal length of $f=250\text{ mm}$. The $1/e^2$ radius of the input beam is $w=8\text{ mm}$. The outside radius of the annular aperture is $b=25\text{ mm}$. Compare the Gouy phase between $a=5\text{ mm}$ and $a=10\text{ mm}$.
- 7.3 The photon energy of an attosecond pulse extends from 25 to 50 eV. When argon is used as the detection atom, what is the corresponding kinetic energy range of the photoelectron? Compare it with the case when neon is used to detect the pulse.
- 7.4 The drift tube of a field free electron TOF spectrometer is 3 m. Suppose the energy resolution of the detector and electrons is 250 ps. What are the energy resolutions for the 100 and 250 eV electrons.
- 7.5 Redo the calculations in Problem 7.4 for detectors with 100 ps and 1 ns time resolution, respectively.
- 7.6 In Problem 7.4, consider two MCP detectors, one has a 25 mm in diameter and the other is 50 mm in diameter. Calculate the solid angles that the electrons can be detected for these two detectors.
- 7.7 In Problem 7.4, if the 100 eV electron is slowed down to 10 eV by a retarding mesh at the entry of the TOF, what will be energy resolution?
- 7.8 Calculate the cyclotron frequency of an electron in a 1 T magnetic field.
- 7.9 Calculate the cyclotron radius of an electron emitted with 45° angle to the z axis and with 10 eV initial energy. Compare it with the result for an electron with 100 eV energy.
- 7.10 Suppose the magnetic fields in the gas target and in the detection regions are 0.5 T and 1 mT, respectively. Calculate the angle of the electron in the drift tube for electrons that are initially emitted perpendicularly to the field lines in the high-field region.

- 7.11** Consider two electrons with 10 eV energy emitted at $\Theta_i = 0^\circ$ and $\Theta_i = 90^\circ$, respectively. Suppose also that $B_f/B_i = 10^{-3}$. For a 3 m long drift tube, calculate their flight time difference.

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Magnetic Bottle TOF

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Single Isolated Attosecond Pulses

When noble gas atoms are driven by linearly polarized intense near-infrared (NIR) laser pulses containing multiple optical cycles, a train of attosecond pulses can be generated, as discussed in Chapter 7. The separation between adjacent pulses is half an optical cycle of the driving laser pulses, which is 1.3 fs for Ti:Sapphire lasers. Attosecond pulse trains with such a short inter-pulse spacing have limited use in pump–probe experiments where the process to be studied may last longer than the pulse spacing. Instead, single isolated attosecond pulses are highly desirable for initiating and probing the dynamics.

Once an attosecond pulse train leaves a gas target, it is extremely difficult to switch out a single isolated pulse from the train like we pick up a femtosecond pulse from a laser oscillator with a Pockels cell. For that to happen, we would need a pulse picker that opens for less than an optical cycle because of short spacing between the pulses. Various sub-cycle gating methods that have been proposed and demonstrated are implemented in the generation stage instead. Three of them are introduced in this chapter. With these schemes, none of the pulses in the train except one get a chance to be born from the generation gas target.

We first discuss the methods for characterizing isolated attosecond pulses.

8.1 Phase Retrieval by Omega Oscillation Filtering

The femtosecond pulse characterization methods described in Chapter 2 are based on nonlinear optical effects such as second harmonic generation, but it is very difficult to apply them to attosecond pulses because of the low XUV photon flux. The attosecond streak camera concept introduced in Chapter 1 works by converting time information into momentum information, which is discussed in the next section. Here, we discuss the methods for measuring both the shape and phase of single isolated attosecond pulses using a scheme called phase retrieval by omega oscillation filtering (PROOF).

8.1.1 Introduction

In the time domain, an isolated attosecond XUV pulse can be described by its electric field as

$$\varepsilon_X(t) = E_X(t)e^{i[\omega_X t + \phi_X(t)]}, \quad (8.1)$$

where ω_X is the central frequency. The pulse envelope $E_X(t)$ and the temporal phase $\phi_X(t)$ are the two quantities that need to be determined.

As an example, a linearly chirped XUV pulse with a Gaussian shape can be expressed by

$$\varepsilon_X(t) = E_{X0} e^{-2 \ln 2 \frac{t^2}{\tau_X^2}} e^{i(\omega_X t + b t^2)}, \quad (8.2)$$

where

E_{X0} is the peak amplitude of the field

b is the chirp parameter of the XUV pulse

τ_X is the pulse duration, defined as the full width at the half maximum (FWHM) of the intensity profile $|\varepsilon_X(t)|^2$

For such pulses, the quantities to be determined are τ_X and b . Pulse duration is the most important parameter for specifying an attosecond pulse.

The attosecond pulse can also be described by the Fourier transform

$$\varepsilon_X(t) = \int_{-\infty}^{\infty} U(\omega) e^{i\varphi(\omega)} e^{i\omega t} d\omega, \quad (8.3)$$

where $U(\omega)$ and $\varphi(\omega)$ are the spectrum amplitude and phase.

The power spectrum of an XUV pulse is

$$I_X(\omega) = |U(\omega)|^2. \quad (8.4)$$

As an example, we consider a chirped Gaussian XUV pulse with 18.25 eV bandwidth centered at $\hbar\omega_X = 121.56$ eV, which supports a 100 as pulse according to

$$\tau_{X0}(\text{as}) = \frac{1835}{\Delta\varepsilon_X(\text{eV})}, \quad (8.5)$$

where τ_{X0} is the transform-limited pulse duration in attoseconds. $\Delta\varepsilon_X$ is the full width at half maximum (FWHM) bandwidth of the power spectrum. The power spectrum with the given bandwidth is shown in [Figure 8.1](#).

We consider two linearly chirped pulses. The degrees of chirp are specified by the spectral phases $-(d^2\varphi/d\omega^2)|_{\omega_X} = +5000 \text{ as}^2$ and $-(d^2\varphi/d\omega^2)|_{\omega_X} = -5000 \text{ as}^2$, respectively. The negative sign before the second derivative is from the convention discussed in Chapter 2. The corresponding spectral phases are shown in [Figure 8.2](#).

The power spectrum distribution $I_X(\omega)$ can be measured using conventional spectroscopy methods such as a grating spectrometer or a photoelectron spectrometer; thus, the main task of attosecond pulse characterization is the measurement of the spectrum phase $\varphi(\omega)$. Once

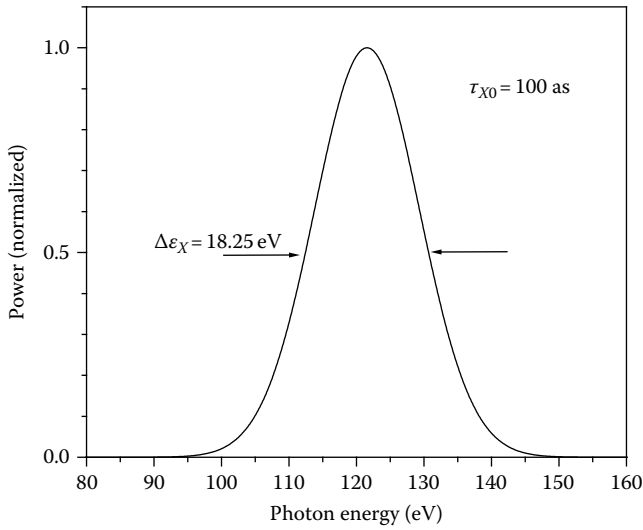


Figure 8.1 The power spectrum of an XUV pulse that supports a 100 as pulse.

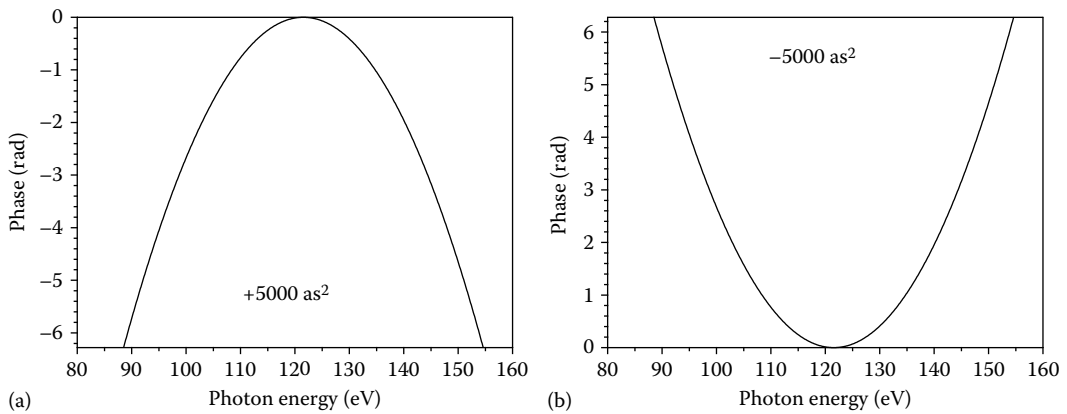


Figure 8.2 The spectrum phases of the (a) positively and (b) negatively chirped pulses.

the spectrum phase is known, the duration of the chirped XUV pulse can be determined. For linearly chirped Gaussian pulses, the pulse duration

$$\tau_X = \tau_{X0} \sqrt{1 + \left(4 \ln 2 \frac{\frac{d^2 \varphi}{d\omega^2} \big|_{\omega_X}}{\tau_{X0}^2} \right)^2}. \quad (8.6)$$

For the chirp values in the example, the duration of the two pulses is $\tau_X = 171$ as, regardless of the sign of the chirp. The comparison between the transform-limited pulse and the chirped pulse is illustrated in [Figure 8.3](#). In general, for an arbitrary power spectrum, once the spectrum phases at each frequency are measured, the XUV pulse in the time domain can be obtained by using Fourier transforms.

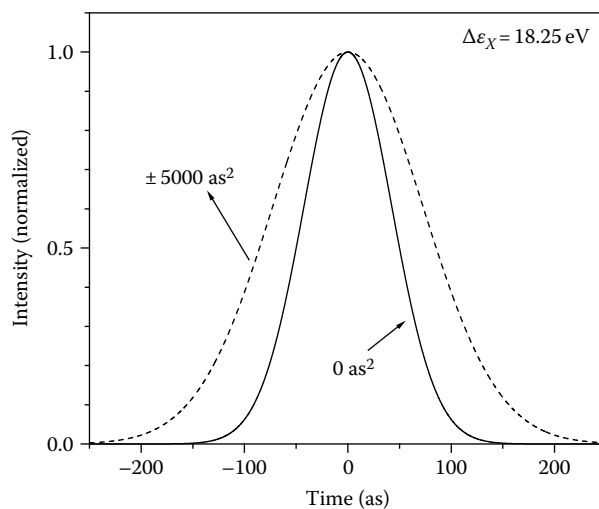


Figure 8.3 The effects of chirp on attosecond pulse duration.

8.1.2 Phase Encoding in Electron Spectrograph

For the characterization of single isolated attosecond XUV pulses, the photon pulse is converted to an electron pulse through the photoelectric effect. The electron pulse interacts with an NIR dressing laser to pull out the XUV phase information. A schematic diagram of the experimental setup is depicted in Figure 8.4.

When an atom with ionization potential I_p absorbs an XUV photon, a free electron with momentum $\vec{v}$ is produced through dipole transition from the ground state. The ionization potential of the commonly used detection

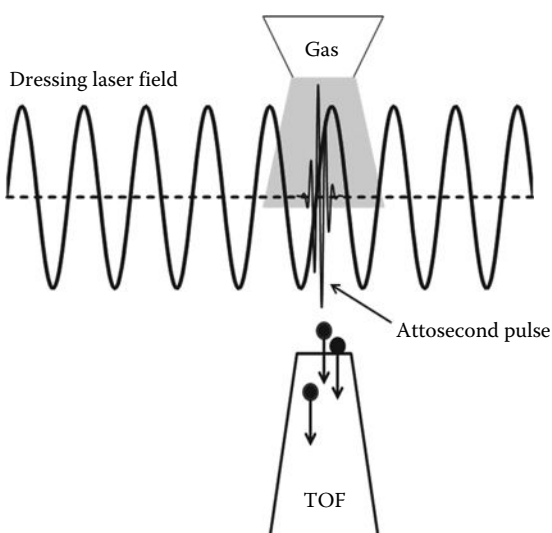


Figure 8.4 Experimental setup for characterizing isolated attosecond pulses.

atom, neon, is $I_p = 21.56$ eV. The ionization rate is proportional to the dipole transition matrix element $d(\vec{v})$. The NIR field changes the momentum of the electron. The spectrum of the photoelectrons produced by the XUV pulse in the presence of an NIR laser field is measured as a function of the time delay τ_d between the XUV and NIR pulses. It turns out that such a spectrogram contains the spectrum phase information of the XUV pulse.

In the high-intensity regime, the change of electron momentum can be considered as due to the electric force of the laser field, as described in Newton's theory of mechanics, which is the foundation of attosecond streaking.

As discussed in Chapter 5, over a rather broad range of dressing laser intensity, the amplitude of the freed electron wave with momentum $\vec{v}$ detected by an electron spectrometer at delay τ_d can be expressed in atomic units as

$$b(\vec{v}, \tau_d) = -i \int_{-\infty}^{\infty} \vec{\epsilon}_X(t - \tau_d) \cdot \vec{d}[\vec{v} + \vec{A}_L(t)] e^{i\Phi_G(t)} e^{-i\left(\frac{v^2}{2} + I_p\right)t} dt, \quad (8.7)$$

and

$$\Phi_G(t) = - \int_t^{\infty} \left[\vec{v} \cdot \vec{A}_L(t') + \frac{1}{2} A_L^2(t') \right] dt', \quad (8.8)$$

where $\vec{A}_L(t)$ is the vector potential of the dressing laser. The phase modulation $\Phi_G(t)$ to the electron wave by the dressing laser, as expressed by Equation 8.8, is the origin of electron spectrum oscillation.

The direction of $\vec{d}$ is the same as the electron momentum, thus $\vec{\epsilon}_X \cdot \vec{d} = \epsilon_X d \cos \theta_X$, where θ_X is the angle between XUV field and the direction of the electron. For simplicity, we assume $\theta_X = 0$, which means that we only detect the electrons emitted along the XUV field direction. If we assume that the dipole matrix element is a constant, then the measured spectrogram trace (i.e., the number of electrons) is

$$S(\vec{v}, \tau_d) = |b(\vec{v}, \tau_d)|^2 \propto \left| \int_{-\infty}^{\infty} \epsilon_X(t - \tau_d) e^{i\Phi_G(t)} e^{-i\left(\frac{v^2}{2} + I_p\right)t} dt \right|^2. \quad (8.9)$$

8.1.2.1 Dressing Laser

A portion of the laser that generates the attosecond pulse can be conveniently used as the dressing laser. For the Ti:Sapphire laser, the center wavelength is 800 nm. For simplicity, we assume that the NIR electric field is monochromatic and is linearly polarized, as depicted in Figure 8.4, i.e.,

$$\vec{\epsilon}_L(t) = \vec{E}_L \cos(\omega_1 t), \quad (8.10)$$

where

ω_1 is the laser frequency

$\vec{E}_L$ is the amplitude of the NIR field

The vector potential becomes

$$\vec{A}_L(t) = -\frac{\vec{E}_L}{\omega_1} \sin(\omega_1 t). \quad (8.11)$$

Consequently, the oscillating terms of the phase modulation are

$$\Phi_G(t) = \cos \theta_L \frac{\sqrt{8KU_p}}{\varepsilon_1} \cos(\omega_1 t) + \frac{U_p}{2\varepsilon_1} \sin(2\omega_1 t), \quad (8.12)$$

where θ_L is the angle between the direction of the electron after leaving the NIR field and the laser polarization direction. For simplicity, we assume $\theta_L = 0$, which means that only the electrons moving along the laser electric field direction are detected. The kinetic energy $K = (1/2)v^2$ and the ponderomotive energy $U_p = E_L^2/4\omega_1^2$. ε_1 is the photon energy of the dressing laser. For Ti:Sapphire laser, $\varepsilon_1 = \omega_1$ (in a.u.) = 1.55 eV.

8.1.2.2 ω_1 Component of Electron Spectrogram

An example of a simulated electron spectrogram assuming the polarization directions of the XUV and NIR photons pointing to the electron detector is shown in Figure 8.5a, which resembles an experimentally

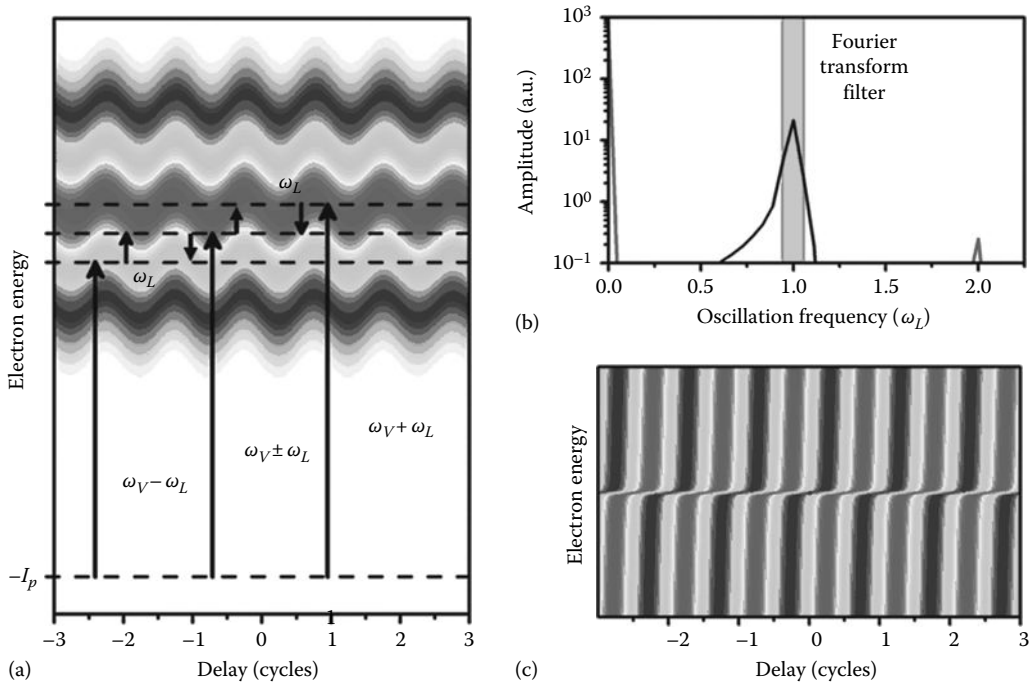


Figure 8.5 Interference inside an attosecond continuum. (a) Electron spectrogram and two-photon (XUV+NIR) transitions. (b) The amplitude of the one-dimensional Fourier transform of the spectrogram. (c) The normalized one-omega component. (Reprinted from M. Chini, S. Gilbertson, S.D. Khan, and Z. Chang, Characterizing ultrabroadband attosecond lasers, *Opt. Express*, 18, 13006, 2010. With permission of Optical Society of America.)

measured spectrogram. The XUV Gaussian pulse spectrum in the simulation supports 90 as the transform-limited duration, and the dressing laser is chosen to be 20 fs in duration, centered at 800 nm, and with a peak intensity of 10^{11} W/cm².

The vertical axis is electron energy $(1/2)v^2$. The horizontal axis is the delay between an isolated attosecond pulse and an intense NIR laser field. The size of the array is on the order of 1000×1000 . The number of grid points in the vertical direction is determined by the attosecond spectrum range divided by the energy resolution of the electron spectrometer. The number of points in the horizontal direction is approximately the NIR laser pulse width divided by the delay steps, which is a fraction of the laser cycle. In the simulations, the value at each grid is calculated by performing the integral 8.9 for the given attosecond pulse and NIR laser parameters.

The most obvious feature is the oscillation of the spectrum as a function of time delay. There are two different ways to understand the energy/momentum change as a function of delay. In the quantum picture, the electron in the continuum state absorbs or emits one or several laser photons, making a transition from one continuum state to another state. When a Fourier transform is performed on the spectrogram in Figure 8.5a, several peaks show in Figure 8.5b. The DC component is from the power spectrum. The peaks at ω_1 and $2\omega_1$ are related to transitions involving absorbing and emitting NIR photons. This photon transition point of view is more appropriate when the laser intensity is low.

When a narrow spectrum filter is applied to filter out the ω_1 component of the spectrogram, a new trace is obtained. It turns out that the attosecond spectrum phase can be extracted from the ω_1 component trace. To see the effects of the XUV spectral phase on the ω_1 component more clearly, one can normalize the value at each delay by the power spectrum of the XUV pulse, as shown in Figure 8.5c.

Simulated electron spectrograms for two different attosecond pulses are shown in Figure 8.6a and d. The power spectra of the two XUV pulses are identical, but the spectral phases are different. In both cases, examples of the filtered spectrograms are shown in Figure 8.6b and e. After spectrum normalization, the traces become Figure 8.6c and f. It is obvious that the normalized patterns are very different for the two pulses with different chirp values.

In Figure 8.6, the ω_1 component traces are obtained by performing the spectral filtering numerically. First, a complex array is obtained, $s_{\omega_1}(\vec{v}) = \int_{-\infty}^{\infty} |b(\vec{v}, \tau_d)|^2 e^{-i\omega_1 \tau_d} d\tau_d$. From its amplitude $|s_{\omega_1}(\vec{v})|$ and phase $\alpha(\vec{v}) = \text{Arg}(s_{\omega_1}(\vec{v}))$, we have a two-dimensional trace $S_{\omega_1}(\vec{v}, \tau_d) = |s_{\omega_1}(\vec{v})| \cos[\omega_1 \tau_d + \alpha(\vec{v})]$, which can be seen as traces in Figure 8.6b and e. In Figure 8.6, the power spectra of the XUV pulses are shown in the plots on the right of (a) and (d). The amplitudes of the ω_1 component are shown in the plots on the right of (b) and (e). Finally, the phases of the ω_1 component are shown in the plots on the right of (c) and (f).

When the dressing laser intensity is low, it is possible to perform the Fourier transform $\int_{-\infty}^{\infty} |b(\vec{v}, \tau_d)|^2 e^{-i\omega_1 \tau_d} d\tau_d$ analytically, which gives an expression that explicitly shows that the XUV spectrum phase can be extracted from the phase angle $\alpha(\vec{v}) = \text{Arg}(s_{\omega_1}(\vec{v}))$.

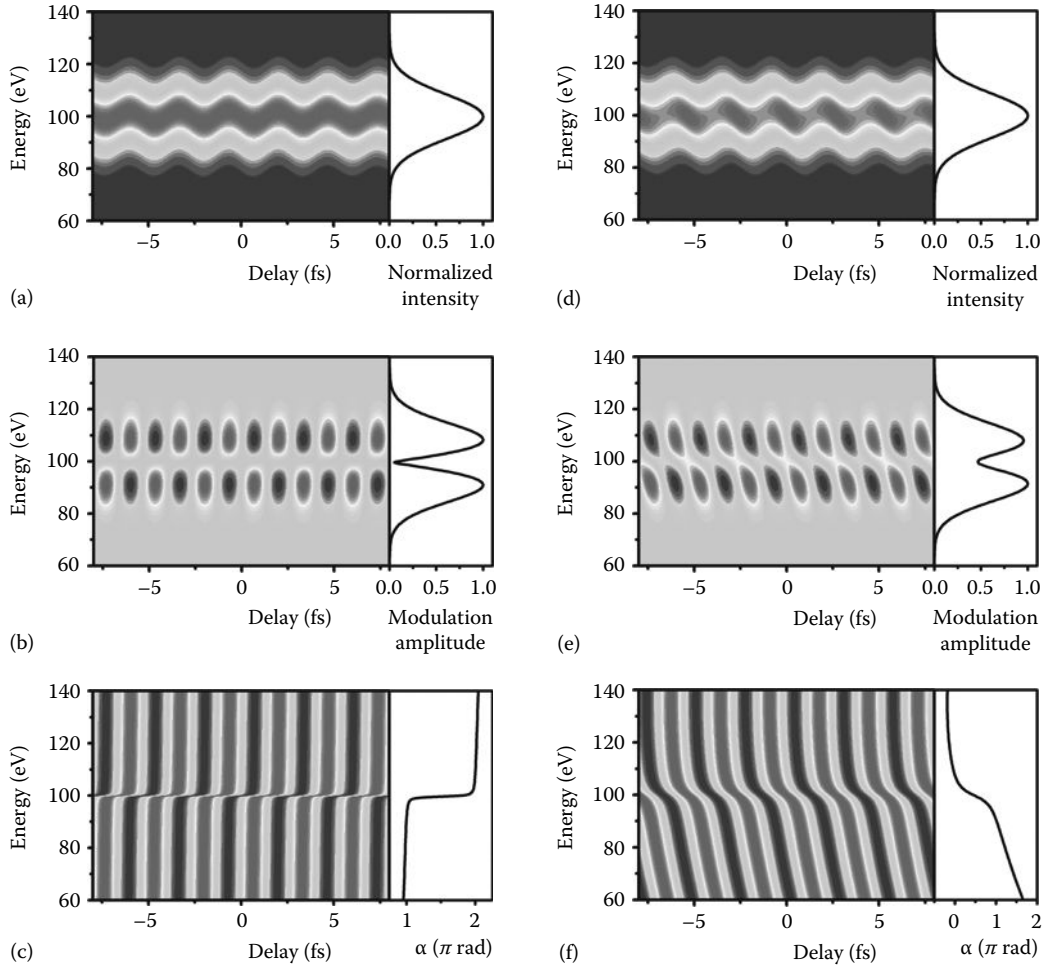


Figure 8.6 Phase encoding in electron spectrogram: (a) is for a nearly transform-limited 95 as pulse and (d) is for a strongly chirped 300 as pulse. (b, e) (left) Filtered ω_L oscillation and (right) extracted modulation amplitude. (c, f) (left) Filtered ω_L oscillation, normalized to the peak signal at each electron energy and (right) extracted $\alpha(v)$. (Reprinted from M. Chini, S. Gilbertson, S.D. Khan, and Z. Chang, Characterizing ultrabroadband attosecond lasers, *Opt. Express*, 18, 13006, 2010. With permission of Optical Society of America.)

8.1.2.3 Perturbative Regime

The laser intensity is proportional to U_p . When the laser intensity is low, $\eta \equiv \sqrt{8KU_p}/2\varepsilon_1 \ll 1$ and $U_p/2\varepsilon_1 \ll \sqrt{8KU_p}/\varepsilon_1$, the phase modulator for the electrons emitted in the direction of the laser polarization is

$$\begin{aligned}
 e^{i\Phi_G(t)} &\approx 1 + i\Phi_G(t) \approx 1 + i \frac{\sqrt{8KU_p}}{2\varepsilon_1} \cos(\omega_1 t) \\
 &= 1 + i\eta(e^{i\omega_1 t} + e^{-i\omega_1 t}).
 \end{aligned} \tag{8.13}$$

In the perturbative regime,

$$b(v, \tau_d) = \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} d\omega U(\omega) e^{i\varphi(\omega)} e^{i\omega(t-\tau_d)} \times [1 + i\eta(e^{i\omega_1 t} + e^{-i\omega_1 t})] e^{-i\left(\frac{v^2}{2} + I_p\right)t}. \quad (8.14)$$

Changing the order of the integration leads to

$$b(v, \tau_d) = \int_{-\infty}^{\infty} d\omega U(\omega) e^{i\varphi(\omega)} e^{-i\omega\tau_d} \times \int_{-\infty}^{\infty} dt [1 + i\eta(e^{i\omega_1 t} + e^{-i\omega_1 t})] e^{i\left[\omega - \left(\frac{v^2}{2} + I_p\right)\right]t}. \quad (8.15)$$

Using the property of the delta function

$$\int_{-\infty}^{\infty} dt e^{i\omega t} = \delta(\omega), \quad (8.16)$$

we have

$$b(v, \tau_d) = \int_{-\infty}^{\infty} d\omega U(\omega) e^{i\varphi(\omega)} e^{-i\omega\tau_d} \times \left\{ \delta\left[\omega - \left(\frac{v^2}{2} + I_p\right)\right] + i\eta\left\{\delta\left[\omega + \omega_1 - \left(\frac{v^2}{2} + I_p\right)\right] + \delta\left[\omega - \omega_1 - \left(\frac{v^2}{2} + I_p\right)\right]\right\} \right\}. \quad (8.17)$$

For the measured electron momentum v , we define an XUV frequency

$$\omega_v \equiv \frac{v^2}{2} + I_p, \quad (8.18)$$

which is the frequency of the XUV photon that produces the electron when the dressing laser is absent. Consequently, at low dressing laser intensity, $\leq 10^{12}$ W/cm², the electron spectrogram is related to

$$b(v, \tau_d) e^{i\omega_v \tau_d} = U(\omega_v) e^{i\varphi(\omega_v)} + i\eta \left[U(\omega_v + \omega_1) e^{i\varphi(\omega_v + \omega_1)} e^{-i\omega_1 \tau_d} + U(\omega_v - \omega_1) e^{i\varphi(\omega_v - \omega_1)} e^{+i\omega_1 \tau_d} \right]. \quad (8.19)$$

8.1.2.4 Flat Spectrum

For simplicity, we assume $U(\omega_v) = U(\omega_v + \omega_1) = U(\omega_v - \omega_1)$, which means that the XUV spectrum amplitude is flat across the whole range. In other words, the power spectrum has a square shape. For such spectra, the measured electron spectrogram is

$$\begin{aligned}
\frac{|b(v, \tau_d)|^2}{I^2(\omega_v)} &= |b(v, \tau_d)|^2 \approx 1 + \frac{2\sqrt{8KU_p}}{\varepsilon_1} \\
&\times \sin\left(\varphi(\omega_v) - \frac{\varphi(\omega_v - \omega_1) + \varphi(\omega_v + \omega_1)}{2}\right) \\
&\times \cos\left(\omega_1 \tau_d + \frac{\varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1)}{2}\right) \\
&+ \frac{4KU_p}{\varepsilon_1^2} [1 + \cos(2\omega_1 \tau_d - \varphi(\omega_v - \omega_1) + \varphi(\omega_v + \omega_1))].
\end{aligned} \tag{8.20}$$

The first term is just the power spectrum.

It is clear from the second and third terms that the electron signal at a given kinetic energy will oscillate with the delay τ_d . The second term, $\cos[\omega_1 \tau_d + (\varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1))/2]$, is due to the interference of transitions from the continuum states $\omega_v + \omega_1 - I_p$ and $\omega_v - \omega_1 - I_p$ to $\omega_v - I_p$, as shown in Figure 8.6a. The phase difference between two frequency components $\omega_v + \omega_1$ and $\omega_v - \omega_1$ can thus be easily obtained from the ω_1 Fourier component of the electron signal with kinetic energy $\omega_v - I_p$. This means that the single isolated attosecond pulse spectrum phase is encoded in the oscillation of the signal counts at a given kinetic energy as a function of delay between the XUV pulse and the NIR field. To decode the spectrum phase difference, one only needs to find the phase angle of a sinusoidal function $(\varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1))/2$.

8.1.2.5 Arbitrary Spectrum

In general, $U(\omega_v) \neq U(\omega_v + \omega_1) \neq U(\omega_v - \omega_1)$, the electron spectrogram

$$\begin{aligned}
|b(v, \tau_d)|^2 &= \left\{ U(\omega_v) e^{i\varphi(\omega_v)} + i\eta \left[U(\omega_v + \omega_1) e^{i\varphi(\omega_v + \omega_1)} e^{-i\omega_1 \tau_d} \right. \right. \\
&\quad \left. \left. + U(\omega_v - \omega_1) e^{i\varphi(\omega_v - \omega_1)} e^{+i\omega_1 \tau_d} \right] \right\} \\
&\times \left\{ U(\omega_v) e^{-i\varphi(\omega_v)} - i\eta \left[U(\omega_v + \omega_1) e^{-i\varphi(\omega_v + \omega_1)} e^{i\omega_1 \tau_d} \right. \right. \\
&\quad \left. \left. + U(\omega_v - \omega_1) e^{-i\varphi(\omega_v - \omega_1)} e^{-i\omega_1 \tau_d} \right] \right\}.
\end{aligned} \tag{8.21}$$

Keeping the DC and ω_1 components, we have

$$\begin{aligned}
|b(v, \tau_d)|^2 &\approx I(\omega_v) + i\eta \left[-U(\omega_v) U(\omega_v + \omega_1) e^{i[\varphi(\omega_v) - \varphi(\omega_v + \omega_1)]} e^{i\omega_1 \tau_d} \right. \\
&\quad \left. + U(\omega_v) U(\omega_v + \omega_1) e^{-i[\varphi(\omega_v) - \varphi(\omega_v + \omega_1)]} e^{-i\omega_1 \tau_d} \right. \\
&\quad \left. + U(\omega_v) U(\omega_v - \omega_1) e^{-i[\varphi(\omega_v) - \varphi(\omega_v - \omega_1)]} e^{i\omega_1 \tau_d} \right. \\
&\quad \left. - U(\omega_v) U(\omega_v - \omega_1) e^{i[\varphi(\omega_v) - \varphi(\omega_v - \omega_1)]} e^{-i\omega_1 \tau_d} \right].
\end{aligned} \tag{8.22}$$

The $-U(\omega_v) U(\omega_v + \omega_1) e^{i[\varphi(\omega_v) - \varphi(\omega_v + \omega_1)]} e^{i\omega_1 \tau_d}$ term can be considered as from the two-photon transition that involves one XUV photon ω_v plus one NIR photon ω_1 . The final state is at $\omega_v + \omega_1$. This transition reduces the signal at ω_v . Similarly, the $U(\omega_v) U(\omega_v + \omega_1) e^{i[\varphi(\omega_v + \omega_1) - \varphi(\omega_v)]} e^{i\omega_1 \tau_d}$ term represents the two-photon transition that involves one XUV photon $\omega_v + \omega_1$ minus one NIR photon ω_1 . The final state is at ω_v , which increases the signal at ω_v .

The other two terms can be explained in the same manner. The $-U(\omega_v)U(\omega_v - \omega_1)e^{i[\varphi(\omega_v) - \varphi(\omega_v - \omega_1)]}e^{i\omega_1\tau_d}$ term can be considered as the two-photon transition that involves one XUV photon ω_v minus one NIR photon ω_1 . The final state is at $\omega_v - \omega_1$. This transition reduces the signal at ω_v . The $U(\omega_v)U(\omega_v - \omega_1)e^{i[\varphi(\omega_v - \omega_1) - \varphi(\omega_v)]}e^{i\omega_1\tau_d}$ term represents the two-photon transition that involves one XUV photon $\omega_v - \omega_1$ plus one NIR photon ω_1 . The final state is at ω_v , which increases the signal at ω_v . In principle, we should be able to obtain the expression in Equation 8.22 by using the second-order perturbation theory.

Using Euler's equation $(e^{ix} - e^{-ix})/2i = \sin x$, Equation 8.22 can be rewritten as

$$|b(v, \tau_d)|^2 \approx I(\omega_v) \left\{ 1 + \frac{\sqrt{8KU_p}}{\varepsilon_1} \left[\sqrt{\frac{I(\omega_v + \omega_1)}{I(\omega_v)}} \times \sin(\omega_1\tau_d + \varphi(\omega_v) - \varphi(\omega_v + \omega_1)) - \sqrt{\frac{I(\omega_v - \omega_1)}{I(\omega_v)}} \sin(\omega_1\tau_d + \varphi(\omega_v - \omega_1) - \varphi(\omega_v)) \right] \right\}. \quad (8.23)$$

Let us define

$$\gamma(v) \sin[\alpha(v)] = \sqrt{\frac{I(\omega_v + \omega_1)}{I(\omega_v)}} \sin(\varphi(\omega_v) - \varphi(\omega_v + \omega_1)) - \sqrt{\frac{I(\omega_v - \omega_1)}{I(\omega_v)}} \sin(\varphi(\omega_v - \omega_1) - \varphi(\omega_v)), \quad (8.24)$$

$$\gamma(v) \cos[\alpha(v)] = \sqrt{\frac{I(\omega_v + \omega_1)}{I(\omega_v)}} \cos(\varphi(\omega_v) - \varphi(\omega_v + \omega_1)) - \sqrt{\frac{I(\omega_v - \omega_1)}{I(\omega_v)}} \cos(\varphi(\omega_v - \omega_1) - \varphi(\omega_v)). \quad (8.25)$$

Then

$$\begin{aligned} & \sqrt{\frac{I(\omega_v + \omega_1)}{I(\omega_v)}} \sin(\omega_1\tau_d + \varphi(\omega_v) - \varphi(\omega_v + \omega_1)) \\ & - \sqrt{\frac{I(\omega_v - \omega_1)}{I(\omega_v)}} \sin(\omega_1\tau_d + \varphi(\omega_v - \omega_1) - \varphi(\omega_v)) \\ & = \gamma(v) \cos[\alpha(v)] \sin(\omega_1\tau_d) + \gamma(v) \sin[\alpha(v)] \\ & \times \cos(\omega_1\tau_d) = \gamma(v) \sin(\omega_1\tau_d + \alpha(v)), \end{aligned} \quad (8.26)$$

which leads to

$$\frac{|b(v, \tau_d)|^2}{I(\omega_v)} - 1 \approx \frac{\sqrt{8KU_p}}{\varepsilon_1} \gamma(v) \sin[\omega_1\tau_d + \alpha(v)] = \frac{s_{\omega_1}(\bar{v}, \tau_d)}{I(\omega_v)}, \quad (8.27)$$

where

$$\begin{aligned} \gamma(v) &= \frac{I(\omega_v + \omega_1)}{I(\omega_v)} + \frac{I(\omega_v - \omega_1)}{I(\omega_v)} \\ & - 2\sqrt{\frac{I(\omega_v + \omega_1)I(\omega_v - \omega_1)}{I(\omega_v)}} \cos[\varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1)], \end{aligned} \quad (8.28)$$

$$\tan[\alpha(v)] = \frac{\sin[\varphi(\omega_v) - \varphi(\omega_v + \omega_1)] - \sqrt{\frac{I(\omega_v - \omega_1)}{I(\omega_v + \omega_1)}} \sin[\varphi(\omega_v - \omega_1) - \varphi(\omega_v)]}{\cos[\varphi(\omega_v) - \varphi(\omega_v + \omega_1)] - \sqrt{\frac{I(\omega_v - \omega_1)}{I(\omega_v + \omega_1)}} \cos[\varphi(\omega_v - \omega_1) - \varphi(\omega_v)]}. \quad (8.29)$$

Equation 8.27 is the analytical expression of the ω_1 component of the electron spectrogram normalized to the power spectrum. It is obtained by keeping the lowest-order term in Equation 8.13, which is equivalent to applying a very narrow spectrum filter at ω_1 . The spectrogram corresponding to the signal inside the filtered region is shown in Figure 8.5b.

8.1.2.6 Modulation Depth

The modulation depth of the oscillation is $(\sqrt{8KU_p}/\varepsilon_1)\gamma(v)$, where the parameter $\gamma(v)$ contains the attosecond chirp information. The $\gamma(v)$ curves for four different chirp values, $-(d^2\varphi/d\omega^2)|_{\omega_x} = \pm 5000 \text{ as}^2$ and $-(d^2\varphi/d\omega^2)|_{\omega_x} = \pm 500 \text{ as}^2$, are shown in Figure 8.7, assuming the power spectrum is the same as that shown in Figure 8.1.

Experimentally, the electron spectrum is measured with a certain energy resolution. In many cases, the measured spectrum at each delay is a data array, spaced by the energy width equal to the resolution. Here, the electron energy takes discrete values to show that γ is a one-dimensional array. For transform-limited pulses, $\varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1) = 0$, $\gamma(v) = \sqrt{I(\omega_v + \omega_1)/I(\omega_v)} - \sqrt{I(\omega_v - \omega_1)/I(\omega_v)}$. The product of the power spectrum and γ is plotted in Figure 8.8, which explains the signal plotted on the right of the filtered two-dimensional spectrogram

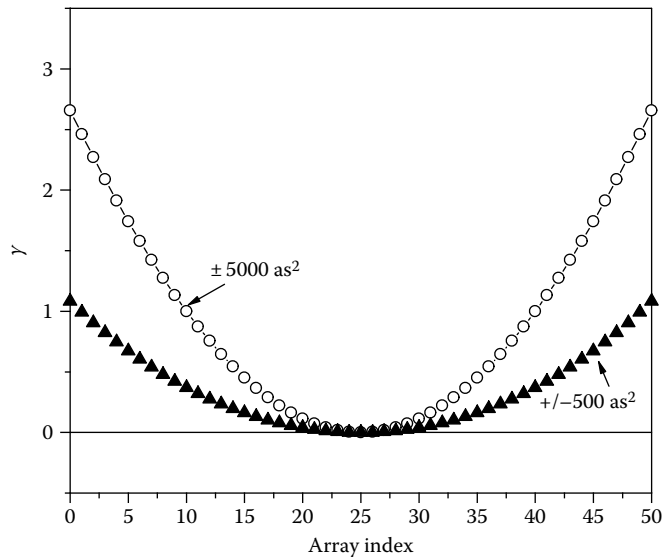


Figure 8.7 Modulation depth parameters corresponding to two chirp values.

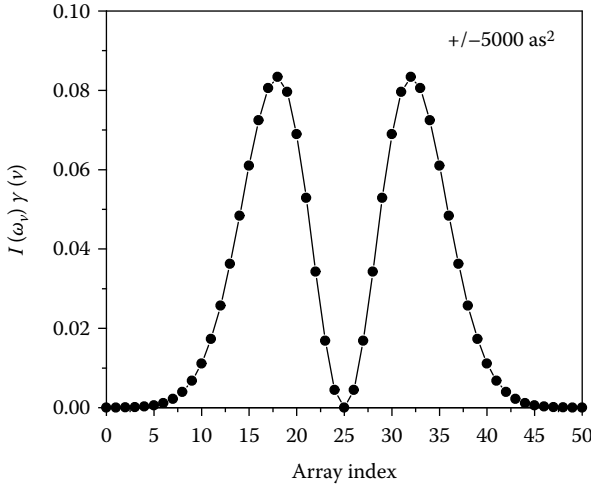


Figure 8.8 Product of the power spectrum and $\gamma(v)$.

in Figure 8.5b and e, i.e., $I(\omega_v)\gamma(v)$. It is worth pointing out that this quantity does not depend on the sign of the chirp.

It can be seen that the phase difference can be obtained from the measured $\gamma(v)$, which is related to the modulation depth of the $\omega_1\tau_d$ oscillation, i.e.,

$$\begin{aligned} & \varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1) \\ &= \arccos \left[\frac{I(\omega_v + \omega_1)}{I(\omega_v)} + \frac{I(\omega_v - \omega_1)}{I(\omega_v)} - \gamma(v) \right] \bigg/ \left[2 \frac{\sqrt{I(\omega_v + \omega_1)I(\omega_v - \omega_1)}}{I(\omega_v)} \right]. \end{aligned} \quad (8.30)$$

8.1.2.7 Phase Angle of the Filtered Spectrogram

In case the modulation depth cannot be accurately measured due to the poor signal-to-noise ratio, one can also obtain the spectral phase from the phase angle $\alpha(v)$ of the sinusoidal oscillation.

For characterizing attosecond pulses using the phase angle, a set of equations need to be solved one by one. The number of equations is the number of grid points in the electron energy direction of the two-dimensional spectrogram matrix, N . The phase angles at all grid points α_j , $j = 1, 2 \dots N$ are determined from the measured spectrogram.

The equation for an arbitrary inner point j is

$$\begin{aligned} & \tan[\alpha_j] \\ &= \frac{\sqrt{\frac{I(\omega_j + \omega_1)}{I(\omega_j)}} \sin[\varphi(\omega_j) - \varphi(\omega_j + \omega_1)] - \sqrt{\frac{I(\omega_j - \omega_1)}{I(\omega_j)}} \sin[\varphi(\omega_j - \omega_1) - \varphi(\omega_j)]}{\sqrt{\frac{I(\omega_j + \omega_1)}{I(\omega_j)}} \cos[\varphi(\omega_j) - \varphi(\omega_j + \omega_1)] - \sqrt{\frac{I(\omega_j - \omega_1)}{I(\omega_j)}} \cos[\varphi(\omega_j - \omega_1) - \varphi(\omega_j)]} \end{aligned} \quad (8.31)$$

or

$$\begin{aligned} & \sqrt{\frac{I(\omega_j + \omega_1)}{I(\omega_j)}} \sin[\varphi(\omega_j) - \varphi(\omega_j + \omega_1) - \alpha_j] \\ &= \sqrt{\frac{I(\omega_j - \omega_1)}{I(\omega_j)}} \sin[\varphi(\omega_j - \omega_1) - \varphi(\omega_j) - \alpha_j]. \end{aligned} \quad (8.32)$$

At the low endpoint $j = 1$, we assume the contribution to the interference from the outside of the matrix is zero, i.e.,

$$\sin[\varphi(\omega_{j=1} - \omega_1) - \varphi(\omega_{j=1}) - \alpha_{j=1}] = 0. \quad (8.33)$$

i.e., $\varphi(\omega_{j=1} - \omega_1) - \varphi(\omega_{j=1}) = \alpha_{j=1}$. Using this boundary condition, all the other phase differences at $j = 2, 3 \dots N - 1$ can be determined by

$$\begin{aligned} & \varphi(\omega_j) - \varphi(\omega_j + \omega_1) \\ &= \alpha_j + \text{asin} \left\{ \sqrt{\frac{I(\omega_j - \omega_1)}{I(\omega_j + \omega_1)}} \sin[\varphi(\omega_j - \omega_1) - \varphi(\omega_j) - \alpha_j] \right\}. \end{aligned} \quad (8.34)$$

Alternatively, one can also start at the high end $j = N$, by assuming $\sin[\varphi(\omega_{j=N}) - \varphi(\omega_{j=N+1}) - \alpha_{j=N}] = 0$. The phase difference values at $j = N - 1$ and other grid points are

$$\begin{aligned} & \varphi(\omega_j - \omega_1) - \varphi(\omega_j) \\ &= \alpha_j + \text{asin} \left\{ \sqrt{\frac{I(\omega_j + \omega_1)}{I(\omega_j - \omega_1)}} \sin[\varphi(\omega_j) - \varphi(\omega_j + \omega_1) - \alpha_j] \right\}. \end{aligned} \quad (8.35)$$

To retrieve the spectral phase of the pulses given in Figures 8.1 and 8.2, the spectral range extending ± 38 eV around the center energy is sampled with 1.55 eV steps, forming an array of size $N = 50$. The phase angle α is plotted as shown in Figure 8.9. The jump in the middle ($j = 25$) is due to the sign change slope of the power spectrum. To retrieve the phase using Equation 8.31, the phase angle needs to be wrapped into the

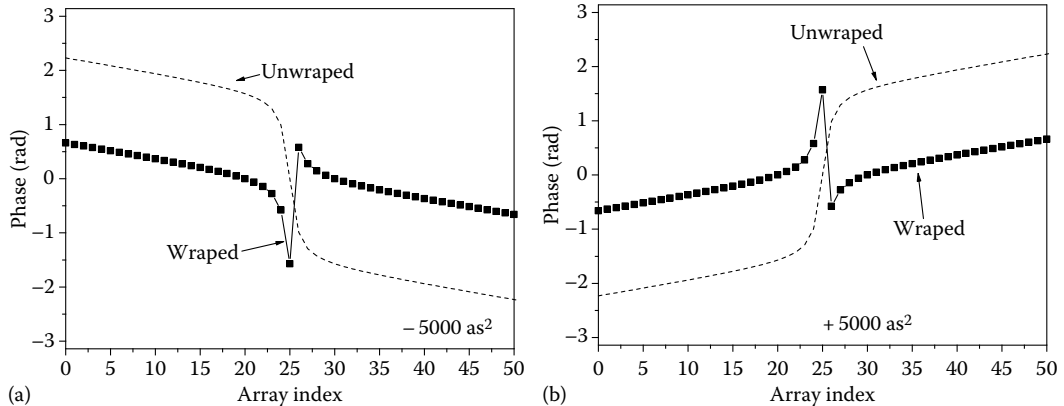


Figure 8.9 The spectral angles.

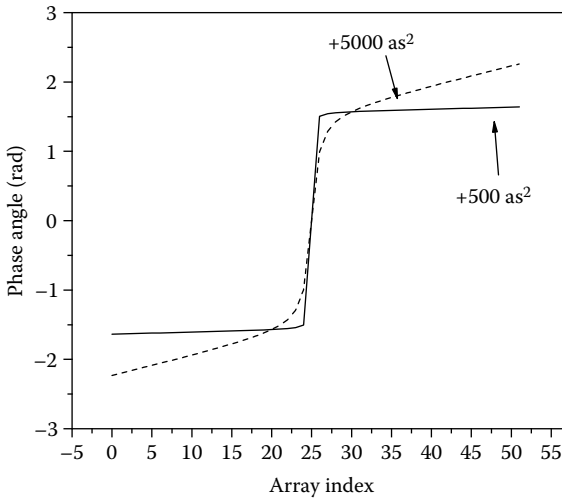


Figure 8.10 Unwrapped phase angles for two different chirp values.

$[-\pi/2, +\pi/2]$ range, the principal values of the sin function. The comparison of the wrapped and unwrapped phases is included in the figure. Figure 8.10 shows the effects of chirp on the unwrapped phase. One can qualitatively tell the degree of the chirp from the shape of the S curve. For transform-limited pulses, it becomes three straight-line segments.

The retrieved spectral phase differences are shown in Figure 8.11. Interestingly, the values of the phase angle α and the spectral phase difference become very close in the far wings of the spectrum. Thus, one can quantitatively find out the chirp by simply drawing a line that is tangential to the phase angle curves near the two ends.

For linearly chirped pulses, the spectral phase

$$\varphi(\omega_v) = \frac{1}{2} \frac{d^2 \varphi}{d\omega^2} \bigg|_{\omega_X} (\omega_v - \omega_X)^2. \quad (8.36)$$

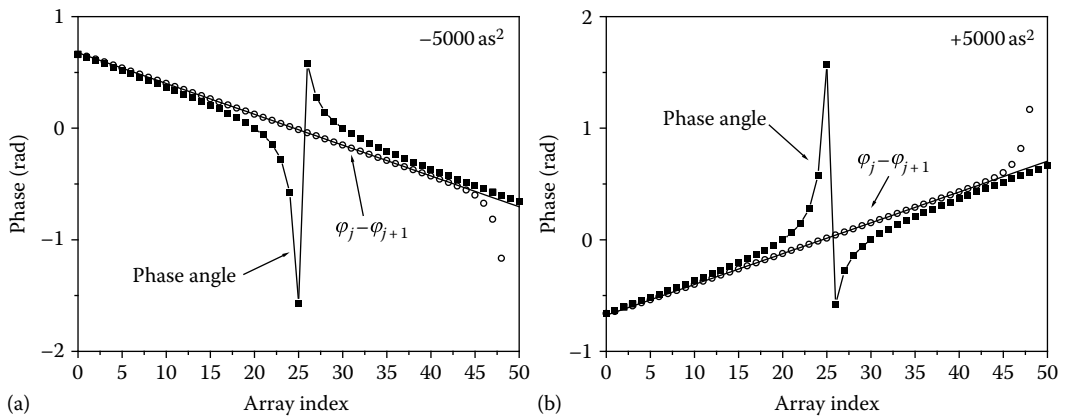


Figure 8.11 Spectral phase reconstruction. The solid line is the original phase and circles represent the retrieved phase.

The spectral phase difference

$$\begin{aligned} & \varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1) \\ &= \frac{1}{2} \frac{d^2 \varphi}{d\omega^2} \bigg|_{\omega_X} [(\omega_v - \omega_X - \omega_1)^2 - (\omega_v - \omega_X + \omega_1)^2], \end{aligned} \quad (8.37)$$

or

$$\varphi(\omega_v + \omega_1) - \varphi(\omega_v - \omega_1) = 2\omega_1 \frac{d^2 \varphi}{d\omega^2} \bigg|_{\omega_X} (\omega_v - \omega_X). \quad (8.38)$$

Thus the spectral phase difference is a linear function of ω_v , which can be seen in Figure 8.11.

When the signal-to-noise ratio of the experimental data is low, Equation 8.31 may not have an analytical solution. The most straightforward way to extract the phase $\varphi(\omega)$ is by minimizing the least square error function between the measured and guessed phase angles

$$\begin{aligned} R[\varphi(\omega_v)] &= \sqrt{\sum_{\omega_v} \left(\alpha(v) - \tan^{-1} \left[\frac{\sqrt{I(\omega_v + \omega_L)} \sin[\varphi(\omega_v) - \varphi(\omega_v + \omega_L)] - \sqrt{I(\omega_v - \omega_L)} \sin[\varphi(\omega_v - \omega_L) - \varphi(\omega_v)]}{\sqrt{I(\omega_v + \omega_L)} \cos[\varphi(\omega_v) - \varphi(\omega_v + \omega_L)] - \sqrt{I(\omega_v - \omega_L)} \cos[\varphi(\omega_v - \omega_L) - \varphi(\omega_v)]} \right] \right)^2}. \end{aligned} \quad (8.39)$$

Various analytical forms of the spectral phase can be assumed. The simplest minimization scheme is to ignore the high-order phases by assuming

$$\varphi(\omega_j) - \varphi(\omega_j + \omega_1) = a + b(\omega_j - \omega_X) + c(\omega_j - \omega_X)^2. \quad (8.40)$$

The coefficients a , b , c can be found out by minimizing the function $R(a, b, c)$. Furthermore, minimization algorithms such as the generic algorithm, which do not require an analytical form of the spectral phase, can be developed.

8.1.2.8 Comparison with Attosecond Streak Camera

The phase retrieval scheme based on the ω_1 component of the spectrogram has many advantages over the streak camera method based on attosecond streaking.

First, it requires much lower dressing laser intensity. The modulation depth of the $\omega_1 \tau_d$ oscillation is on the order of $\eta = \sqrt{8KU_p}/2\varepsilon_1$. As an example, for $K = 10$ eV, when the intensity of the dressing laser is 1×10^{11} W/cm², the modulation depth $\sqrt{8KU_p}/2\varepsilon_1 \approx 23\%$. Experimentally, such a large modulation is sufficient for determining the phase angle $\alpha(v)$, because it only requires a signal-to-noise ratio of ~ 10 , or electron counts on the order of 100 per pixel of the two-dimensional spectrogram.

In the attosecond streaking picture, the attosecond phase is encoded in the broadening of the photoelectron spectrum at the delay point where the vector potential crosses zero. The maximum amount of electron kinetic energy change due to the dressing laser is $\sqrt{8KU_p}$. Under the same condition as the previous example, $\sqrt{8KU_p} \approx \varepsilon_1 \approx 1.55$ eV, which means the streaking amplitude is on the order of the laser photon energy in

the two-dimensional spectrogram. This is not sufficient for characterizing attosecond pulses by streaking. In other words, a phase encoding scheme that relies on attosecond streaking needs much higher dressing laser intensity than one based on electron count oscillation caused by the interference of two-photon transitions.

High NIR laser intensity is needed for a streak camera so that the amount of broadening of the electron spectrum width is comparable to the bandwidth of the attosecond pulse to be measured, which requires intensity greater than 10^{14} W/cm² to characterize a 70 as pulse centered at 100 eV. For characterizing 25 as pulses centered at 100 eV, the required laser intensity would produce high-energy photoelectrons through multiphoton and field ionization of the target atoms, which would overlap with the attosecond photoelectron spectrum and destroy much of the information encoded in the streaked spectrogram.

Second, this attosecond phase encoding scheme is not susceptible to dressing laser intensity variations in the laser focal volume as well as the shot-to-shot laser intensity fluctuation, which is obvious, as the phase angle of the oscillation does not depend on dressing laser intensity.

8.1.3 Modulation Depth for Gaussian Pulses

The modulation depth of $|b(\nu, \tau_d)|^2$ is

$$\frac{\sqrt{8KU_p}}{\varepsilon_1} \gamma = \frac{\sqrt{8(\hbar\omega_v - I_p)U_p}}{\varepsilon_1} \times \left\{ \frac{I(\omega_v + \omega_1)}{I(\omega_v)} + \frac{I(\omega_v - \omega_1)}{I(\omega_v)} - 2 \frac{\sqrt{I(\omega_v + \omega_1)I(\omega_v - \omega_1)}}{I(\omega_v)} \right. \\ \left. \times \cos \left[2\omega_1 \frac{d^2\varphi}{d\omega^2} \Big|_{\omega_X} (\omega_v - \omega_X) \right] \right\}. \quad (8.41)$$

For Gaussian pulses with a linear chirp

$$I(\omega_v) = I_0 e^{-4 \ln 2 \frac{(\omega_v - \omega_X)^2}{\Delta\omega^2}}, \quad (8.42)$$

we have

$$\gamma(\omega_v - \omega_X) e^{4 \ln 2 \frac{\omega^2}{\Delta\omega^2}} = e^{4 \ln 2 \frac{2(\omega_v - \omega_X)\omega_1}{\Delta\omega^2}} + e^{-4 \ln 2 \frac{2(\omega_v - \omega_X)\omega_1}{\Delta\omega^2}} \\ - 2 \cos \left[2 \frac{d^2\varphi}{d\omega^2} \Big|_{\omega_X} \omega_1 (\omega_v - \omega_X) \right], \quad (8.43)$$

or

$$\frac{1}{2} \gamma(\omega_v - \omega_X) e^{4 \ln 2 \frac{\omega^2}{\Delta\omega^2}} = \cosh \left[2\omega_1 (\omega_v - \omega_X) \frac{4 \ln 2}{\Delta\omega^2} \right] \\ - \cos \left[2\omega_1 (\omega_v - \omega_X) \frac{d^2\varphi}{d\omega^2} \Big|_{\omega_X} \right], \quad (8.44)$$

which is an even function and equals zero at the center frequency, i.e., $\omega_v = \omega_X$.

$$\text{For } 2\omega_1(\omega_v - \omega_X) \frac{4 \ln 2}{\Delta\omega^2} \ll 1 \text{ and } 2\omega_1(\omega_v - \omega_X) \frac{d^2\varphi}{d\omega^2} \Big|_{\omega_X} \ll 1,$$

$$\frac{1}{2} \gamma(\omega_v - \omega_X) e^{4 \ln 2 \frac{\omega_v^2}{\Delta\omega^2}} = 2 \left[\left(\frac{4 \ln 2}{\Delta\omega^2} \right)^2 + \left(\frac{d^2\varphi}{d\omega^2} \Big|_{\omega_X} \right)^2 \right] \omega_1^2 (\omega_v - \omega_X)^2. \quad (8.45)$$

When the XUV pulse is linearly chirped, the modulation depth varies with the XUV frequency almost quadratically, which can be seen in [Figure 8.7](#). For the transform-limited XUV pulse, $(d^2\varphi/d\omega^2)|_{\omega_X} = 0$, the modulation is due to the power spectrum difference at $\omega_v + \omega_1$ and $\omega_v - \omega_1$. The contribution from chirp becomes significant when $(d^2\varphi/d\omega^2)|_{\omega_X}$ is comparable to $4 \ln 2 / \Delta\omega^2 = \tau_0^2 / 4 \ln 2 \approx 0.36 \tau_0^2$, where τ_0 is the transform-limited pulse duration corresponding to the bandwidth.

Sometimes, photon energy ε is preferred instead of frequency. For that purpose, Equation 8.44 is expressed as

$$\frac{1}{2} \gamma(\varepsilon_v - \varepsilon_X) e^{4 \ln 2 \frac{\omega_v^2}{\Delta\omega^2}} = \cosh \left[2\varepsilon_1(\varepsilon_v - \varepsilon_X) \frac{4 \ln 2}{\Delta\varepsilon^2} \right] - \cos \left[2\varepsilon_1(\varepsilon_v - \varepsilon_X) \frac{d^2\varphi}{d\omega^2} \Big|_{\omega_X} \times 2.3 \times 10^{-6} \right], \quad (8.46)$$

where the unit of ε is eV and the unit of $(d^2\varphi/d\omega^2)|_{\omega_X}$ is as^2 .

8.1.3.1 High-Order Effects

When the intensity of the dressing laser is further increased, high-order terms need to be added to describe the measured spectrogram. For higher dressing laser intensity

$$e^{i\Phi_G(t)} \approx 1 + i\Phi_1(t) - \frac{\Phi_1^2(t)}{2}, \quad (8.47)$$

where $\Phi_1(t) = (\sqrt{8KU_p}/\varepsilon_1) \cos(\omega_1 t)$. Here we keep $\Phi_1^2(t)/2 = (8KU_p/2\varepsilon_1^2) \cos^2(\omega_1 t)$ but neglect the $U_p/2\varepsilon_1 \sin(2\omega_1 t)$ in Equation 8.12 because $K/\varepsilon_1 \gg 1$.

The electron wave amplitude becomes

$$b(v, \tau_d) = \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} U(\omega) e^{i\varphi(\omega)} e^{i\omega(t-\tau_d)} d\omega$$

$$\times \left[1 + i\eta(e^{i\omega_1 t} + e^{-i\omega_1 t}) - \frac{\eta^2}{2}(2 + e^{i2\omega_1 t} + e^{-i2\omega_1 t}) \right] e^{-i(\frac{v^2}{2} + I_p)t}. \quad (8.48)$$

or

$$\begin{aligned}
 b(v, \tau_d) = & \int_{-\infty}^{\infty} d\omega U(\omega) e^{i\varphi(\omega)} e^{-i\omega\tau_d} \left\{ \delta \left[\omega - \left(\frac{v^2}{2} + I_p \right) \right] \right. \\
 & + i\eta \left\{ \delta \left[\omega + \omega_1 - \left(\frac{v^2}{2} + I_p \right) \right] + \delta \left[\omega - \omega_1 - \left(\frac{v^2}{2} + I_p \right) \right] \right\} \\
 & - \frac{\eta^2}{2} \left\{ 2\delta \left[\omega - \left(\frac{v^2}{2} + I_p \right) \right] + \delta \left[\omega + 2\omega_1 - \left(\frac{v^2}{2} + I_p \right) \right] \right. \\
 & \left. \left. + \delta \left[\omega - 2\omega_1 - \left(\frac{v^2}{2} + I_p \right) \right] \right\} \right\}, \quad (8.49)
 \end{aligned}$$

which lead to

$$\begin{aligned}
 b(v, \tau_d) e^{i\omega_v \tau_d} = & (1 - \eta^2) U(\omega_v) e^{i\varphi(\omega_v)} + i\eta [U(\omega_v + \omega_1) e^{i\varphi(\omega_v + \omega_1)} e^{-i\omega_1 \tau_d} \\
 & + U(\omega_v - \omega_1) e^{i\varphi(\omega_v - \omega_1)} e^{+i\omega_1 \tau_d}] \\
 & - \frac{\eta^2}{2} [U(\omega_v + 2\omega_1) e^{i\varphi(\omega_v + 2\omega_1)} e^{-i2\omega_1 \tau_d} \\
 & + U(\omega_v - 2\omega_1) e^{i\varphi(\omega_v - 2\omega_1)} e^{+i2\omega_1 \tau_d}]. \quad (8.50)
 \end{aligned}$$

For extremely short attosecond pulses, the power spectrum changes slowly with the frequency, we can assume $U(\omega_v) = U(\omega_v + \omega_1) = U(\omega_v - \omega_1) = U(\omega_v + 2\omega_1) = U(\omega_v - 2\omega_1)$.

Consequently

$$\begin{aligned}
 b(v, \tau_d) = & U(\omega_v) e^{-i\omega_v \tau_d} \left\{ [1 - \eta^2] e^{i\varphi(\omega_v)} + i\eta [e^{i\varphi(\omega_v + \omega_1)} e^{-i\omega_1 \tau_d} \right. \\
 & + e^{i\varphi(\omega_v - \omega_1)} e^{+i\omega_1 \tau_d}] - \frac{\eta^2}{2} [e^{i\varphi(\omega_v + 2\omega_1)} e^{-i2\omega_1 \tau_d} \\
 & \left. + e^{i\varphi(\omega_v - 2\omega_1)} e^{+i2\omega_1 \tau_d}] \right\}. \quad (8.51)
 \end{aligned}$$

The measured electron signal

$$\begin{aligned}
 |b(v, \tau_d)|^2 \approx & I(\omega_v) \left\{ 1 + \frac{2\sqrt{8KU_p}}{\varepsilon_1} \sin \left(\varphi(\omega_v) - \frac{\varphi(\omega_v - \omega_1) + \varphi(\omega_v + \omega_1)}{2} \right) \right. \\
 & \times \cos \left(\omega_1 \tau_d + \frac{\varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1)}{2} \right) \\
 & + \frac{4KU_p}{\varepsilon_1^2} \left[1 + \cos(2\omega_1 \tau_d + \varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1)) \right. \\
 & - \cos \left(\varphi(\omega_v) - \frac{\varphi(\omega_v - 2\omega_1) + \varphi(\omega_v + 2\omega_1)}{2} \right) \\
 & \left. \left. \times \cos \left(2\omega_1 \tau_d + \frac{\varphi(\omega_v - 2\omega_1) - \varphi(\omega_v + 2\omega_1)}{2} \right) \right] \right\}. \quad (8.52)
 \end{aligned}$$

The ω_1 component is determined by the phase difference of the two frequency components $\varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1)$. However, the $2\omega_1$

oscillation is affected by both $\varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1)$ and $\varphi(\omega_v - 2\omega_1) - \varphi(\omega_v + 2\omega_1)$. Thus, it is easier to retrieve the spectral phase from the former.

For an arbitrary spectrum,

$$\begin{aligned} \frac{s_{\omega_1}(\vec{v}, \tau_d)}{I(\omega_v)} &= \frac{\sqrt{8KU_p}}{\varepsilon_1} \gamma(v) \sin[\omega_1 \tau_d + \alpha(v)] + \left(\frac{\sqrt{8KU_p}}{\varepsilon_1} \right)^2 \\ &\times \left[\frac{\sqrt{I(\omega_v + \omega_1)I(\omega_v + 2\omega_1)}}{I(\omega_v)} \right. \\ &\times \sin[\omega_1 \tau_d + (\varphi(\omega_v + \omega_1) - \varphi(\omega_v + 2\omega_1))] \\ &- \frac{\sqrt{I(\omega_v - \omega_1)I(\omega_v - 2\omega_1)}}{I(\omega_v)} \\ &\left. \times \sin[\omega_1 \tau_d - (\varphi(\omega_v - \omega_1) - \varphi(\omega_v - 2\omega_1))] \right]. \quad (8.53) \end{aligned}$$

In this case, the ω_1 component is affected by the phases $\varphi(\omega_v - 2\omega_1)$, $\varphi(\omega_v + 2\omega_1)$ and the amplitudes $U(\omega_v + \omega_1)$, $U(\omega_v - \omega_1)$. To reduce the error caused by these extra terms, we should keep $(\sqrt{8KU_p}/\varepsilon_1)^2 \ll 1$ or $(\sqrt{8KU_p}/\varepsilon_1) < 1$. These transitions involve three NIR photons, i.e., $\omega_v \pm 2\omega_1$ and $\omega_v \pm \omega_1$. The interferences at ω_v are from $(\omega_v + 2\omega_1) - 2\omega_1$ and $(\omega_v + \omega_1) - \omega_1$, as well as from $(\omega_v - 2\omega_1) + 2\omega_1$ and $(\omega_v - \omega_1) + \omega_1$. Transitions that affect the signal at ω_v also include $\omega_v + 2\omega_1$ and $\omega_v + \omega_1$, as well as $\omega_v - 2\omega_1$ and $\omega_v - \omega_1$. The transition rates from these transitions are smaller than the transitions that involve one NIR photon at low intensity. As the dressing laser intensity increases, transitions that involve more and more photons contribute to the ω_1 component. Thus, the error of using Equation 8.28 increases with dressing laser intensity.

At the dressing laser intensity 2×10^{11} W/cm², centered at 0.8 μ m, the ponderomotive energy $U_p = 9.33 \times 10^{-14} \times 1 \times 10^{12} \times 0.8^2 = 0.06$ eV. For $K = 100$ eV, $(\sqrt{8KU_p}/\varepsilon_1) = 1$. This intensity is too high for the approximation to be valid. However, numerical simulations reveal that the error is acceptable even at 10^{12} W/cm².

8.1.4 Effect of Dipole Transition Element

For low laser intensity, $\vec{\mathcal{A}}_L(t) < \vec{v}$, the dipole transition element

$$d[\vec{v} + \vec{\mathcal{A}}_L(t)] \approx d(\vec{v}). \quad (8.54)$$

Including this approximate expression in the electron wave in the continuum

$$\begin{aligned} b(v, \tau_d) &= \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} d\omega U(\omega) d(\vec{v}) e^{i\varphi(\omega)} e^{i\omega(t - \tau_d)} \\ &\times [1 + i\eta(e^{i\omega_1 t} + e^{-i\omega_1 t})] e^{-i\left(\frac{v^2}{2} + I_p\right)t}, \quad (8.55) \end{aligned}$$

we get

$$b(v, \tau_d) = e^{-i\omega_v \tau_d} \left\{ U(\omega_v) d(\vec{v}) e^{i\varphi(\omega_v)} + i\eta [U(\omega_v + \omega_1) d(\vec{v}) e^{i\varphi(\omega_v + \omega_1)} e^{-i\omega_1 \tau_d} + U(\omega_v - \omega_1) d(\vec{v}) e^{i\varphi(\omega_v - \omega_1)} e^{+i\omega_1 \tau_d}] \right\}. \quad (8.56)$$

For target atoms with a real dipole transition element,

$$U(\omega_v) d(\vec{v}) = U_E \left(\frac{v^2}{2} \right) = \sqrt{I_E \left(\frac{v^2}{2} \right)}, \quad (8.57)$$

where

$U_E(v^2/2)$ is the amplitude of the electron wave with kinetic energy $v^2/2$
 $I_E(v^2/2)$ is the NIR laser field free photoelectron signal

Strictly, the transition interference is affected by the phase of both the attosecond wave and phase of the dipole transition element. This effect can be explored to measure the phase of the dipole transition element by comparing the results from two detection targets.

The measured electron spectrum

$$b(v, \tau_d) = e^{-i\omega_v \tau_d} \left\{ U_E \left(\frac{v^2}{2} \right) e^{i\varphi(\omega_v)} + i\eta \left[U_E \left(\frac{v^2}{2} + \omega_1 \right) e^{i\varphi(\omega_v + \omega_1)} e^{-i\omega_1 \tau_d} + U_E \left(\frac{v^2}{2} - \omega_1 \right) e^{i\varphi(\omega_v - \omega_1)} e^{+i\omega_1 \tau_d} \right] \right\} \quad (8.58)$$

or

$$\frac{|b(v, \tau_d)|^2}{I_E(v)} \approx 1 + \frac{\sqrt{8KU_p}}{\varepsilon_1} [\gamma(v) \sin(\omega_1 \tau_d + \alpha(v))], \quad (8.59)$$

where

$$\gamma(v) = \frac{I_E \left(\frac{v^2}{2} + \omega_1 \right)}{I_E \left(\frac{v^2}{2} \right)} + \frac{I_E \left(\frac{v^2}{2} - \omega_1 \right)}{I_E \left(\frac{v^2}{2} \right)} - 2 \frac{\sqrt{I_E \left(\frac{v^2}{2} + \omega_1 \right) I_E \left(\frac{v^2}{2} - \omega_1 \right)}}{I_E \left(\frac{v^2}{2} \right)} \times \cos[\varphi(\omega_v - \omega_1) - \varphi(\omega_v + \omega_1)], \quad (8.60)$$

$\tan[\alpha(v)]$

$$= \frac{\sqrt{I_E \left(\frac{v^2}{2} + \omega_1 \right)} \sin[\varphi(\omega_v) - \varphi(\omega_v + \omega_1)] - \sqrt{I_E \left(\frac{v^2}{2} - \omega_1 \right)} \sin[\varphi(\omega_v - \omega_1) - \varphi(\omega_v)]}{\sqrt{I_E \left(\frac{v^2}{2} + \omega_1 \right)} \cos[\varphi(\omega_v) - \varphi(\omega_v + \omega_1)] - \sqrt{I_E \left(\frac{v^2}{2} - \omega_1 \right)} \cos[\varphi(\omega_v - \omega_1) - \varphi(\omega_v)]}. \quad (8.61)$$

Thus, both the amplitude modulation and the phase angle can be expressed by the photoelectron spectrum signal measured when only XUV light is present.

8.2 Complete Reconstruction of Attosecond Bursts for Isolated Attosecond Pulses

Characterization of isolated attosecond XUV pulses by complete reconstruction of attosecond bursts (CRAB) trace is a two-dimensional phase retrieval problem, which is similar to the frequency-resolved optical gating (FROG) measurement of femtosecond laser pulses. In principle, the temporal envelope and phase of an attosecond pulse can be retrieved from the photoelectron spectrogram using various phase retrieval algorithms. In practice, however, most of them are too slow. The one chosen by Mairesse and Quéré for CRAB is the principal component generalized projection algorithm (PCGPA), which can be executed on personal computers in a reasonable amount of time.

For the generalized projection algorithms to work, momentum and time cannot be inseparable terms inside the integrand

$$b(\vec{v}, \tau_d) = -i \int_{-\infty}^{\infty} \varepsilon_X(t - \tau_d) d[\vec{v} + \vec{A}_L(t)] e^{i\Phi_G(t)} e^{-i\left(\frac{v^2}{2} + I_p\right)t} dt, \quad (8.62)$$

and

$$\Phi_G(\vec{v}, t) = - \int_t^{\infty} \left[\vec{v} \cdot \vec{A}_L(t') + \frac{1}{2} A_L^2(t') \right] dt'. \quad (8.63)$$

Obviously, the terms $d[\vec{v} + \vec{A}_L(t)]$ and $\Phi_G(\vec{v}, t)$ contain both momentum and time.

The time dependence of the dipole transition matrix element can be removed by replacing it with $d[\vec{v} + \vec{A}_L(\tau_d)]$, which was derived in Chapter 5. As a result, the modified spectrogram fed into the reconstruction algorithm is given by the following expression:

$$S(v, \tau_d) = \frac{|b(v, \tau_d)|^2}{d^2[\vec{v} + \vec{A}_L(\tau_d)]}. \quad (8.64)$$

8.2.1 Central Momentum Approximation

The momentum dependence of $\Phi_G(\vec{v}, t)$ can be removed by making the substitution $\vec{v} \rightarrow \vec{v}_0$, where $\vec{v}_0$ is the central momentum of the unstreaked electrons. This is called central momentum approximation, which is valid when the width of the XUV spectrum is much smaller than the center energy of the photoelectron. At a given center energy, this approximation puts a limit on the shortest XUV pulse that can be characterized. Let us assume that the maximum bandwidth $\Delta\varepsilon_X$ is equal to 50% of the center energy $K_0 = (1/2)v_0^2$. Then, the shortest pulse that can be characterized by CRAB is

$$\tau_{X0}(\text{as}) = \frac{1825}{\Delta\varepsilon_X(\text{eV})} = \frac{3650}{K_0(\text{eV})}. \quad (8.65)$$

Under the central momentum approximation, the phase modulation becomes

$$\Phi_G(\vec{v}, t) = \Phi_G(\vec{v}_0, t) = - \int_t^\infty \left[\vec{v}_0 \cdot \vec{\mathcal{A}}_L(t') + \frac{1}{2} \mathcal{A}_L^2(t') \right] dt'. \quad (8.66)$$

Finally, we have

$$S(\vec{v}, \tau_d) = \left| \int_{-\infty}^{\infty} \varepsilon_X(t - \tau_d) e^{i\Phi_G(\vec{v}_0, t)} e^{-i\left(\frac{v^2}{2} + I_p\right)t} dt \right|^2. \quad (8.67)$$

This expression has the same form as the FROG trace for conventional femtosecond laser pulse characterization. The phase modulation, $\Phi_G(\vec{v}_0, t)$, acts as a temporal phase gate in the FROG language.

The difference between the exact phase of the modulator and the approximate one can be expressed as

$$\Delta\Phi_G(v, t) = (v - v_0) \int_t^\infty \mathcal{A}_L(t') dt'. \quad (8.68)$$

For the central momentum approximation to be valid, $\Delta\Phi_G(v, t)$ must vary less than π during the interaction with the XUV field. This condition can be written as

$$\epsilon(v) = \max_t |\Delta\Phi_G(v, t)| \leq \pi. \quad (8.69)$$

For a single isolated attosecond XUV pulse with a duration τ_x , the NIR vector potential is approximately linear over the duration of the pulse, so that the error can be estimated as

$$\epsilon(v) = \tau_x |(v - v_0) \mathcal{A}_L(t_0)|, \quad (8.70)$$

where the XUV pulse is assumed to be centered at a moment zero-crossing point t_0 . For a maximum value of the vector potential, $\mathcal{A}_{L, \max} = 0.25$ a.u. (7.2×10^{12} W/cm²), Equation 8.70 yields an error of $\epsilon(v) \leq 0.4$ rad in the energy window between 80 and 130 eV.

8.2.1.1 Effects of Experimental Conditions

We will discuss the effects of two factors on the pulse retrieval using PCGPA: the shot noise and the NIR laser intensity. We wish to determine the minimum electron counts in the spectrogram and the lowest laser intensity acceptable for the CRAB reconstruction to guide experiments. The first one is related to the signal accumulation time. The second one determines the level of background noise from laser field ionization of the target atoms.*

Except where otherwise noted, the NIR pulse is assumed to be 5 fs in duration and centered at 800 nm in the simulation and in the discussion to follow. The XUV spectrum is assumed to support a 90 as transform-limited pulse with a 5000 as² linear chirp. The observation angle is set at zero. The grid size for the CRAB trace is 512×512 pixels. The delay range is chosen to be 10.67 fs and the spectral range is 200 eV.

* A more compressive analysis of the various effects on CRAB can be found in Wang, H., Chini, M., Khan, S.D., Chen, S., Gilbertson, S., Feng, X., Mashiko, H., and Chang, Z., Practical issues of retrieving isolated attosecond pulses, *J. Phys. B At. Mol. Opt. Phys.* 42, 134007 (2009).

8.2.1.2 Shot Noise

Due to the low XUV photoelectron signal level, the shot noise in the experimental CRAB traces obtained in a reasonable amount of time (less than 1 h) using 1 kHz lasers could be substantial. The situation will improve with the 10 kHz lasers that are becoming available.

The low electron counts are caused by several factors. The number of attosecond XUV photons generated in noble gases is on the order of 10^7 – 10^8 per laser shot (the pulse energy is typically 0.1–1 nJ). This is comparable to the output of a femtosecond oscillator, but much lower than the amplified femtosecond pulses (mJ or higher).

Another limiting factor is the probability that an XUV photon is absorbed at the detection gas jet to generate a photoelectron. The atomic gases used for photoelectron production have photoabsorption cross sections on the order of 10^{-17} cm² at XUV wavelengths. The absorption probability is also a function of the pressure–length product. The length of the gas target must be much smaller than the Rayleigh range of the NIR dressing laser focal spot to avoid significant carrier-envelope (CE) phase variation. The gas pressure is limited by the microchannel plate detector, which requires a vacuum better than 10^{-5} torr to avoid damage. Differential pumping is commonly implemented to allow a high target pressure. Even if the local gas density at the laser focus is maximized by using small gas jets, the gas density is limited by electron scattering, as discussed in Chapter 7.

In a typical attosecond streak camera, the overall probability of detecting a photoelectron for an XUV photon striking the target is $\sim 10^{-7}$. The number of detected photoelectrons is ~ 1 per laser shot. For such a small number of photoelectrons, shot noise can be significant unless the spectra are accumulated for a very long time. It is thus important to determine how many photoelectron counts are necessary for an accurate reconstruction of the duration and the contrast of attosecond pulses with CRAB.

8.2.1.3 Array Dimension of CRAB Trace

We start with a simulated noise-free CRAB trace with 512×512 pixels, which is shown in Figure 8.12a. The signal intensity changes smoothly from pixel to pixel. The spectrogram shown is re-sampled to 80 delay steps with a step size of $\Delta\tau_d = 130$ as, which is comparable to the achievable experimental delay resolution. The required step size should decrease with NIR intensity as more and more high-frequency peaks show up in Figure 8.6b. To include the signal peaked at $n\omega_1$, the delay step size must be smaller than $\pi/n\omega_1$, to satisfy the Sampling theorem.

The dimension in the electron energy direction is kept at 512 samples. The resolution is $\Delta K = 0.4$ eV, which is similar to the energy resolution of typical experiments. In PCGPA, the delay step size $\Delta\tau_d$ and the energy step size ΔK must satisfy the constraint $\Delta\tau_d \Delta K = 2\pi/N_E$, where N_E , a power of two, is the total number of samples in the chosen energy range. Thus, the CRAB trace is interpolated from the 80 to 512 pixels in the delay direction in order to satisfy this condition.

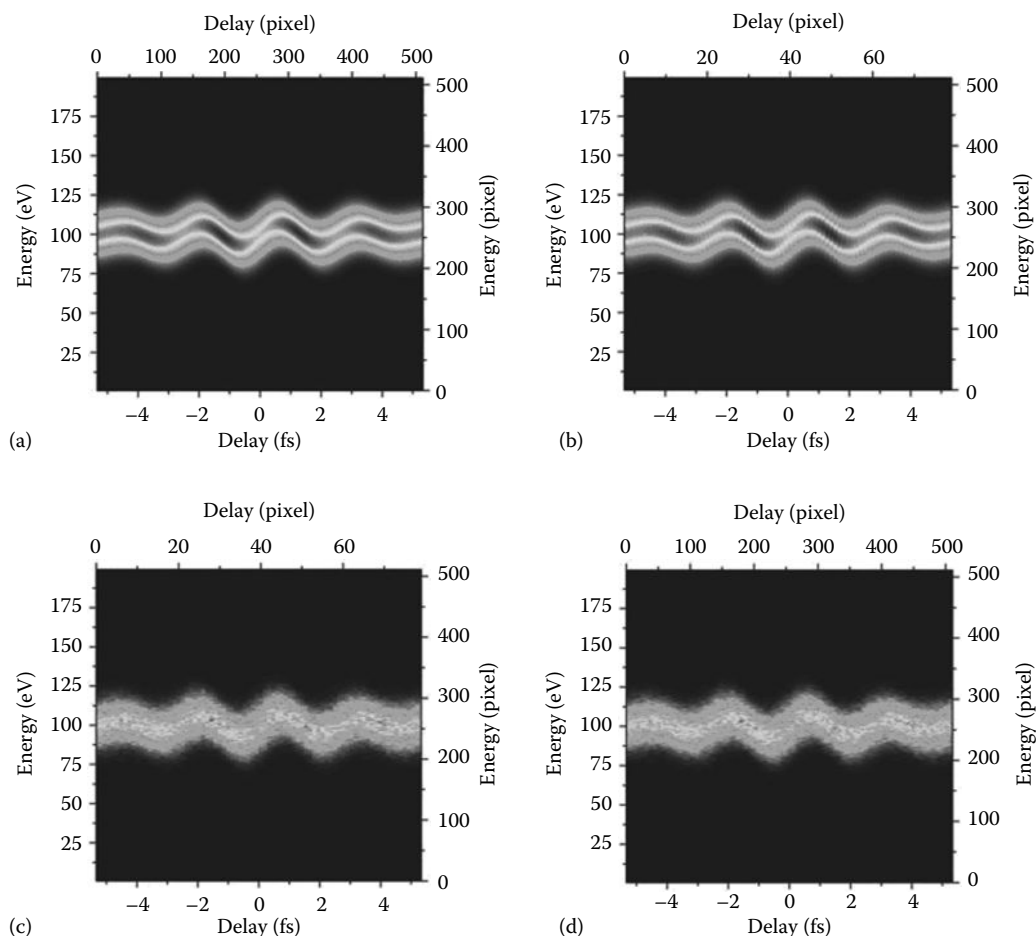


Figure 8.12 Preparation of simulated CRAB traces. All plots are normalized. The XUV spectrum supported a 90 as transform-limited pulse duration and had a 5000 as² linear chirp. The NIR streaking field had peak intensity of 1×10^{12} W/cm². (a) Noise-free trace on a square grid size of 512² pixels. (b) The trace was first re-sampled to 80 delay steps of ~130 as with integer values of counts at each pixel. Here, the maximum pixel count was set to 50. (c) After re-sampling, shot noise was added as integer values taken from a Poisson distribution. (d) The trace was finally interpolated (using a bicubic spline method) back to a square grid of 512² pixels, as required for PCGPA. (From H. Wang, M. Chini, S.D. Khan, S. Chen, S. Gilbertson, X. Feng, H. Mashiko, and Z. Chang, Practical issues of retrieving isolated attosecond pulses, *J. Phys. B: At. Mol. Opt. Phys.*, 42, 134007, 2009.)

8.2.2 Simulation of Shot Noise in CRAB Traces

The simulations are conducted at five signal levels, with a maximum count at the pixel where the count is the highest of 200, 100, 50, 25, and 10 counts, which approximately corresponds to 10,000, 5,000, 2,500, 1,250, and 500 total photoelectron counts per delay step. The number of counts on each grid point is quantized to integer values. As an example, the trace with a maximum count of 50 is shown in Figure 8.12b.

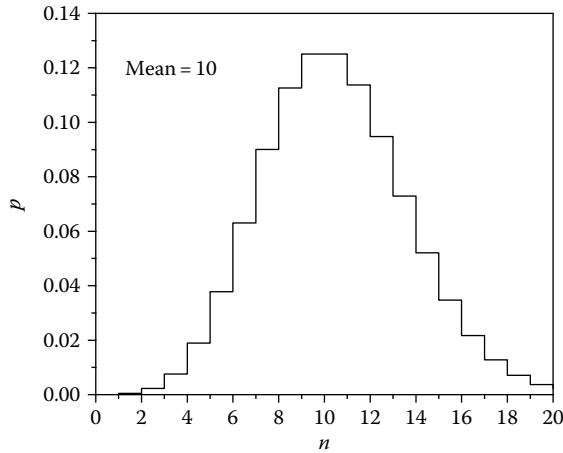


Figure 8.13 Poisson probability distribution for $\bar{n} = 10$.

The shot noise follows a Poisson distribution. Suppose the mean value of the electron count at a pixel is $\bar{n}$, then the probability of detecting n electrons is

$$p(n, \bar{n}) = \frac{\bar{n}^n}{n!} e^{-\bar{n}}. \quad (8.71)$$

The probability distribution for $\bar{n} = 10$ is illustrated in Figure 8.13. The electron counts in all pixels are calculated using a Monte Carlo method, which results in the trace shown in Figure 8.12c, which corresponds to Figure 8.12b. Finally, interpolation is performed on the trace to return it to a grid size of 512×512 pixels, suitable for reconstruction with PCGPA, as shown in Figure 8.12d.

8.2.3 Effects of Shot Noise on XUV Pulse Retrieval

The retrieved attosecond XUV pulse intensity and phase for the cases of 200 and 10 counts are shown in Figure 8.14a. The 200 count results are close to the actual values. Also, as is shown in Figure 8.14b, the noise in the wings of the pulse is 10^{-6} of the main pulse, which is lower than experiments can measure.

To determine the minimum counts and lowest laser intensities required for faithful reconstruction, this process is performed for several dressing laser intensities. The FWHM pulse duration and the linear chirp for our retrievals for two XUV pulses with different bandwidths but the same chirp (5000 as^2) are plotted in Figure 8.15. For CRAB traces with at least 50 counts for the brightest pixel, the retrieved pulse duration and linear chirp are within 5% of their actual values when the streaking intensity is greater than W/cm^2 for XUV pulses with a spectrum supporting 90 as transform-limited durations, as shown in Figure 8.15a. When the XUV spectrum supports 180 as transform-limited pulses, the retrieved pulse duration is within 3% and the linear chirp is within 5% for traces with 50 counts or more, as shown in Figure 8.15a. Thus, 50 counts is the minimum required value for the chosen XUV and laser pulse parameters.

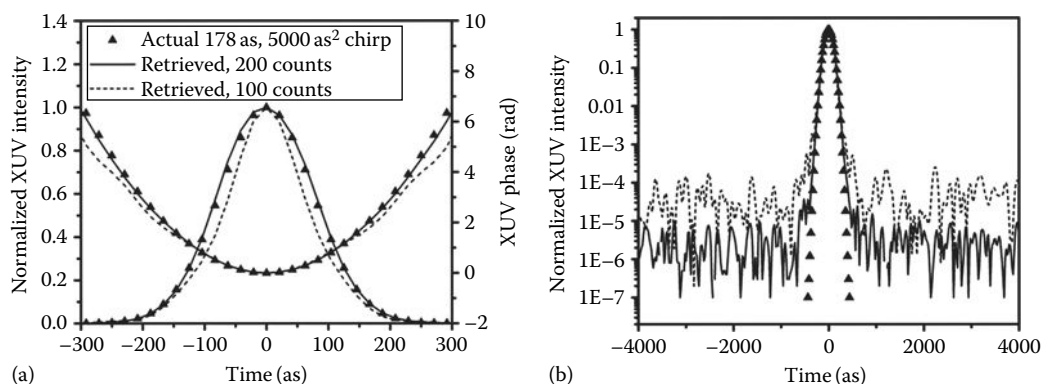


Figure 8.14 (a) Normalized XUV intensity and phase. (b) Normalized XUV intensity on log scale. The streaking intensity was $1 \times 10^{12} \text{ W/cm}^2$. The triangles show the noise-free pulse, whereas the solid line and dashed line show the retrieved pulses with 200 and 10 counts at the peak of the energy spectrum, respectively. (From H. Wang, M. Chini, S.D. Khan, S. Chen, S. Gilbertson, X. Feng, H. Mashiko, and Z. Chang, Practical issues of retrieving isolated attosecond pulses, *J. Phys. B: At. Mol. Opt. Phys.*, 42, 134007, 2009.)

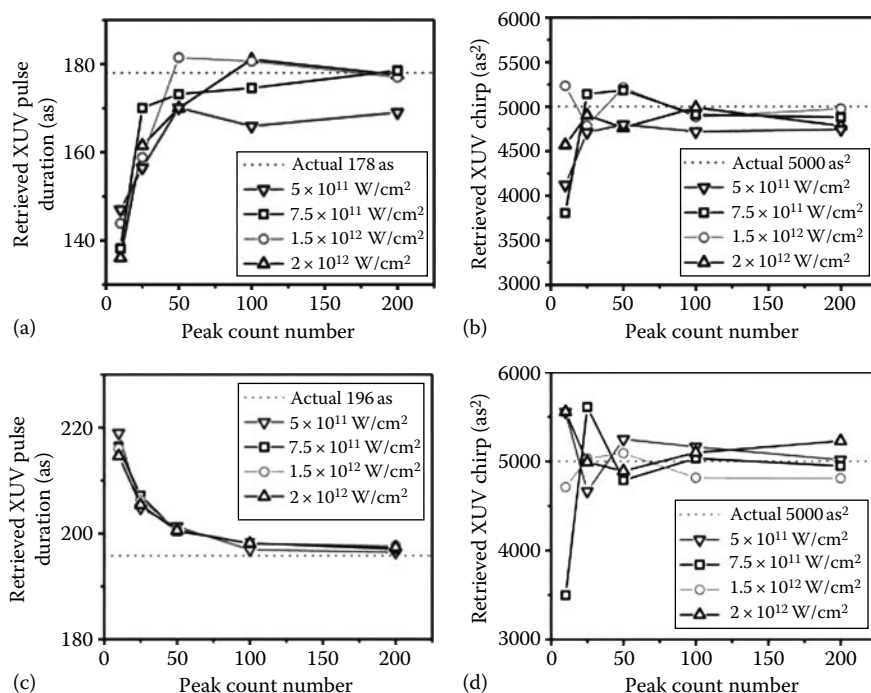


Figure 8.15 (a) Retrieved XUV pulse duration and (b) retrieved linear chirp for pulses with spectrum supporting 90 as transform-limited pulses. For peak count numbers above 50, the pulse duration and linear chirp are retrieved within 5% of their actual values for streaking intensities greater than $5 \times 10^{11} \text{ W/cm}^2$, indicating convergence of the algorithm. (c) Retrieved XUV pulse duration and (d) retrieved linear chirp for pulses with spectrum supporting 180 as transform-limited pulses. For peak count numbers above 50, the pulse duration is retrieved within 3% and the linear chirp within 5% of their actual values. (From H. Wang, M. Chini, S.D. Khan, S. Chen, S. Gilbertson, X. Feng, H. Mashiko, and Z. Chang, Practical issues of retrieving isolated attosecond pulses, *J. Phys. B: At. Mol. Opt. Phys.*, 42, 134007, 2009.)

8.2.4 Dressing Laser Intensity

There are two different ways to understand how the temporal structure information is encoded in the CRAB trace and how it is extracted. We can consider the CRAB trace as a group of streak camera traces with different streaking speeds obtained at different delays between the XUV pulse and the NIR streaking field. The temporal information of the XUV attosecond pulse is extracted by comparing the distorted electron spectrum at the maximum streaking speed with the undistorted one at the minimum streaking speed. The temporal resolution is determined by the maximum streaking speed.

We can also think of the CRAB trace as an assembly of interference traces taken at difference delays. The spectrum of isolated attosecond pulses is an XUV continuum. When the laser dressed spectrum is measured as a function of the delay between the XUV pulse and the NIR field, the spectral phase difference between any two frequency components separated by one NIR photon energy affects the interference and the spectrogram, as we discussed in PROOF.

Numerical simulations on the effects of dressing laser intensity on the isolated attosecond pulse reconstruction are done to find out which of these two interpretations is better. We first work out the intensity requirement from the streak camera perspective.

8.2.4.1 NIR Intensity and Streaking Speed

In a streak camera, the attosecond pulse duration is obtained by the photoelectron spectrum broadening in the NIR field. For this to work, the amount of broadening should be comparable to the spectrum width of the pulse. The energy width of an attosecond pulse with time duration τ_X is

$$\Delta\omega_X = \frac{1}{\tau_X}. \quad (8.72)$$

Suppose the streaking speed is dK/dt , then the energy width of the streaked trace is $(dK/dt)\tau_X$. To unambiguously determine the pulse duration, we set

$$\frac{dK}{dt}\tau_X = \Delta\omega_X. \quad (8.73)$$

Thus, the required streaking speed

$$\frac{dK}{dt} = \frac{1}{\tau_X^2}. \quad (8.74)$$

The streaking speed is related to the maximum energy shift ΔK_{max} at a given laser intensity,

$$\frac{dK}{dt} = \frac{d}{dt}\left(\frac{v^2}{2}\right) = v\frac{d}{dt}[\Delta v_{max} \sin(\omega_1 t)] = \omega_1 v \Delta v_{max} = \omega_1 \Delta K_{max}. \quad (8.75)$$

Thus, the required energy shift

$$\Delta K_{max} = \frac{1}{\omega_1 \tau_X^2}. \quad (8.76)$$

Since

$$\Delta K_{\max} = \sqrt{8K_0 U_p}, \quad (8.77)$$

the ponderomotive energy corresponds to the required NIR intensity to satisfy

$$U_p(I_L)\omega_1^2 = \frac{1}{8K_0\tau_X^4}. \quad (8.78)$$

Using this criterion for an NIR laser centred at 800 nm, 28 eV of $\Delta K_{\max}$ is required to resolve a 90 as XUV pulse, which corresponds to the minimum NIR laser peak intensity of $5.5 \times 10^{13} \text{ W/cm}^2$, if the center energy of the electron is at 35 eV.

Such high intensity of the NIR field can produce free electrons through multiphoton ionization and above-threshold ionization (ATI). Since the cutoff energy of rescattered electrons from ATI can reach $10U_p$ ($\sim 33 \text{ eV}$), the ATI electrons overlap with the streaked XUV photoelectrons and add significant background noise to the CRAB trace.

8.2.4.2 Dependence of Minimum NIR Intensity on XUV Chirp

To examine the minimum streaking intensity requirement, CRAB reconstruction for an XUV pulse with spectrum supporting a 90 as transform-limited pulse is performed over a large chirp range at three different NIR intensities. The peak electron count is set at 100. As shown in Figure 8.16a and b, even with the linear chirp of $20,000 \text{ as}^2$, for which the XUV pulse duration was broadened to one quarter of the NIR cycle, both the pulse duration and the linear chirp of the XUV pulse can be retrieved with high accuracy at the intensity $7.5 \times 10^{11} \text{ W/cm}^2$.

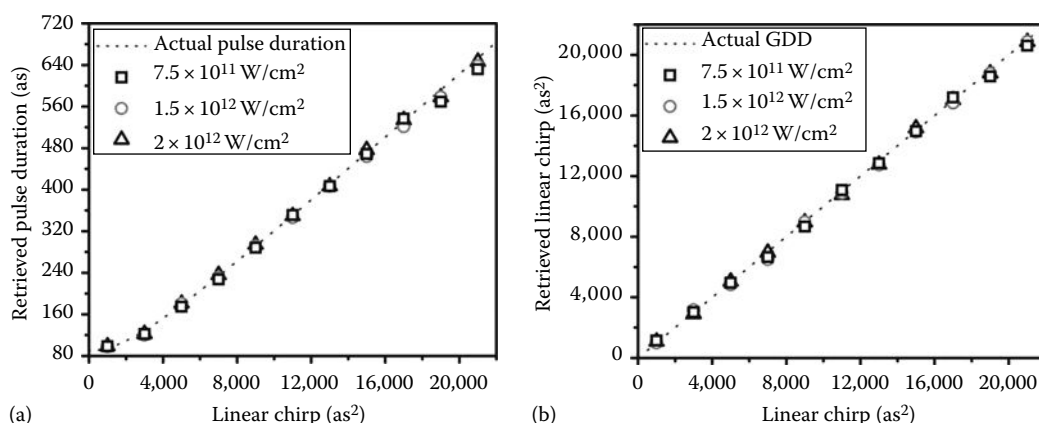


Figure 8.16 (a) Retrieved XUV pulse duration as a function of input linear chirp for different streaking intensities. (b) Retrieved linear chirp as a function of input linear chirp for different streaking intensities. (From H. Wang, M. Chini, S.D. Khan, S. Chen, S. Gilbertson, X. Feng, H. Mashiko, and Z. Chang, Practical issues of retrieving isolated attosecond pulses, *J. Phys. B: At. Mol. Opt. Phys.*, 42, 134007, 2009.)

Thus, the CRAB method requires much lower streaking intensity than the classical streaking camera method to measure an isolated XUV pulse. This conclusion removes the NIR intensity constraint of measuring attosecond pulses, which is important for characterization of even shorter attosecond pulses. The required low dressing laser intensity is consistent with the interference interpretation of the CRAB trace.

8.2.4.3 Comparison between PROOF and CRAB

Both PROOF and CRAB start with the same photoelectron spectrogram. However, PROOF only uses the ω_1 component. In PROOF, the central momentum approximation is not needed. The approximation poses a limitation on the shortest attosecond pulses that can be characterized at a given center photon energy. Thus, PROOF can be applied to pulses whose bandwidths are much larger than the center photon energies, which is important for measuring pulses close to 1 as.

8.3 Amplitude Gating

The strong-field approximation predicts that a pair of XUV attosecond pulses are generated in one laser cycle when atoms are driven by a linearly polarized laser field containing many cycles. This is the case when the cutoff region of the XUV spectrum is examined. The number of attosecond pulses per laser cycle doubles in the plateau region of the spectrum, because there are two energy-degenerate electron trajectories per one half of a laser cycle. Consequently, up to four XUV pulses can be produced per laser cycle. However, the phase-matching process suppresses the long trajectory. As a result, only one attosecond pulse per half a laser cycle can reach the detector.

At the present time, the shortest intense laser pulse is ~ 4 fs and centered at 750 nm, which is approximately two laser periods long. When it excites atoms, approximately three to four pulses with attosecond duration are produced in the plateau region of the XUV spectrum. The interference of the attosecond pulses produces discrete and well-resolved harmonic peaks. Single attosecond pulses have been produced at the cutoff region, i.e., in the laser cycle near the peak of the few-cycle laser pulses. This approach is known as amplitude gating.

The harmonic spectrum is simulated with the strong-field approximation discussed in Chapter 5. Helium atoms are excited by two-cycle linearly polarized laser pulses to demonstrate the amplitude gating. In this case, the electric field of the pulse

$$\varepsilon(t) = E_0 e^{-2 \ln 2 \left(\frac{t}{\tau_p}\right)^2} \cos\left(\frac{2\pi}{\lambda_0} t + \varphi_{CE}\right). \quad (8.79)$$

We choose the center wavelength $\lambda_0 = 1.6 \mu\text{m}$ for generating attosecond pulses in the keV range. The pulse duration $\tau_p = 10$ fs, which corresponds to approximately two laser cycles at the chosen wavelength. The electric fields for two CE phase values, $\varphi_{CE} = 0$ and $\pi/2$, are shown in [Figure 8.17](#). The power spectrum is shown in [Figure 8.18](#).

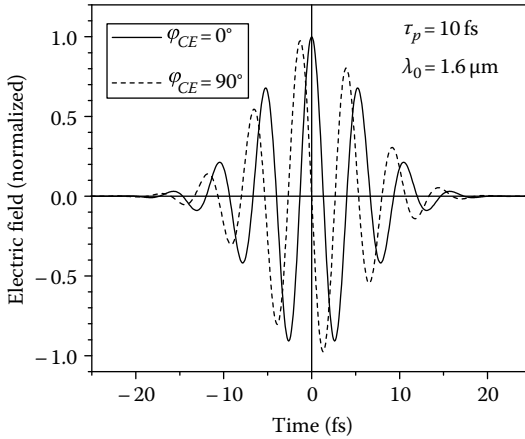


Figure 8.17 Two-cycle laser field centered at 1.6 μm with different CE phases.

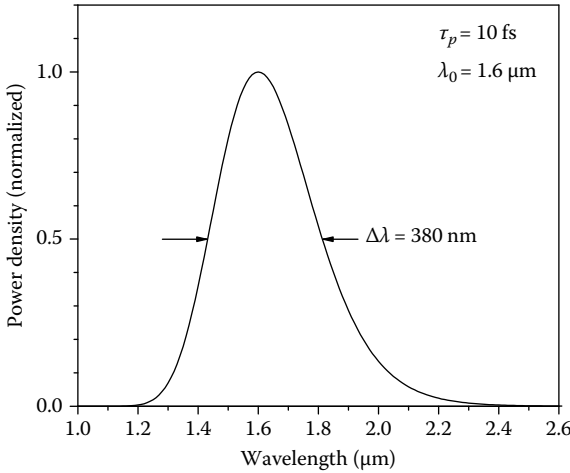


Figure 8.18 The power spectrum of the pulse in Figure 8.17.

At 4.1×10^{15} W/cm², the spectra for the two CE phases are shown in Figures 8.19 and 8.20. There are two to three distinct staircase regions, with different signal strengths. For such a strong laser field, the ground state population varies significantly from one laser half-cycle to the next. The region just above 1 keV is more intense, because the ground state population is high in the half laser cycle where the soft x-ray is generated.

The attosecond pulses in the different spectrum region for $\varphi_{CE} = 0$ are shown in Figure 8.21, which was obtained by applying a square window with the width $\Delta E = 62$ eV to the spectrum, and then performing the Fourier transform. The number of attosecond pulses reduces as the center photon energy of the window increases. At 1007.5 eV central photon energy, there are approximately five pulses. The first two, generated in the -5 to -2.5 fs time range, are from the short and long trajectories

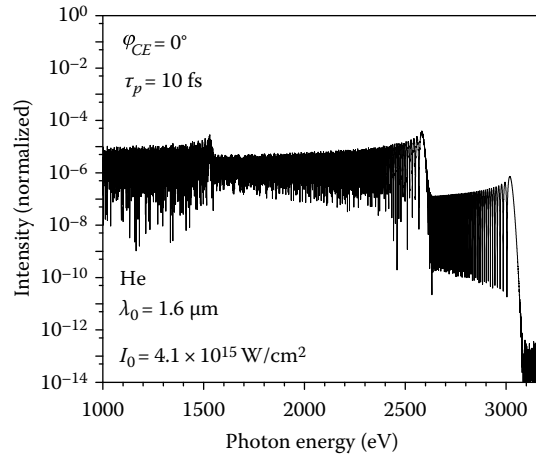


Figure 8.19 Soft x-ray spectrum generated with two-cycle laser fields centered at 1.6 μm . The CE phase $\varphi_{CE} = 0^\circ$.

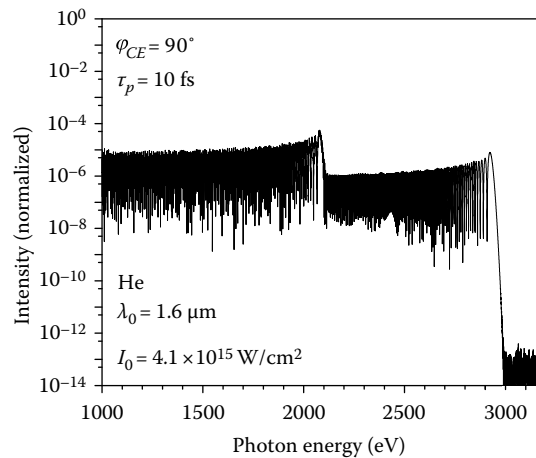


Figure 8.20 Soft x-ray spectrum generated with two-cycle laser fields centered at 1.6 μm . The CE phase $\varphi_{CE} = 90^\circ$.

generated in a low-intensity half laser cycle. The next two pulses, in the -2.5 to 0 fs range, are generated in the next half-cycle. The spectrum in the cutoff region would become a continuum when the propagation effects are taken into account. The propagation suppresses the contribution from the long trajectories. The output pulse from a gas target near the cutoff region is an isolated one from the short trajectory. The shorter the driving laser, the broader the cutoff region. Ideally, one would use a half-cycle laser that generates the broadest continuous spectrum after propagation to support the shortest attosecond pulse.

At center energy $\varepsilon_0 = 387.5$ eV, the pulse emitted at -2.5 fs is much stronger than the attosecond pulse emitted at -5 fs. The width of the pulse is 60 as for the given spectrum window, as illustrated in Figure 8.22. At $\varepsilon_0 = 1937.5$ eV, the duration of the pulse from the short trajectory emitted

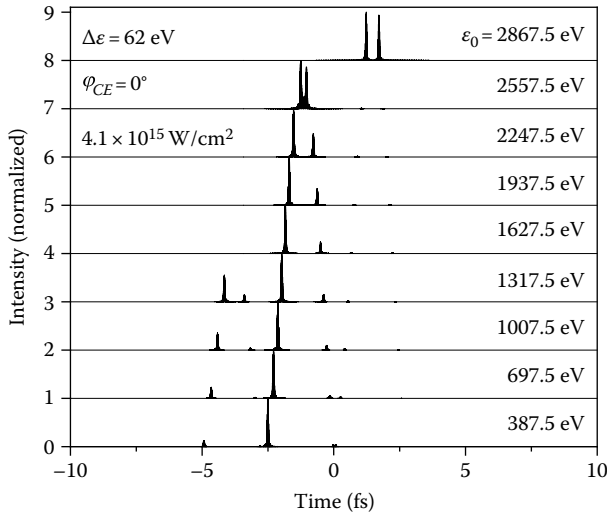


Figure 8.21 The number of attosecond pulses generated at different central photon energies.

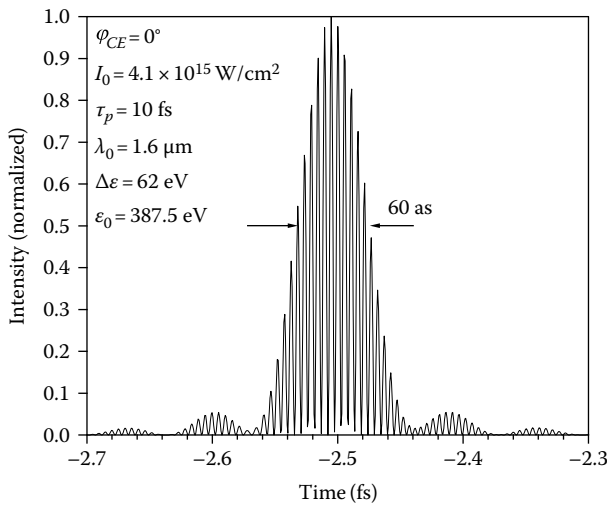


Figure 8.22 The duration of the attosecond pulse centered at 387.5 eV.

at -1.7 fs is only 25 as when a 155 eV spectrum window is applied, as shown in [Figure 8.23](#).

The single atom spectrum is sensitive to the laser intensity. The harmonic spectrum from a single atom at the peak intensity 2×10^{15} W/cm² is shown in [Figure 8.24a](#), where the CE phase of the laser is assumed to be $\varphi_{CE} = 0$ rad. The intensity modulation in the cutoff region, 1.3–1.5 keV, is due to the interference of two x-ray attosecond pulses. The noise-like spectrum in the plateau region, 1–1.3 keV, is formed by the interference between the many attosecond pulses from the long and short trajectories and from different half laser cycles. When the CE phase is changed to $\pi/2$, the whole spectrum exhibits irregular peaks due to the

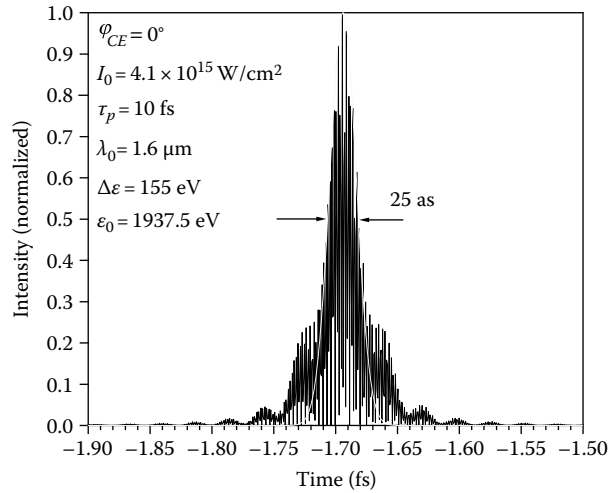


Figure 8.23 The duration of the attosecond pulse centered at 1937.5 eV.

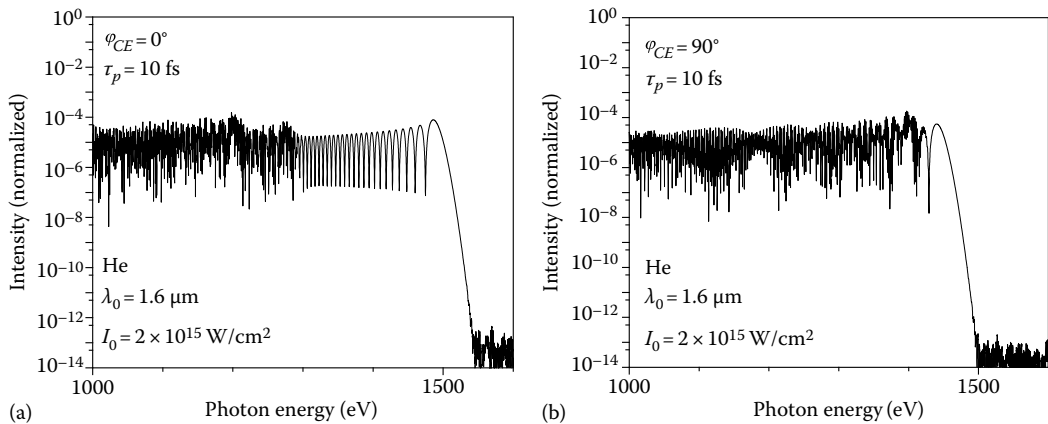


Figure 8.24 The soft-ray spectra generated at a lower intensity $2 \times 10^{15} \text{ W/cm}^2$.

multipulse interference, as shown in Figure 8.24b. In amplitude gating, spectrum filters are used to select the highest photon energy region, which is generated at the higher laser intensity region of the focal volume.

8.4 Polarization Gating

There are drawbacks to producing single attosecond pulses at the cutoff. The usable spectrum range is limited and becomes narrower as the laser pulse duration increases. The signals in the plateau region and originated at the low laser intensity region of laser focus are dumped, which sacrifices the photon flux. The desirable half-cycle laser is very difficult to construct and operate.

Polarization gating can relax the stringent requirement for the laser pulse duration to a certain degree. Its principle, using the semiclassical model, is discussed in Chapter 4. In Chapter 5, polarization gating is simulated under the strong-field approximation. Here, we present some experimental results.

8.4.1 Setup for Measuring Polarization Gated XUV Spectrum

Figure 8.25 depicts the experimental setup in the author's lab to study the attosecond generation in the spectrum domain. Sub-10 fs lasers are required for polarization gating, to reduce the ground state population of the target atoms. Such pulses were generated by focusing the 25 fs lasers from a chirped pulse amplification (CPA) system into a hollow-core fiber, as discussed in Chapter 2. The fused silica fiber had an inner diameter of 0.25 mm and a length of 1.6 m. It was filled with argon gas with a pressure gradient from ~ 0.1 torr at the laser entry to ~ 1 atm at the exit. The white-light pulse from the fiber passed through two pairs of chirp mirrors and became negatively chirped, with a center wavelength at 750 nm. To observe the XUV spectrum changes with the laser pulse duration, the negative chirp of the pulse was compensated by a fused silica plate so that the pulse duration became tunable by adjusting the plate thickness.

The polarization gating field with a time-dependent ellipticity was then formed when the linearly polarized laser beam propagated through

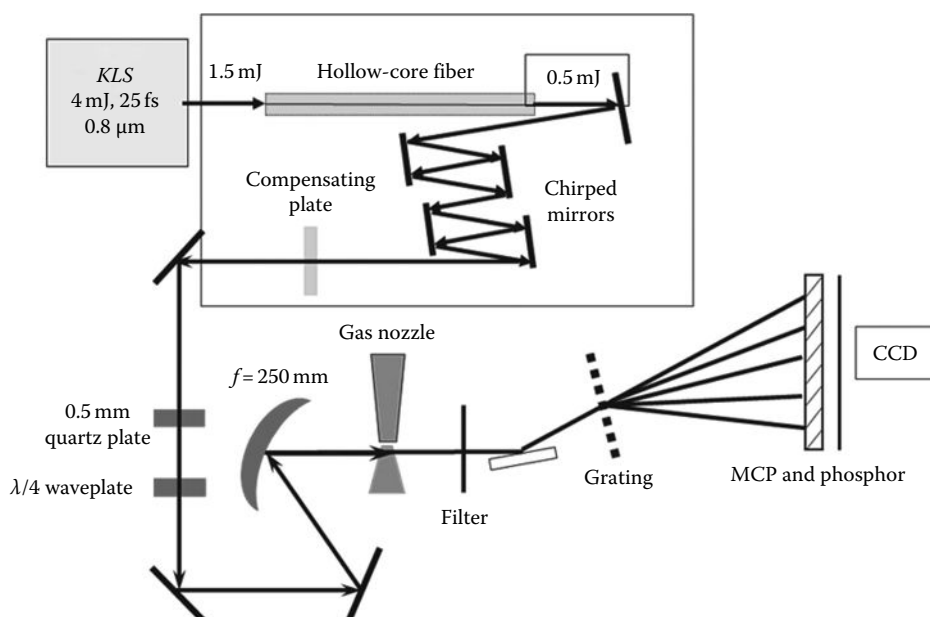


Figure 8.25 The overall system layout for producing a pulse with a time-dependent ellipticity. (Reprinted from B. Shan, S. Ghimire, and Z. Chang, Generation of attosecond XUV supercontinuum by polarization gating, *J. Mod. Opt.*, 52, 277, 2005.)

a 0.5 mm quartz plate and a quarter wave plate. Finally, the laser beam was focused by a parabolic mirror ($f=250$ mm) into an argon gas jet with a local density of $\sim 10^{18}/\text{cm}^3$. The laser pulse energy was $\sim 260 \mu\text{J}$. The generated XUV signal was dispersed by a transmission grating (2000 lines/mm) and recorded by a microchannel plate detector and a cooling CCD camera.

8.4.2 Effects of Laser Pulse Duration

When the polarization gating field is created by the superposition of two counter-rotating circularly polarized pulses, the polarization gate width

$$\delta t_G \approx 0.3 \frac{\tau_p^2}{T_d}, \quad (8.80)$$

where

τ_p is the duration of each pulse

T_d is the delay between them

In the experiments, the delay was fixed at 15 fs by the 0.5 mm quartz plate. The pulse duration could be varied from 8 to 15 fs. The shortest polarization gate width was estimated to be 1.3 fs, corresponding to the 8 fs pulse, which is approximately one half-cycle of the NIR laser field. In this case, we expected that the gated XUV spectrum would become a continuum in the plateau region of the spectrum, corresponding to a single isolated attosecond pulse. This was indeed the case, as Figure 8.26 demonstrated.

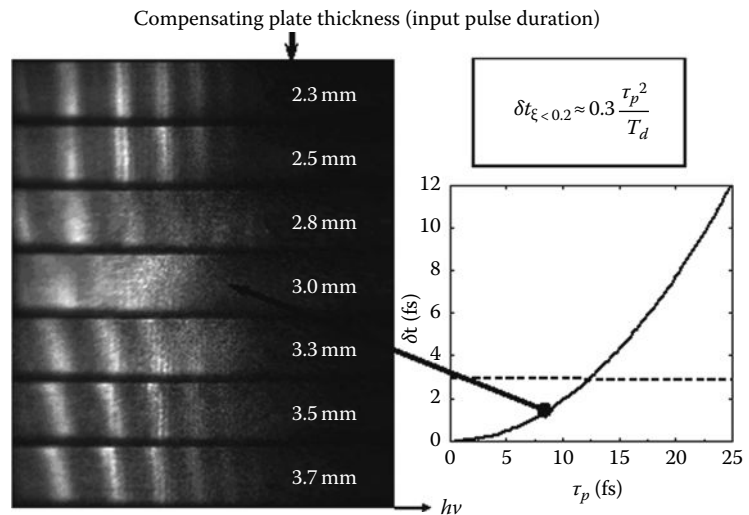


Figure 8.26 The measured XUV supercontinuum at the high-order harmonic generation plateau and cutoff. (Reprinted from B. Shan, S. Ghimire, and Z. Chang, Generation of attosecond XUV supercontinuum by polarization gating, *J. Mod. Opt.*, 52, 277, 2005.)

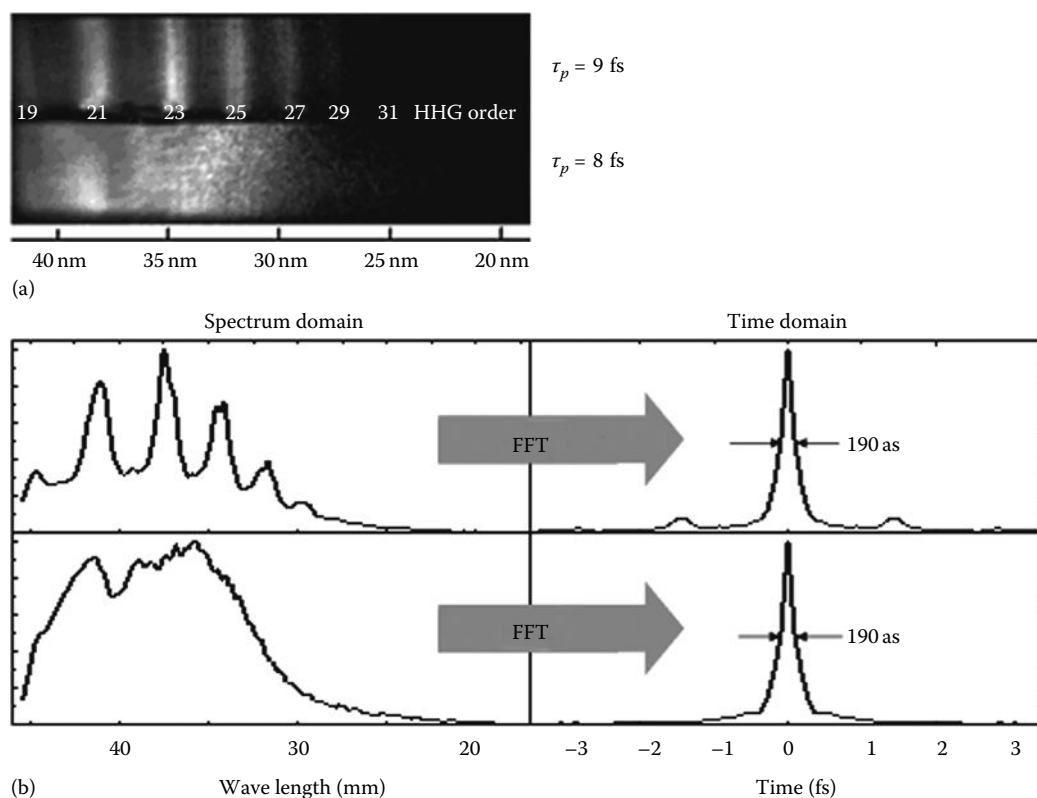


Figure 8.27 (a) The lineout of gated spectra and (b) their Fourier transforms. (Reprinted from B. Shan, S. Ghimire, and Z. Chang, Generation of attosecond XUV supercontinuum by polarization gating, *J. Mod. Opt.*, 52, 277, 2005.)

In the experiments, compensating plates with seven different thicknesses were tried out. When the plate was either too thin (2.3 mm) or too thick (3.7 mm), high-order harmonic spectra with well-resolved peaks showed up. In both cases, the NIR pulse duration was too long. As the thickness approached the optimum value (3.0 mm), the harmonic peaks merged to a supercontinuum. This is the thickness at which the NIR laser pulse reaches the minimum duration (8 fs).

The XUV supercontinuum covered a range from 25 to 40 nm, corresponding to the harmonic order of 19–31. The lineout of two XUV spectrographs taken at 8 and 9 fs, as seen in Figure 8.26, is shown in Figure 8.27. The spectrum measurement alone cannot determine the XUV pulse shape. To get some idea of the XUV pulse, these two spectra were Fourier transformed assuming flat phases. The resultant XUV pulses are also shown in Figure 8.27. As expected, the discrete spectrum resulted in three XUV attosecond pulses with a time interval of a half-cycle of the NIR laser field. The XUV supercontinuum yielded a single attosecond pulse with the transform-limited duration of 190 as.

Polarization gating can be implemented with pulses longer than what is required for amplitude gating. However, the conversion efficiency from laser to XUV decreases with the laser pulse duration. To achieve usable

XUV photon flux, the laser pulse should be close to two cycles, as the simulation in Chapter 5 revealed. Isolated 130 as pulses have been generated by using 5 fs lasers in Nisoli's lab. The CE phase of the laser was stabilized, and the attosecond pulses were characterized with CRAB.

8.5 Double Optical Gating

Double optical gating (DOG) is another gating method for generating isolated attosecond pulses using multi-cycle lasers, although it also works with two-cycle lasers. The phase matching of DOG is discussed in Chapter 6. Here, we focus on the other aspects of the gating, such as the attosecond pulse width and contrast.

8.5.1 Principle of Double Optical Gating

DOG is a combination of two-color gating and polarization gating. The idea is to increase the spacing between the adjacent attosecond pulses in a train by a factor of two (from half of a laser cycle to a full laser cycle) so that a wide polarization gate can be used. This helps to reduce the ground state population depletion when multi-cycle NIR lasers are used to drive the attosecond pulse generation.

When atoms are driven by strong, linearly polarized NIR laser pulses containing many optical cycles, a train of attosecond pulses can be generated, as illustrated by Figure 8.28a. In two-color gating, a weak second harmonic field (10%–20% of the fundamental amplitude), polarized in the same direction as the driving field, is superimposed on the fundamental laser field to break the symmetry of the driving field. The relative phase between the two fields is chosen to be either zero or π , so that the ionization rate difference between the two adjacent field maxima differs by more than two orders of magnitude. In other words, electrons are essentially only ionized when the field is pointing in one direction. As a result, when the combined laser pulse is focused on a gas target, the spacing between the adjacent attosecond pulses is a full fundamental cycle, i.e., 2.6 fs for Ti:Sapphire lasers, as shown in Figure 8.28b.

The laser pulse used for polarization gating is linearly polarized in the middle of the envelope and is elliptically polarized in the leading and trailing edges. When the width of the linearly polarized portion equals the spacing between adjacent attosecond pulses in the pulse train, only a single isolated attosecond pulse can be generated, as depicted in Figure 8.28c.

The polarization gating pulse can be considered to be a combination of two orthogonally polarized fields, where one serves as a driving field while the other acts as a gating field, as discussed in Chapter 4. Here, the driving laser field component is plotted in red. The blue shading represents the suppressing power of the polarization gating. Deep color means the strong suppression of satellite pulses.

When a linearly polarized second harmonic field is added to the driving field, the time interval between adjacent attosecond pulses becomes one full optical cycle of the fundamental wave. To allow a single attosecond pulse emission, the width of the polarization gating should be close to one

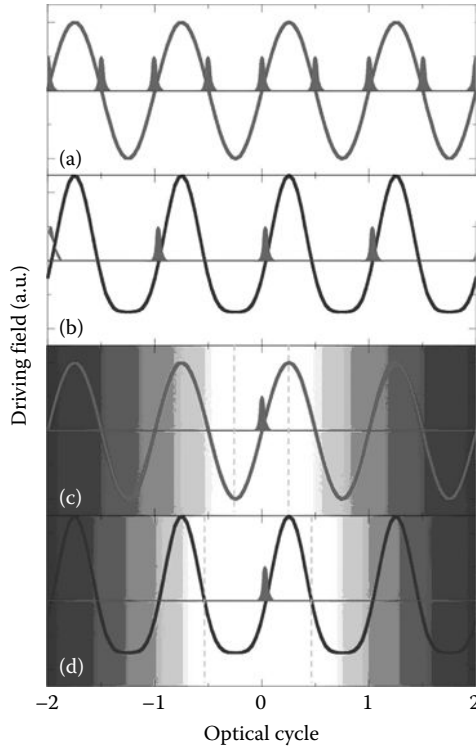


Figure 8.28 Principle of DOG. (a) Attosecond pulse train generated by the laser field at the fundamental frequency. (b) Increase of attosecond pulses spacing by breaking the driving field symmetry. (c) Polarization gating using one color driving laser. (d) Double optical gating.

full cycle of the driving field, which is two times what is required by the conventional polarization gating, as illustrated in Figure 8.28d. The wider gate width allows the use of longer lasers without increasing the ionization probability by the leading edge of the laser.

8.5.2 Gate Width

To be more general, we considered the case that the pulse with a time-dependent ellipticity for polarization gating is generated by the superposition of a left-elliptically polarized and a right-elliptically polarized Gaussian pulse. The field amplitude, E_0 , ellipticity, ε , carrier frequency, ω_0 , and pulse duration, τ_p , are the same for the two pulses. The delay between them is T_d , which is an integer number, n , of the laser optical period. The field can be resolved into a driving field and an orthogonally polarized gating field,

$$\varepsilon_{drive}(t) = E_0 \varepsilon \left[e^{-2 \ln 2 \left(\frac{t - T_d/2}{\tau_p} \right)^2} + e^{-2 \ln 2 \left(\frac{t + T_d/2}{\tau_p} \right)^2} \right] \cos(\omega_0 t + \varphi_{CE}), \quad (8.81)$$

$$\varepsilon_{gate}(t) = E_0 \left[e^{-2 \ln 2 \left(\frac{t-T_d/2}{\tau_p} \right)^2} - e^{-2 \ln 2 \left(\frac{t+T_d/2}{\tau_p} \right)^2} \right] \sin(\omega_0 t + \varphi_{CE})(-1)^n, \quad (8.82)$$

where φ_{CE} is the CE phase of the driving field. The attosecond pulses are generated by the driving field, but are suppressed by the gating field outside of the polarization gate.

The time-dependent ellipticity of the pulse is

$$\xi(t) = \min \left[\frac{\left| 1 - e^{-4 \ln 2 \frac{T_d t}{\tau_p}} \right|}{\varepsilon \left[1 + e^{-4 \ln 2 \frac{T_d t}{\tau_p}} \right]}, \frac{\varepsilon \left[1 + e^{-4 \ln 2 \frac{T_d t}{\tau_p}} \right]}{\left| 1 - e^{-4 \ln 2 \frac{T_d t}{\tau_p}} \right|} \right], \quad (8.83)$$

where the function min takes the smaller value of the two terms. During the time range where the time-dependent ellipticity is less than a threshold value, the suppression power of the gating field is sufficiently small enough for attosecond pulses to be efficiently generated. This range is called the polarization gate. The gate width can be estimated by

$$\delta t_G \approx \varepsilon \frac{\xi_{th}(\eta, q)}{\ln(2)} \frac{\tau_p^2}{T_d}, \quad (8.84)$$

where ξ_{th} is the threshold ellipticity at which the q th harmonic intensity drops to a certain value, η , of that generated by linearly polarized lasers. For $\xi_{th} = 0.2$, the polarization gate width is

$$\delta t_G \approx 0.3 \frac{\varepsilon \tau_p^2}{T_d}. \quad (8.85)$$

Equations 8.84 and 8.85 suggest that the polarization gate can be narrowed by introducing ellipticity to the input pulses.

When polarization gating is combined with two-color gating for the generation of single isolated pulses, the gate width δt_G should equal one optical cycle, which is ~ 2.5 fs for Ti:Sapphire laser pulses compressed by hollow-core fibers. To satisfy this condition for a given laser pulse duration, the ratio must be

$$\frac{\varepsilon}{T_d} \approx \frac{1}{0.3} \frac{T_0}{\tau_p^2} = \frac{8.1}{\tau_p^2}. \quad (8.86)$$

ε cannot be chosen too small, otherwise harmonics are generated outside of the gate. For driving lasers at 800 nm, the minimum ε is approximately 0.3.

For $\varepsilon = 0.35$ and $\varepsilon = 1$, the calculated required delay time for producing a single isolated pulse as a function of the laser pulse duration with DOG is shown in Figure 8.29. Here we assume that the laser period $T_0 = 2.5$ fs. For example, for 25 fs lasers, the required gate width can be obtained by two extreme combinations, i.e., $\varepsilon = 1$ and $T_d \approx 75$ fs or $\varepsilon = 0.35$ and $T_d \approx 26$ fs.

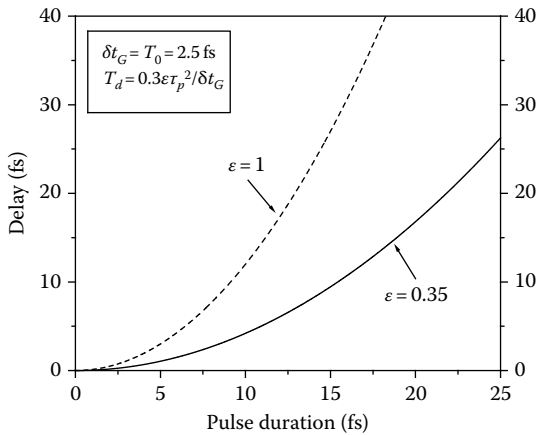


Figure 8.29 The required delay between two counter-rotating elliptically polarized pulses for extracting a single isolated attosecond pulse from a pulse train. Solid line: with conventional polarization gating, the gate width equals one half of an optical cycle. Dashed line: with double gating, the gate width equals one full optical cycle.

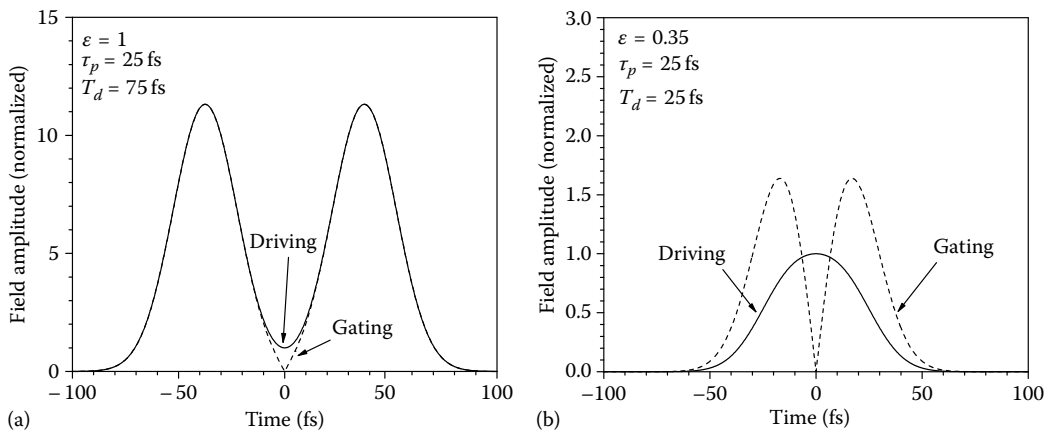


Figure 8.30 The laser field components for DOG with 25 fs lasers and one cycle gate width. The driving and gating fields formed by the combination of (a) a left-hand circularly polarized pulse and a delayed right-hand circularly polarized pulse or (b) a left-hand elliptically polarized pulse and a delayed right-hand elliptically polarized pulse.

8.5.3 Upper Limit of NIR Laser Pulse Duration

The field components corresponding to these two cases are shown in Figure 8.30a and b, respectively. The strength of the driving field inside the polarization gate is the same for the two cases. In the $\varepsilon = 0.35$ case, the delay for the 25 fs laser is close to the pulse duration, which is preferred to reduce ionization of the attosecond by the leading edge of the polarization gating pulse. On the contrary, when $\varepsilon = 1$, the field inside the polarization gate is much weaker than the outside field. The field in the leading edge

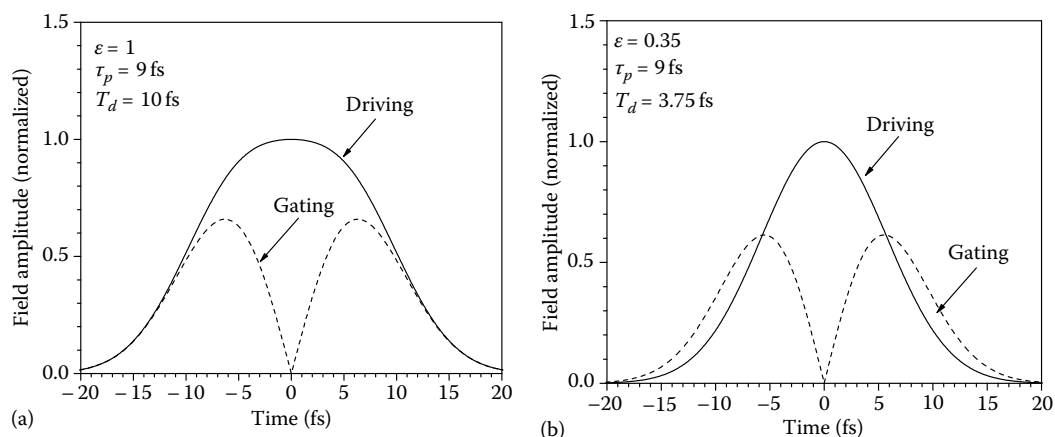


Figure 8.31 The laser field components for DOG with 9 fs lasers and one cycle gate width. The driving and gating fields formed by the combination of (a) a left-hand circularly polarized pulse and a delayed right-hand circularly polarized pulse or (b) a left-hand elliptically polarized pulse and a delayed right-hand elliptically polarized pulse.

may completely deplete the ground state population before the gating starts. With the reduction of NIR laser pulse duration, the ratio between the field amplitude inside and outside the polarization gate increases, which favors the efficiency of attosecond generation, as shown in Figure 8.31.

The dipole moment of the driven atom responsible for attosecond light emission is proportional to the ground state population. The ionization probability for an argon atom by the leading edge of the NIR laser pulse for three different gating situations is shown in Figure 8.32, which was calculated by using the Ammosov–Delone–Krainov (ADK) rate introduced in Chapter 4. The intensity at the center of the gate was chosen as $1.9 \times 10^{14} \text{ W/cm}^2$. The upper limit of pump pulses duration was set by the duration at which the ground state is almost completely depleted.

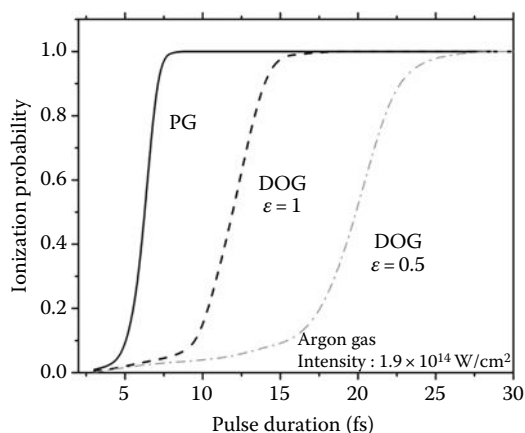


Figure 8.32 Comparison of the ionization probabilities of the target atom argon for three gating conditions. PG stands for polarization gating.

For the same driving field strength in the middle of the gate, the same gate width, and the same laser pulse duration, calculations have shown that the depletion of the ground state population by the laser field before the polarization gate decreases with the ellipticity, ε . This is because both driving field strength and gating field strength before the gate are lower for the smaller ellipticity, ε , which can be seen in Figure 8.30. It can be seen that for sufficiently small ε values, NIR laser pulses as long as 25 fs can still be used for generating isolated attosecond pulses with DOG.

8.5.4 Creating the Gating Laser Field

The two counter-rotating elliptically polarized pulses and the second harmonic pulses can be created using the setup shown in Figure 8.33. There are a total of four optical elements that transform a linearly polarized pulse into the field for DOG. These include a full-wave plate, a fused silica Brewster window (BW), a second quartz plate, and a barium borate (BBO) crystal. A photo of the experimental setup for creating the field for DOG is shown in Figure 8.34.

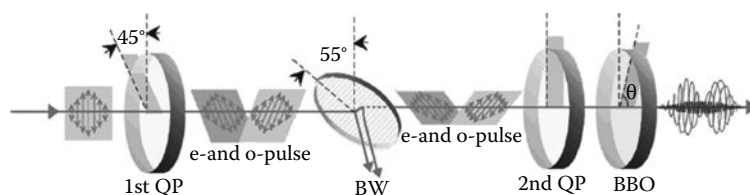


Figure 8.33 Optics for creating the field for DOG. A multi-order whole-wave plate followed by a tunable BW. Afterwards, there is a second quartz plate and BBO crystal that form a quarter-wave plate. (Adapted from X. Feng, S. Gilbertson, H. Mashiko, H. Wang, S.D. Khan, M. Chini, Y. Wu, K. Zhao, and Z. Chang, *Phys. Rev. Lett.*, 103, 183901, 2009.)

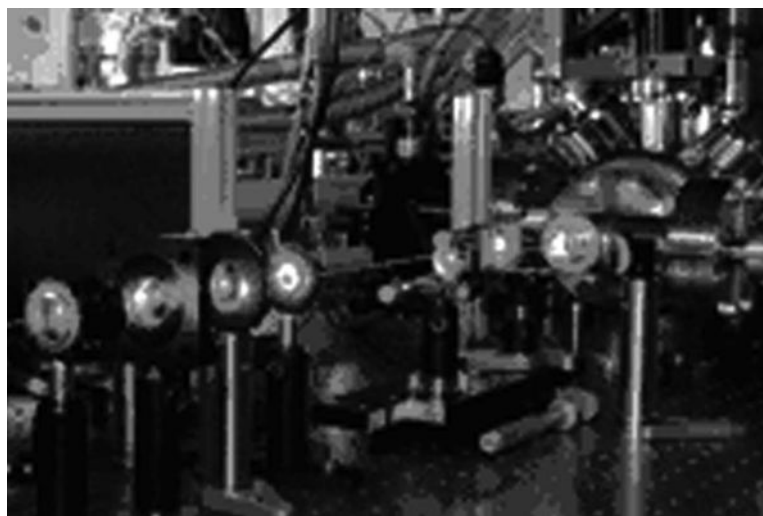


Figure 8.34 Experimental setup for creating the field for DOG.

Assume that the input laser is vertically polarized. The first quartz plate has its optic axis set at 45° with respect to the input polarization, which splits the input pulse into two orthogonal linearly polarized pulses with a delay between them. We chose the delay to be an integer number of the optical cycle. The BW is used to reduce the vertical component of the two pulses so that they can become two elliptical counter-rotating pulses after passing through the subsequent quartz plate and the BBO crystal, which combine to form a quarter-wave plate.

8.5.4.1 Controlling the Delay by the Whole Wave Plate

The delay between the two counter-rotating elliptically polarized pulses is introduced by the difference in group velocity between the ordinary rays (o-rays) and extraordinary rays (e-rays), v_{og} and v_{eg} , in the first quartz plate. Of course, other birefringence materials can also be used.

The amount of delay can be changed by the varying the thickness of the plate, L ,

$$T_d = L \left(\frac{1}{v_{eg}} - \frac{1}{v_{og}} \right). \quad (8.87)$$

The values v_{og} and v_{eg} are slightly different from the phase velocities. Experimentally, the thickness can be tuned with a pair of quartz wedges or by using a set of plates with different thicknesses.

The group velocities can be calculated by

$$\frac{1}{v_{og}} = \frac{n_o}{c} - \frac{\lambda}{c} \frac{dn_o}{d\lambda}, \quad (8.88)$$

$$\frac{1}{v_{eg}} = \frac{n_e}{c} - \frac{\lambda}{c} \frac{dn_e}{d\lambda}, \quad (8.89)$$

where $n_o(\lambda)$ and $n_e(\lambda)$ are expressed by the Sellmeier equations of quartz. For quartz, $v_{og}(0.8 \mu\text{m}) = 0.192984 \mu\text{m/fs}$, $v_{eg}(0.8 \mu\text{m}) = 0.191806 \mu\text{m/fs}$. At 750 nm, which is the center wavelength of the hollow-core fiber compressor, $v_{og}(0.75 \mu\text{m}) = 0.191545 \mu\text{m/fs}$, $v_{eg}(0.75 \mu\text{m}) = 0.1912725 \mu\text{m/fs}$. The dependence of the delay on the thickness is shown in [Figure 8.35](#).

8.5.4.2 Controlling the Ellipticity by Brewster Window

The ellipticity, ε , is controlled by the incident angle of the BW. The Fresnel reflections on the two uncoated surfaces allow the amplitude of the driving field (s wave) to be reduced, while maintaining the gating field (p wave). At each surface, the Brewster angle is

$$\theta_B = \arctan(n_{\text{glass}}). \quad (8.90)$$

At the center wavelength of the Ti:Sapphire CPA lasers, 800 nm, the indices of refraction n_{glass} for fused silica and SF11 are 1.45332 and 1.76475, respectively. The $\theta_B = 55.47^\circ$ for the former and 60.46° for the latter.

The Fresnel reflection is zero for the p wave. For the s wave, the reflectance

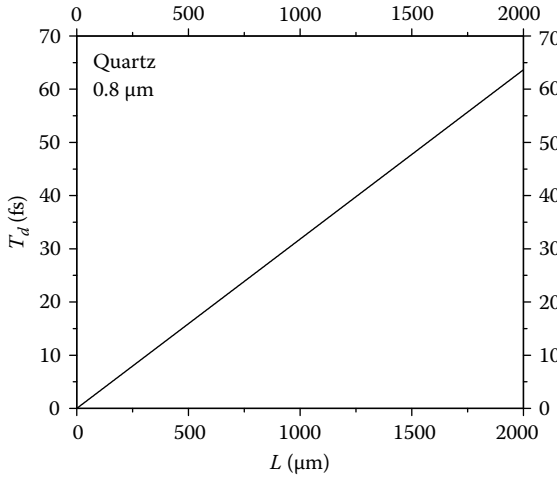


Figure 8.35 The group delay between the pulses of the o-ray and e-ray introduced by a quartz plate.

$$R = \left[\frac{\sin \left(\theta_B - \text{asin} \left(\frac{\sin \theta_B}{n_{\text{glass}}} \right) \right)}{\sin \left(\theta_B + \text{asin} \left(\frac{\sin \theta_B}{n_{\text{glass}}} \right) \right)} \right]^2, \quad (8.91)$$

which is 0.128 for fused silica and 0.264 for SF11.

Taking into account the reflection loss of both surfaces, the ellipticity of the transmitted beam

$$\varepsilon = \sqrt{(1 - R)^2} = 1 - R, \quad (8.92)$$

which is 0.872 for fused silica and 0.736 for SF11. This is the minimum ε that can be achieved by using one BW. Adding another window can further reduce the value of ε

$$\varepsilon = \sqrt{(1 - R)^4} = (1 - R)^2, \quad (8.93)$$

which gives 0.76 and 0.542 for the two materials. The angle of incidence can be changed to tune the ellipticity.

8.5.4.3 BBO Crystal

The BBO crystal serves two purposes. First, it generates the second harmonic wave from the gating field component. The second harmonic field is polarized in the vertical direction as a result of the type I phase-matching configuration.

Second, it combines with the second quartz plate to make a zero-order quarter-wave plate over a broad wavelength range, taking advantage of the fact that quartz is a positive crystal but BBO is a negative uniaxial crystal,

$$|(n_{o,q}L_q + n_{o,BBO}L_{BBO}) - (n_{e,q}L_q + n_{e,BBO}(\theta)L_{BBO})| = \frac{\lambda_0}{4}, \quad (8.94)$$

where $n_{o,q}$ and $n_{e,q}$ are the indices of refraction of quartz for the o-ray and e-ray, respectively. $n_{o,BBO}$ is the index of refraction of BBO for the o-ray. $\theta = 29^\circ$ is the phase-matching angle of BBO at the laser wavelength $\lambda_0 = 800$ nm. $n_{e,BBO}(\theta)$ is the index of refraction of BBO for the e-ray at the phase-matching angle.

8.5.5 Numerical Simulations

It is assumed that a Gaussian laser beam is focused 3.5 mm before a 1 mm long cell filled with 5 torr of argon gas. As discussed in Chapter 6, the photon emission from the long trajectory is suppressed because of poor phase matching with such a configuration. The center wavelengths of the fundamental wave and second harmonic wave are 750 and 375 nm, respectively, and the radius of the focal spot is 30 μm . The gas cell length is shorter than the Rayleigh range, 3.77 mm, to avoid large CE phase variation inside the cell.

The laser beam propagates along the z direction. At a given position of the target, the laser field is expressed as

$$\vec{\varepsilon}(t) = \varepsilon_{drive}(t)\hat{i} + \varepsilon_{gate}(t)\hat{j}, \quad (8.95)$$

where $\hat{i}$ and $\hat{j}$ are the unit vectors in the x and y directions, respectively.

We consider the case where $\varepsilon = 1$. When the delay between the two counter-rotating Gaussian pulses T_d is an integer times of the laser period, the driving field can be expressed as

$$\begin{aligned} \varepsilon_{drive}(t) = E_0 \left[\left(e^{-2 \ln 2 \frac{(t+T_d/2)^2}{\tau_p^2}} + e^{-2 \ln 2 \frac{(t-T_d/2)^2}{\tau_p^2}} \right) \cos(\omega_0 t + \varphi_{CE}) \right. \\ \left. + a_{\omega,2\omega} \left(2e^{-2 \ln 2 \left(\frac{T_d/2}{\tau_p} \right)^2} \right) e^{-2 \ln 2 \frac{t^2}{\tau_{2\omega}^2} \cos(2\omega_0 t + 2\varphi_{CE} + \phi_{\omega,2\omega})} \right] \end{aligned} \quad (8.96)$$

and the gating field as

$$\varepsilon_{gate}(t) = E_0 \left(e^{-2 \ln 2 \frac{(t+T_d/2-T_0/4)^2}{\tau_p^2}} - e^{-2 \ln 2 \frac{(t-T_d/2-T_0/4)^2}{\tau_p^2}} \right) \sin(\omega_0 t + \varphi_{CE}), \quad (8.97)$$

where E_0 is the amplitude of the circularly polarized fundamental laser field with a carrier frequency ω_0 and a pulse duration τ_p . The delay, $T_0/4$, between the gating and the driving fields is introduced by the quarter-wave plate, i.e., the quartz plate and BBO combination in the experimental setup, as is examined in Problem 8.9.

The CE phase of the fundamental laser field changes with z due to the Gouy phase shift. The value of φ_{CE} is referenced to that at the laser focus. The CE phase of the second harmonic field is $2\varphi_{CE}$, while $a_{\omega,2\omega}$ is the ratio of the amplitudes between the second harmonic field and the driving field at the center of the polarization gate ($t=0$). The relative phase delay between the two fields is $\phi_{\omega,2\omega}$ when $\varphi_{CE}=0$.

The gate width is set to 2.4 fs by assuming $\tau_p = 10$ fs and $T_d = 12.5$ fs. The duration of the linearly polarized second harmonic field is $\tau_{2\omega} = 25$ fs, which is not a critical parameter.

The simulation took into account propagating effects in the target. First, the femtosecond driving laser field for the double optical gating was calculated for all spatial grid points by solving a three-dimensional wave equation that included the plasma defocusing effects. The ionization of the atoms by the laser field was calculated by the ADK model. Next, the dipole moment of a single atom at each grid point was calculated by making the strong-field approximation. Finally, the dipole moments were inserted as sources into the wave equation in the frequency domain to yield the harmonic field (near-field). The XUV spectra in the far-field were obtained by performing the Hankel transform. The signals below 33 eV (the 20th harmonic) were blocked by a high-pass filter when the XUV field was Fourier transformed to the time domain.

For comparison, the calculated attosecond pulses and the corresponding XUV spectra with different gating schemes are shown in [Figure 8.36](#).

8.5.5.1 One-Color Linearly Polarized NIR Laser

When argon gas was driven by a 10 fs linearly polarized fundamental wave (one color), a train of eight XUV attosecond pulses was generated, as shown in the bottom panel in the upper graph of [Figure 8.36](#). The spacing between the adjacent pulses is one half of an optical cycle. The pulses are from the short quantum trajectories, while the long trajectory contributions are suppressed by the phase mismatch during the propagation. In the spectral domain, odd-order high harmonic peaks are observed, as shown in the bottom panel in the lower graph of [Figure 8.36](#).

8.5.5.2 One-Color Polarization Gating

The number of pulses is reduced to three with polarization gating using the fundamental wave laser. The CE phase chosen here favors the pulse in the middle. The separation between the satellite pulses from the main pulse is 1.25 fs. They slip through the soft edge of the polarization gate. The transmission of the gate as a function of time is a Gaussian shape. The intensity of the satellite pulses is $\sim 15\%$ of the main pulse. Such pulses are not considered single isolated pulses when a strict definition is used.

8.5.5.3 Two-Color Gating

Alternately, when a second harmonic field with an amplitude ratio, $a_{\omega,2\omega}$, of 15% of the fundamental wave is added to the linearly polarized fundamental wave with the same polarization direction, the number of pulses in the train is also reduced from eight to three, as a result of the two-color gating effects. However, the intensity of the satellite pulses is much higher than in the case of polarization gating. They are almost comparable to the main pulse in the middle. The spacing between the attosecond pulses in the train is a full fundamental wave cycle, which corresponds to an XUV spectrum consisting of both odd and even high harmonics.

8.5.5.4 Double Optical Gating

When the second harmonic field is added to the driving field of the polarization gating pulse, it turns off the two satellite pulses left by the polarization gating. Only a single isolated attosecond pulse survives

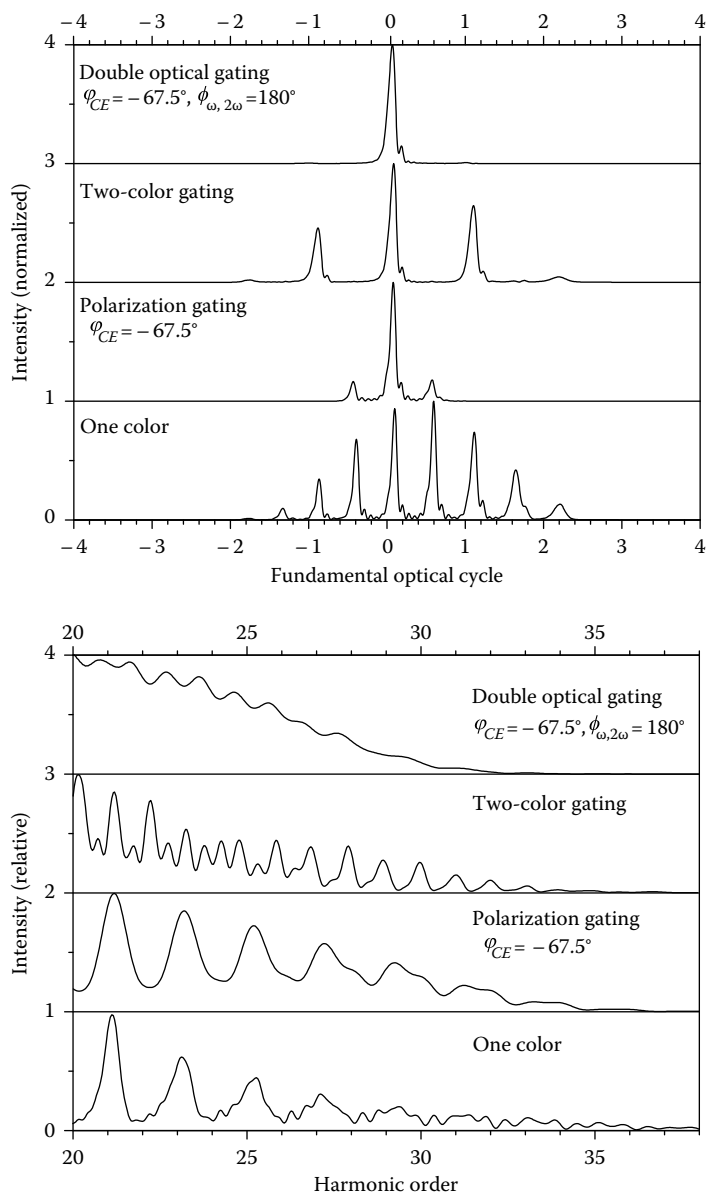


Figure 8.36 Attosecond pulses (a) and their spectra (b) generated by four different types of laser fields. From the bottom panel to the top: with a linearly polarized one color field, with conventional polarization gating, a second harmonic field is added to the linearly polarized fundamental field, and a second harmonic field is added to the polarization gating field. (From Z. Chang, *Phys. Rev. A*, 76, 051403(R), 2007.)

the power of DOG, as shown in the top panel of the upper graph in Figure 8.36, which corresponds to a supercontinuum in the spectrum domain that covers more than 10 eV. There is a small modulation on the spectrum, which comes from satellite pulses that are too low to be seen on the linear scale. The duration of the single isolated pulse is ~ 200 as.

The mixing of the second harmonic field with the polarization gating field introduces many “knobs” that can control the electron trajectories, which includes the second harmonic field strength $a_{\omega,2\omega}$, the relative phase between the fundamental and the second harmonic fields $\phi_{\omega,2\omega}$ as well as the CE phase.

It was found that the second harmonic field amplitude required to reduce the satellite pulse down to 0.7% of the strong pulse is 10% of the NIR field, which corresponds to 1% of NIR intensity. Such a weak second harmonic field is generated by the BBO crystal in the setup shown in Figure 8.33. The relative phase should be set close to 180° when the CE phase chosen is 67.5° .

8.5.5.5 Effects of CE Phase

Figure 8.37 shows the attosecond pulses when the CE phase changes from 0° to 360° . Here the relative phase is set at $\phi_{\omega,2\omega} = 180^\circ$. The CE phase shift moves the main pulse inside the polarization gate, because the electron re-collision time is determined by the CE phase. When the pulse is generated at the center of the gate, the intensity is the highest and the two satellite pulses are suppressed by the gating actions. Otherwise, two weaker pulses separated by one fundamental cycle are produced. The time-integrated signals repeat every 360° . As a comparison, the periodicity is 180° for conventional polarization gating.

For other relative phase $\phi_{\omega,2\omega}$ values, the number of attosecond pulses generated at a given CE phase is different from that in Figure 8.37. It is therefore necessary to stabilize and control the value of both the relative phase and the CE phase in order to generate single isolated attosecond pulses with DOG. In the setup shown in Figure 8.33, the relative phase can be tuned by rotating the phase-matching angle of the BBO crystal. The CE phase is set by the input pulse.*

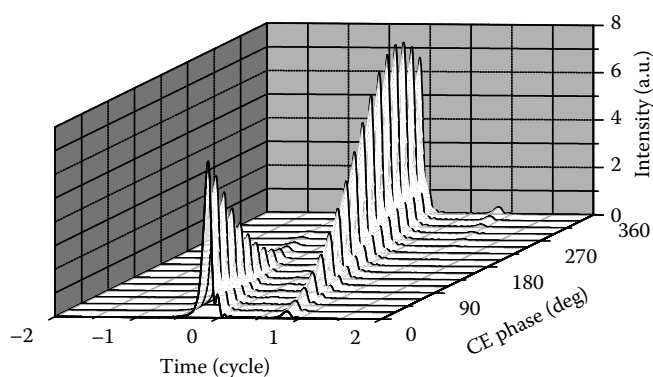


Figure 8.37 The dependence of the attosecond pulse number and intensity on the CE phase on the fundamental wave pulse. (From Z. Chang, *Phys. Rev. A*, 76, 051403(R), 2007.)

* The principle of double optical gating is discussed in more details in Chang, Z., Controlling attosecond pulse generation with a double optical gating, *Phys. Rev. A* 76, 051403(R) (2007).

8.6 Measurement of the XUV Pulse Duration*

8.6.1 Experimental Setup

The experimental setup for characterizing the isolated attosecond pulses generated with DOG in the author's lab is shown in [Figure 8.38](#), which consists of three parts: the CPA laser, the femtosecond FROG, and the attosecond streak camera.

8.6.1.1 Chirped Pulse Amplification with Spectral Shaping

The driving laser system is shown in [Figure 8.38a](#). The front end is a Kerr lens mode-locked Ti:Sapphire oscillator. It produces 3 nJ, 12 fs pulses at 78 MHz. The CE offset frequency of the oscillator was locked to a quarter of the repetition rate. The pulses with the identical CE phase were picked from the oscillator pulse train by a Pockels cell. These were then stretched to ~ 80 ps by a grating-based Martinez type stretcher.

The stretched pulses were amplified at 1.5 kHz with a Ti:Sapphire 14-pass amplifier. The Ti:Sapphire crystal was cooled to liquid nitrogen temperature to reduce the thermal lens effect. After the first seven-pass preamplification, the pulses with ~ 10 μ J were passed through a telescope to match the pump laser size in the crystal so that the pump energy could be efficiently extracted by the second stage seven-pass power amplification. Another Pockels cell with a 10 ns window was used to suppress the amplified spontaneous emission generated during the preamplification stage. After the power amplification, the 5 mJ pulses were compressed by a pair of gratings to 2 mJ, 33 fs, with 26 nm spectrum bandwidth. The output fluctuation was reduced to $<1\%$ by feedback controlling the transmission of the first Pockels cell, as discussed in Chapter 2. The slow drift of the CE phase in the amplifier was compensated through feedback control of the grating separation in the stretcher, as discussed in Chapter 3.

Because the ground state depletion of the target gas is reduced by using shorter lasers, the energy conversion efficiency from the laser to the attosecond pulse is higher. There are two major factors that limit the pulse duration from a CPA. The first one is the dispersion compensation. The material dispersions in multipass amplifiers, such as the laser described in this section, have less material dispersion than regenerative amplifiers. It is therefore easier to generate shorter femtosecond pulses with a multipass configuration.

The second factor is gain narrowing. The width of the stimulated emission cross section curve of the Ti:Sapphire crystal at liquid nitrogen temperature is narrower than that at room temperature. The 33 fs duration was set by the gain narrowing in the amplifier when the crystal is cooled. A simple scheme was implemented here to shorten the laser pulse from the CPA laser. A 300 μ m thick birefringent quartz plate (BP) was inserted as a spectrum shaping filter before the polarizer between the first and second seven passes. It introduced higher loss at the central frequency than in the wings of the

* This section is adapted from the publication Gilbertson, S., Wu, Y., Khan, S.D., Chini, M., Zhao, K., Feng, X., and Chang, Z., Isolated attosecond pulse generation using multicycle pulses directly from a laser amplifier, *Phys. Rev. A* 81, 043810 (2010).

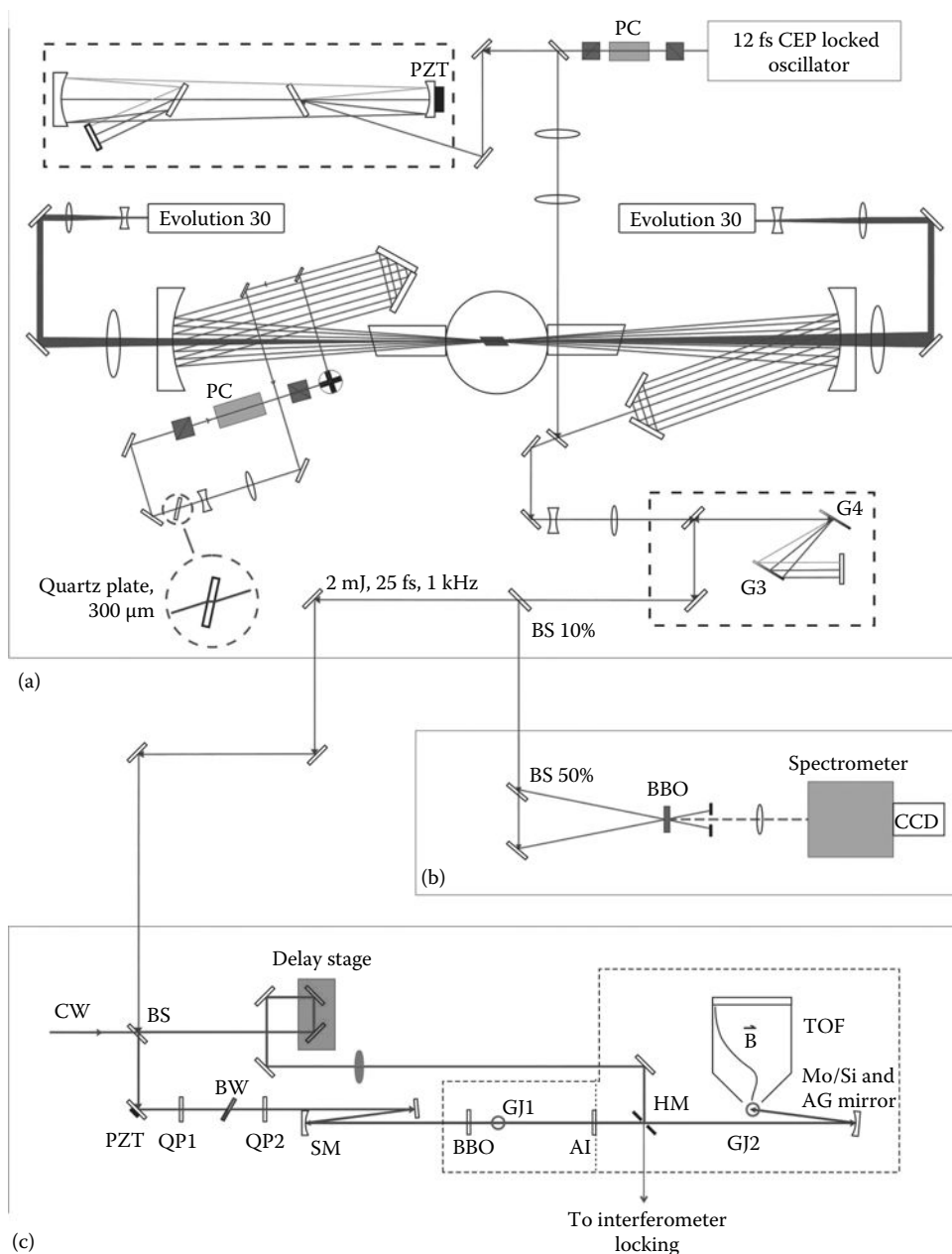


Figure 8.38 (a) Schematic diagram of the laser system. (b) Schematic diagram of the SHG-FROG. (c) The attosecond streak camera setup. BS, beamsplitter; PZT, piezoelectric transducer; QP1 and QP2, quartz plates; BW, Brewster window; SM, spherical mirror; GJ1 and GJ2, gas targets; AI, aluminum filter; HM, hole-drilled mirror; TOF, time-of-flight detector. (From S. Gilbertson, Y. Wu, S.D. Khan, M. Chini, K. Zhao, X. Feng, and Z. Chang, *Phys. Rev. A*, 81, 043810, 2010.)

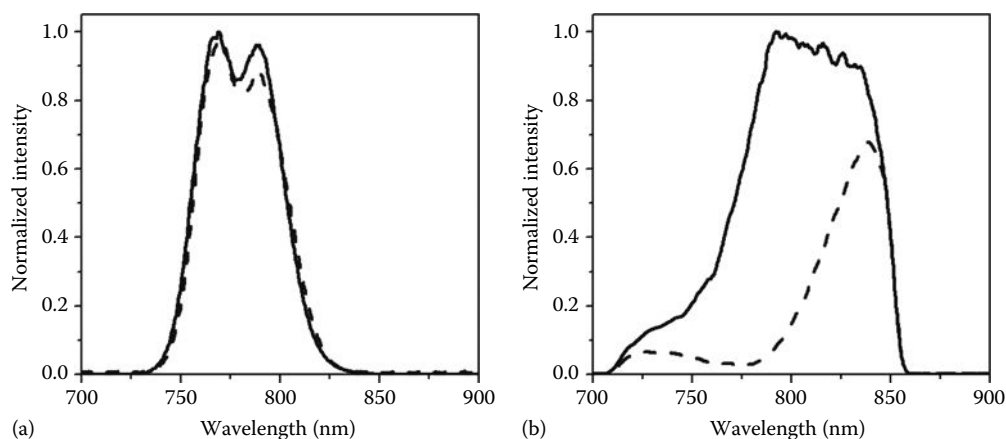


Figure 8.39 Calculated and experimental spectrum for the BP spectrum shaping. (a) Calculated (solid) and measured (dashed) amplified output spectrum with BP. (b) Measured seven-pass spectrum before the plate (solid curve), after the plate and a polarizer (dashed). (From S. Gilbertson, Y. Wu, S.D. Khan, M. Chini, K. Zhao, X. Feng, and Z. Chang, *Phys. Rev. A*, 81, 043810, 2010.)

gain, thus compensating the effects of the gain narrowing. The amplified output spectrum with the spectral shaping reached 50 nm, as shown in Figure 8.39. By improving the spectral shaping with additional optics (not shown), 20 fs pulses with 2 mJ energy are generated from this laser.

8.6.1.2 Femtosecond FROG

The shape and phase of the pulses from the CPA laser are measured by using a single-shot, second-harmonic generation frequency-resolved optical gating (SHG-FROG). The FROG setup is shown in Figure 8.38b. Ten percent of the output beam from the amplifier, approximately 0.2 mJ, is diverted to the SHG-FROG, where it is split evenly into two beams with a broadband beam splitter. The reflected beam, after passing through a compensating plate, and the transmitted beam cross with a small angle at a 5- μm -thick, type I BBO crystal (phase-matching angle of 29.2°) with a 1 cm^2 aperture for the second-harmonic generation.

The finite laser beam size and the angle between the two beams make it possible to generate 400 nm light across the BBO, with different delays between the pulses from the two beams. The 400 nm light distribution on the BBO crystal is imaged onto the entrance slit of the grating spectrometer with a lens of focal length 250 mm and demagnification of 1/2. The output of the imaging spectrometer is recorded on a UV CCD camera. One dimension of the image is the wavelength, whereas the other dimension is the time delay.

Finally, a FROG algorithm is used to reconstruct the laser pulse from the measured FROG trace. The laser pulse is 25 fs long when the quartz spectrum filter is installed. Figure 8.40a shows the measured FROG trace and Figure 8.40b the retrieved FROG trace. Figure 8.40c shows the retrieved pulse shape in the time domain along with the phase. The marginal compression is shown in Figure 8.40d. The reconstructed fundamental wave spectrum agrees well with the laser spectrum directly measured with another spectrometer. The excellent agreement validates the FROG reconstruction.

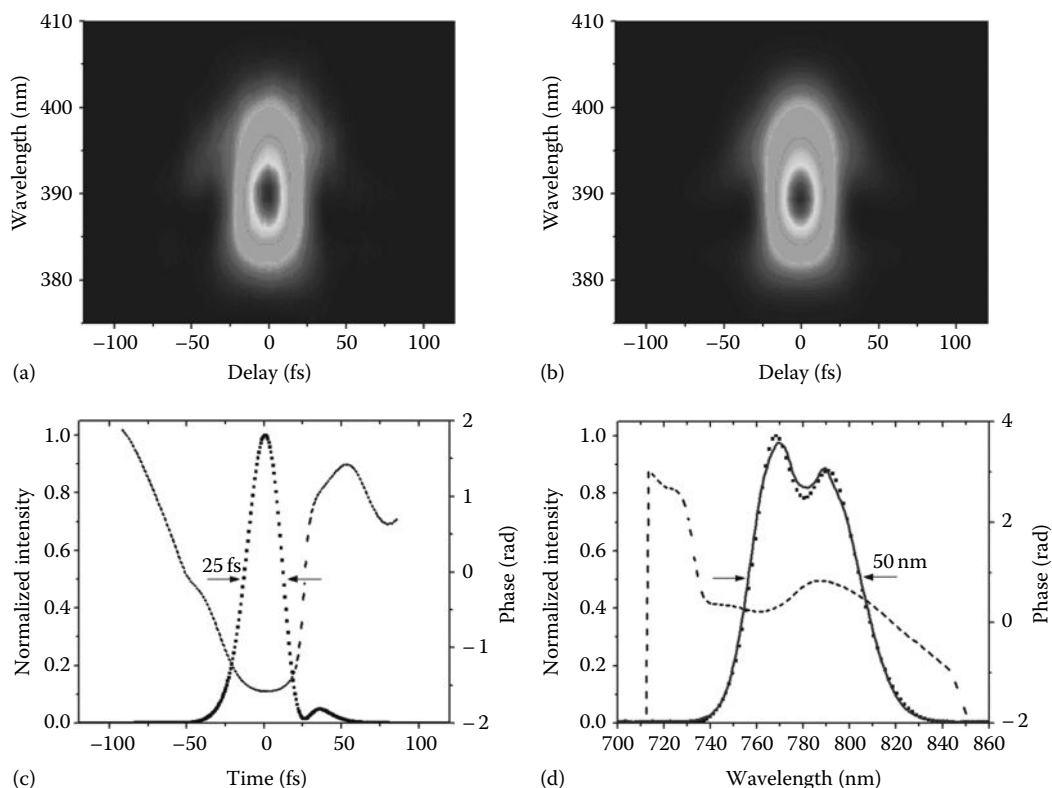


Figure 8.40 Characterization of the 25 fs laser pulse by FROG. (a) The measured FROG trace. (b) The reconstructed FROG trace. (c) The retrieved pulse shape (dotted line) and phase (dashed line). (d) The retrieved power spectrum (dotted line) and phase (dashed line) and independently measured spectrum (solid line).

8.6.1.3 Attosecond Streak Camera

The isolated XUV pulses are characterized using the FROG-CRAB method based on attosecond streaking. A Mach-Zehnder interferometer configuration is used to control the temporal and the spatial overlap of the attosecond XUV beam and the NIR streaking field, as illustrated in Figure 8.38c and in Figure 8.41. The two arms of the Mach-Zehnder interferometer are stabilized by co-propagating a continuous wave (CW) laser with a center wavelength of 532 nm through the interferometer, as illustrated by Figure 8.41. The interference signal of the green beam is used to feedback control a mirror.

The 25 fs pulses directly from the CPA amplifiers are used to generate the attosecond pulses with the DOG technique. The 2 mJ linearly polarized laser pulse is split by a broadband beam splitter, with 80% of the beam being used for the attosecond pulse generation. The other 20% is for streaking the photoelectrons.

The NIR laser beam for attosecond pulse generation passes through the four birefringence optics (the multi-order whole-wave plate, the fused silica BW, and the second quartz plate and the BBO) before being focused by an $f=375$ mm spherical mirror on the gas target. The BBO is located

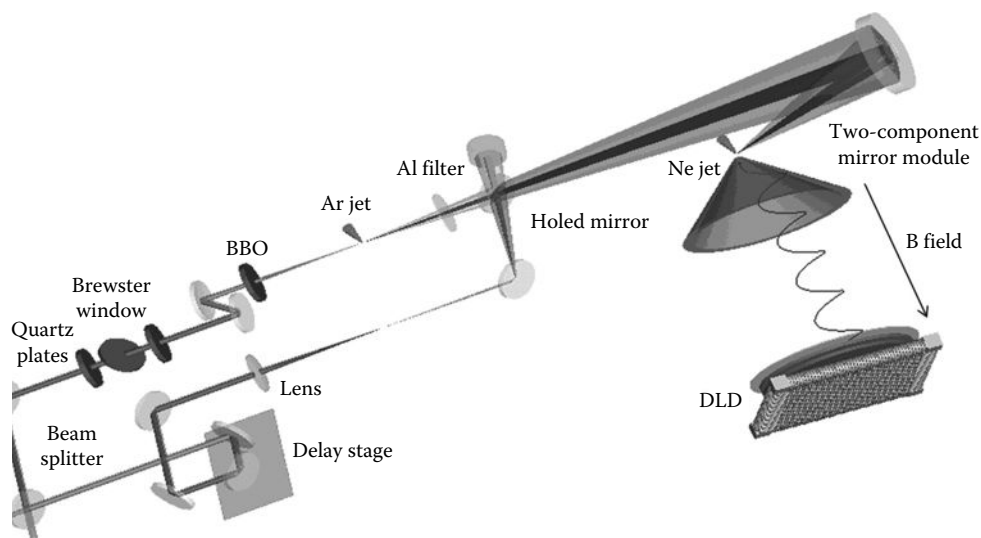


Figure 8.41 Schematic diagram of the attosecond streaking setup.

inside the vacuum to avoid the large group delays between the fundamental and second harmonic beams when passing through the vacuum window. The polarization gate width is estimated to be approximately 2.6 fs, which is one cycle at the fundamental laser wavelength to satisfy the requirement of DOG.

The gas target is placed ~ 2 mm after the laser focus to optimize the phase matching of the attosecond pulse generation process from the short trajectory. The interaction length of the first gas jet is 0.5 mm, which is shorter than the Rayleigh range of the laser focus, so that the variation of the CE phase is small inside the target. The attosecond XUV pulses from the target then pass through an aluminum (Al) filter to reject the residual laser light. The group delay dispersion of the Al is negative in the XUV range of interest, which compensates the positive chirp of the attosecond pulse from the short trajectory.

The linearly polarized streaking NIR laser beam passes through a delay stage and recombines with the attosecond XUV beam at a hole-drilled mirror. The mirror allows the attosecond pulse to pass the hole and reflects the fundamental laser so that their propagation after the mirror becomes collinear.

The two beams are then focused to a second gas target by a concentric set consisting of a molybdenum/silicon (Mo/Si) multilayer XUV mirror and a silver (Ag) laser mirror. The Mo/Si mirror has a reflectivity of $\sim 8\%$ over a 30–60 eV bandwidth, which reflects the attosecond beam. The Ag mirror with a hole in the center reflects the streaking NIR beam.

The second target is krypton gas. The ionization potential is 14 eV, which is suitable for characterizing attosecond pulses with relatively low photon energy. The lower ionization potential shifts the photoelectrons up in energy so that they are easily distinguishable from the ATI peaks found at near-zero energy. The photoelectrons generated by the attosecond pulses are given a momentum shift whose magnitude depends on the vector potential of the fundamental laser. A closed-loop piezoelectric transducer

(PZT) unit, to which the Mo/Si mirror is attached, is used to introduce delay between the XUV and NIR pulses with resolution better than 4 as.

The XUV photoelectrons emitted from the detection gas target are next detected by an electron time-of-flight (TOF) spectrometer, as discussed in Chapter 7. A uniform magnetic field (~ 1.6 gauss) generated by a set of Helmholtz coils is applied along the flight axis between the gas jet and the detector in order to increase the acceptance angle to more than 28° . The TOF detector in this setup has angular resolution over a 28° collection angle. The photoelectrons pass a region of uniform magnetic field and are incident onto a position-sensitive microchannel plate and a delay-line position-sensitive detector. The energy resolution of the TOF detector is better than 0.6 eV at 35 eV electron energy.

The energy spectrum of the photoelectron is then collected as a function of the delay between the attosecond pulse and the streaking laser field, yielding a two-dimensional streaked spectrogram. The attosecond pulse and phase are reconstructed using the iterative PCGPA.

8.6.2 Dependence of Attosecond Electron Spectrum on CE Phase

The measurement of the photoelectron spectrum without the NIR streaking depends strongly on the CE phase of the NIR lasers, as shown in Figure 8.42. The XUV spectra in Figure 8.42a and b are generated from argon and neon, respectively. The CE phase is varied by changing the separation of the gratings in the stretcher of the CPA system.

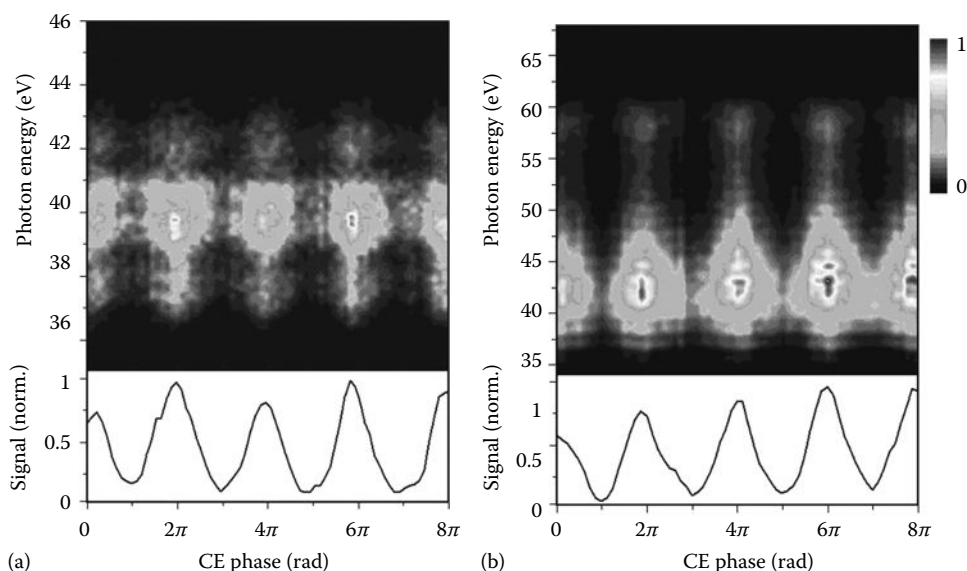


Figure 8.42 (a) The photoelectron energy spectrum plotted as a function of the CE phase of the input laser. The generation gas is argon. The plot across the bottom is the signal strength integrated along the energy axis. (b) The photoelectron spectrum from a generation gas target of neon plotted as a function of the CE phase. The lower plot represents the signal integrated along the energy axis. (From Gilbertson, S. et al., *Phys. Rev. A*, 81, 043810, 2010.)

Each CE phase slice is integrated for 30–60 s, while the CE phase is slowly swept from 0 to 2π . The spectrum is fairly continuous, covering a broad range of the CE phase, which means that isolated attosecond pulses are generated in the CE phase range. In both cases, the variation of the spectrum intensity shows a 2π periodicity, as the simulations in the previous section predict. The width of the integrated spectrum curve (lower plots of (a) and (b)) in the 2π CE phase range is a measure of the gate width. The signal nearly reaches zero for certain values of the CE phase, implying that the gate width is less than one cycle. At those values of the CE phase, the attosecond pulse generation is strongly suppressed, since they would occur outside of the gate. The strongest attosecond pulse is produced for the CE phase value that generates the pulse at the center of the gate. Locking the CE phase to the value of the highest photoelectron yield will maximize the count rate, which is always crucial for attosecond application experiments.

For the terawatt (TW) laser systems that operate at 10 Hz, it is difficult to stabilize the CE phase. The results in [Figure 8.42](#) suggest that one should set the gate width at much less than one optical cycle. In that case, isolated attosecond pulses are always generated, although the flux may change significantly from shot to shot.

8.6.3 Reconstruction of the Attosecond Pulse

The measured CRAB trace for attosecond pulses generated from argon gas, which is the spectrogram of photoelectron energy as a function of the delay between the XUV and NIR fields, is shown in [Figure 8.43a](#). It contains 10 laser cycles of delay to increase the accuracy of the reconstruction. At each time delay slice, the signal is accumulated for 60 s to reduce the shot noise. [Figure 8.43b](#) shows the reconstructed trace. [Figure 8.43c](#) shows the temporal intensity profile of the XUV pulse and the temporal phase. The FWHM of the pulse is of 163 as duration. The inset figure shows the temporal profile on a logarithmic scale over several laser cycles. It is clear that the contributions from pre- and post-pulses are less than 0.1% of the main peak, demonstrating that the pulse is indeed an isolated attosecond pulse.

[Figure 8.43d](#) shows a comparison between the experimental XUV-only spectrum (dotted line) and the retrieved XUV spectrum (solid line). The flat spectrum phase indicates that the pulse is nearly a transform-limited pulse. The good agreement between the measured and reconstructed spectra is an indication of a valid reconstruction. However, unlike the femtosecond SHG-FROG, the measured CRAB pattern already contains the fundamental spectrum. In fact, as the Fourier analysis of [Figure 8.6b](#) shows, most of the signal in the CRAB pattern is the DC component, which is the XUV spectrum without laser. Therefore, the significance of the marginal comparison for CRAB is not as meaningful as for SHG-FROG.

The generation of isolated attosecond pulses with 25 fs lasers offers two advantages. First, the driving lasers without the hollow-core fibers are much easier to work with than the ≤ 5 fs lasers used in attosecond generation with amplitude gating. Second, they can be generated from high

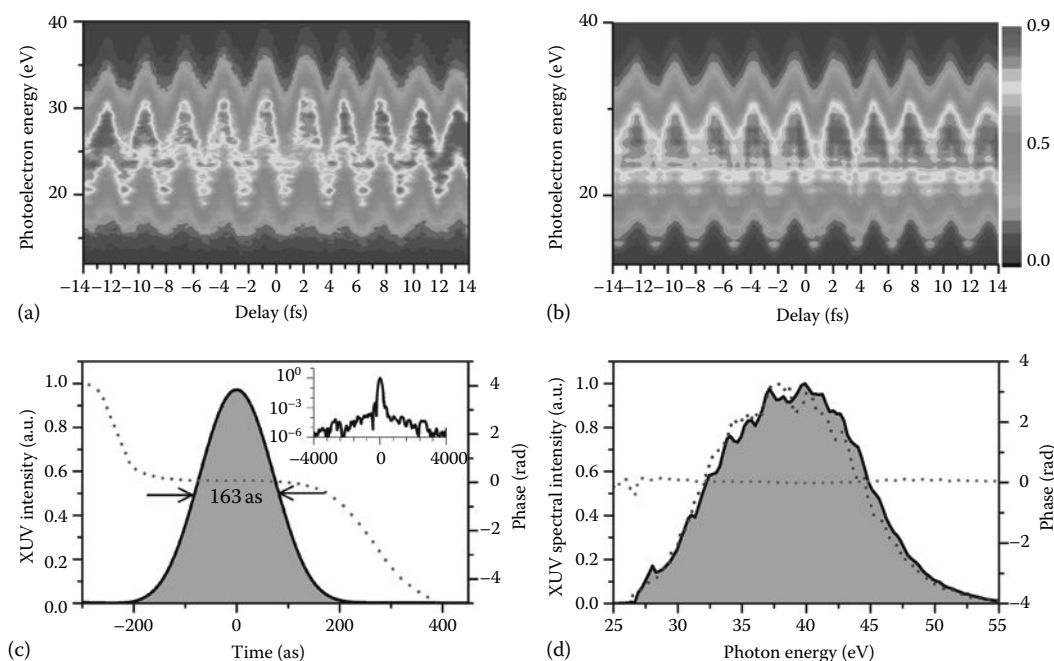


Figure 8.43 The experimentally obtained (a) and retrieved (b) streaked spectrograms of a multi-cycle laser pulse. The temporal profile (solid line) and temporal phase (dashed line) are shown in Figure 8.3. The inset figure shows the same temporal profile, but over an extended temporal range. The pre- and post-pulses located at ± 2600 as are less than 0.1% of the main pulse. (d) The experimental (dotted line) and retrieved (solid line) XUV-only spectrum. The dashed line shows the spectral phase and in this case indicates that the pulse is nearly transform limited. (From Gilbertson, S. et al., *Phys. Rev. A*, 81, 043810, 2010.)

power (TW to PW) Ti:Sapphire CPA lasers directly, which allows the scaling of isolated attosecond pulses to the energy level.

8.7 XUV Pulses with One Atomic Unit of Time Duration and keV X-Ray Pulses

8.7.1 Generation of Pulse with 25 as Duration

The DOG technique works over a wide range of laser pulse duration. With the decrease of the pulse duration, the ground state population of the atoms is also reduced, which allows the use of higher laser intensity, as shown in Figure 8.44.

The higher saturation intensity leads to the extension of cutoff energy as the pulse duration decreases, as shown in Figure 8.45. The corresponding pulse durations supported by the spectra are shown in Figure 8.46. For laser pulses shorter than 10 fs, it is expected that XUV pulses below 20 as will be generated from both neon and helium gases.

The experimentally generated XUV spectrum from neon with DOG is shown in Figure 8.47. The driving laser is 8 fs, centered at 750 nm. The

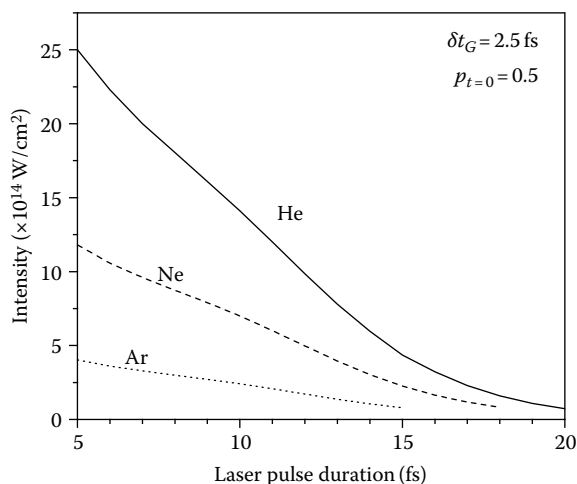


Figure 8.44 The saturation intensity for three types of atoms as a function of laser pulse duration. The ground state population in the center of the gate is 50%.

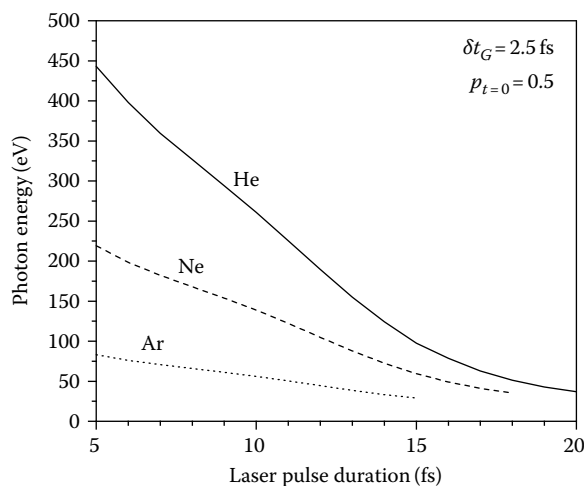


Figure 8.45 The cutoff photon energy for three types of atoms as a function of laser pulse duration. The ground state population in the center of the gate is 50%.

spectrum intensity and shape changes strongly with the CE phase of the NIR laser, indicating that the sub-cycle gating is in action. The continuous spectrum covers the 28–620 eV range, which supports an isolated 16 as pulse that is below one atomic unit of time.

8.7.2 keV Attosecond Pulses

The bandwidth of a 1 as Gaussian pulse is close to 1.9 keV. As discussed in Chapter 4, the cutoff photon energy of high-order harmonics from

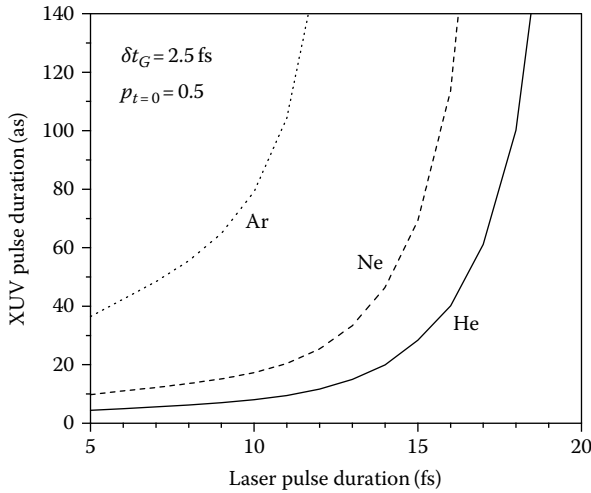


Figure 8.46 The XUV pulse duration for three types of atoms as a function of laser pulse duration. The ground state population in the center of the gate is 50%.

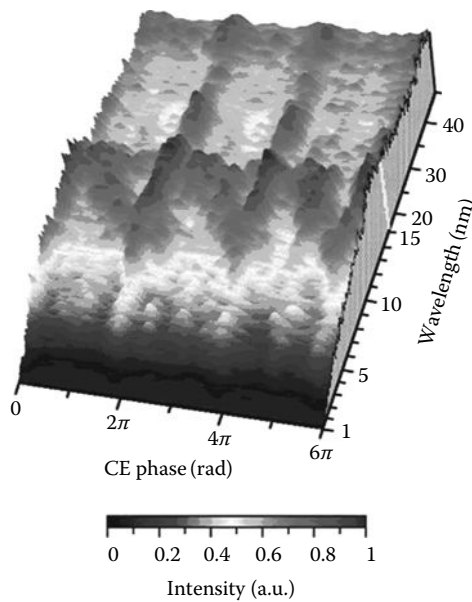


Figure 8.47 CE phase dependence of the XUV spectrum generated from neon with DOG, using 8 fs lasers centered at 750 nm. (From Mashiko, H. et al., *Opt. Lett.*, 34, 3337, 2009.)

helium driven by mid-infrared (mid-IR) lasers can reach a few keV, because the cutoff energy is proportional to the square of the driving laser wavelength. However, the harmonic spectrum from linearly polarized multi-cycle IR lasers mentioned in Chapter 4 corresponds to an attosecond pulse train. By combining amplitude gating and polarization

gating, which is another type of DOG, it is possible to generate isolated keV soft x-ray pulses with mid-IR lasers. When the laser pulse width and center wavelength are chosen properly, it is even feasible to generate isolated coherent pulses less than 1 as, or zeptosecond pulses.

As an example, we consider using 10 fs laser pulses centered at 1.6 μm to perform the polarization gating. In this case, the polarization gate should equal half an optical cycle of the fundamental field, which is 2.7 fs. Choosing the ellipticity of the two counter-rotating fields to be $\varepsilon = 0.5$ and the delay between them, T_d , to be one laser cycle, the calculated gate width is 2.7 fs, using the equation $\delta t_G = 0.3 \left(\varepsilon \tau_p^2 / T_d \right)$, which should allow the generation of isolated attosecond pulses with high contrast.

In a single atom simulation, the acceleration of the electron is calculated using the equation introduced in Chapter 5

$$\begin{aligned} \frac{d^2 \vec{r}}{dt^2} = & i \int_0^\tau d\tau \left(\frac{\pi}{\epsilon + \frac{i\tau}{2}} \right)^{\frac{3}{2}} a^*(t) \langle 0 | \nabla V(\vec{r}) | \vec{p}_s(t, \tau) \\ & - \vec{A}_L(t) \rangle e^{iS(\vec{p}_s, t, \tau)} \vec{E}_L(t - \tau) \cdot \vec{d} [\vec{p}_s - \vec{A}_L(t - \tau)] a(t - \tau) + c.c., \end{aligned} \quad (8.98)$$

which takes into account the effects of the Coulomb potential of the atom on the recombination. For the helium atom considered here, we use the approximate expression for the matrix element

$$\langle 0 | \nabla V(\vec{r}) | \vec{v} \rangle = 0.26 \alpha^{\frac{5}{4}} \frac{\vec{v}}{(v^2 + \frac{3}{2} \alpha)^{\frac{3}{4}}}. \quad (8.99)$$

The numerical simulations show that one only needs to set the gate width to a full laser cycle. The added power of amplitude gating strongly suppresses the intensity of the satellite pulse. Since the optical cycle of the 1.6 μm , T_0 , is 5.3 fs, the pulse has only two cycles, which is short

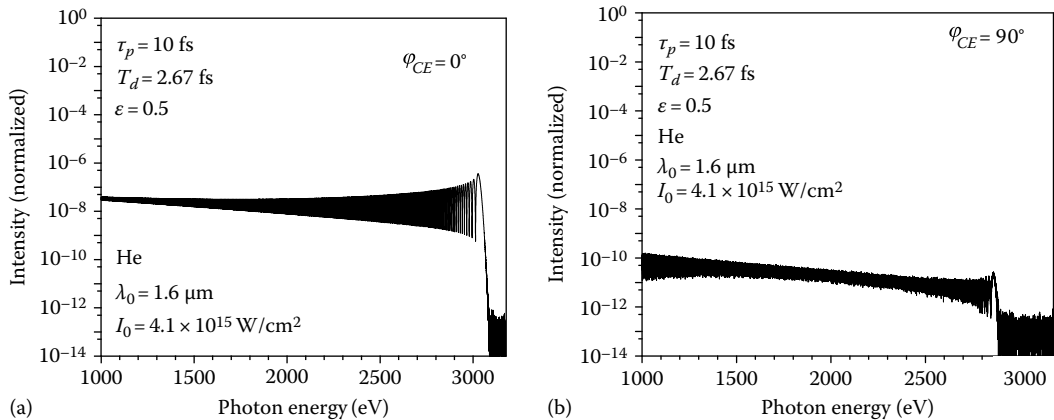


Figure 8.48 The x-ray spectrum generated under two different laser CE phases.

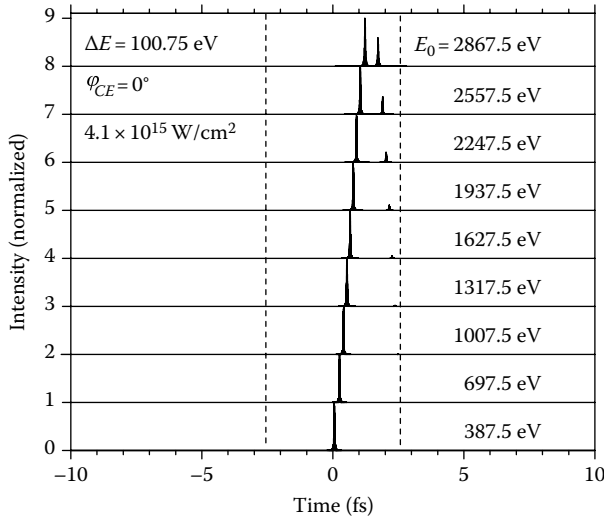


Figure 8.49 The chirp of the attosecond pulse.

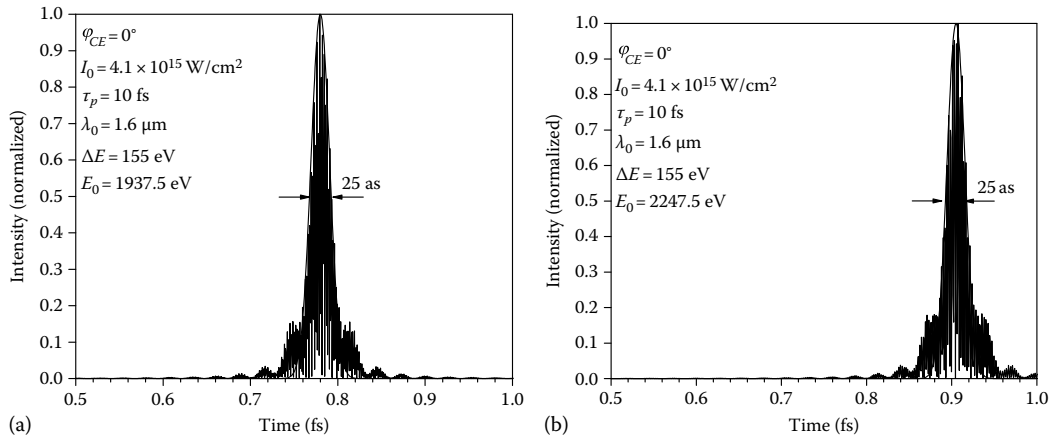


Figure 8.50 Isolated attosecond pulse generation without dispersion compensation.

enough for amplitude gating to be effective. In this case, the delay T_d can be set to half an optical cycle to obtain the required gate width.

For such laser pulses, the ionization saturation intensity of helium is $4.5 \times 10^{15} \text{ W/cm}^2$, which corresponds to a cutout photon energy of 3.4 keV, as shown in Figure 8.48. The spectrum is sufficiently broader to support soft x-ray pulses less than 1 as. The short trajectory of the whole x-ray spectrum is emitted within 0–1.5 fs and the pulse is almost linearly chirped, as shown in Figure 8.49. When a 155 eV width spectrum is filtered out, the pulse duration would be 25 as, as shown in Figure 8.50. It is interesting that no chirped compensation is needed to obtain such extremely short x-ray pulses.

8.8 Summary

Various gating techniques have been developed for generating isolated attosecond pulses. The DOG technique works for both few-cycle and multi-cycle lasers. However, the attosecond pulse energy is still very low (on the order of nanojoules). For performing attosecond pump–probe experiments and for studying nonlinear processes, even higher attosecond pulse energy is needed.

Problems

- 8.1** An XUV Gaussian spectrum supports a 25 as pulse. If the chirp of the pulse is $+1000 \text{ as}^2$, what is the pulse duration? Plot the spectral phase of the chirped pulse.
- 8.2** An XUV Gaussian spectrum supports a 25 as pulse. If the chirp of the pulse is -1000 as^2 , what is the pulse duration? Plot the spectral phase of the chirped pulse.
- 8.3** An XUV Gaussian spectrum supports a 25 as pulse. If the chirp of the pulse is 0 as^2 , what is the pulse duration? Plot the spectral phase of the chirped pulse.
- 8.4** What is the typical conversion efficiency of isolated attosecond pulses using amplitude gating?
- 8.5** What is the typical conversion efficiency of isolated attosecond pulses using polarization gating?
- 8.6** What is the typical conversion efficiency of isolated attosecond pulses using DOG?
- 8.7** In [Figure 8.33](#), suppose that the input pulse is linearly polarized in the vertical (y) direction. The first quartz plate is a multi-order whole-wave plate. Write down the expressions for the electric field components in the vertical and horizontal direction after the plate.
- 8.8** If the first quartz plate in [Figure 8.33](#) is a multi-order half-wave plate, what are the expressions for the electric field components in the vertical and horizontal directions?
- 8.9** A zero-order quarter-wave plate made of quartz is placed right after the first quartz plate in the cases described in Problems 8.7 and 8.8. Its optic axis is oriented vertically. Derive the expressions for the electric field components in the vertical and horizontal directions after the quarter-wave plate.

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Applications of Attosecond Pulses

9.1 Introduction

The ultimate goal of attosecond light-pulse applications is to study electron dynamics in atoms and molecules. Such an experiment would need a strong attosecond pulse to excite the system and another attosecond pulse to probe the evolution of the system. These two kinds of pulses are referred to as the attosecond pump and attosecond probe, respectively.

9.1.1 Attosecond Pump–Probe Experiments

An example of such experiments is proposed by Hu and Collins to study two-electron dynamics in helium. The energy diagram of helium is depicted in the left graph of [Figure 9.1](#). When an intense (6×10^{14} W/cm²), long attosecond pulse (~ 500 as) centered at 21.2 eV pumps a helium atom, it is possible to excite 50% of the ground state ($1s^2\ ^1S^e$) electron to the excited state ($1s2p, 1s3p \dots$) through single-photon absorption. The $1s2s, 1s3s$, and other $1sns$ states are not populated because of the dipole-transition rule. A wave packet is created by mixing the $1s^2\ ^1S^e$ and $1s2p\ ^1P^o$ as well as other excited states. The wave packet in quantum mechanics corresponds to the classical motion of electron in space. In this case, we can imagine the distance of an electron to the nucleus changes with time. It is worth pointing out that an atom in the ground state is a steady state; therefore, it is meaningless to talk about its dynamics.

If another linearly polarized attosecond pulse (the probe pulse) centered at 100 eV doubly ionizes the helium atom in the mixed state as illustrated in the right graph of [Figure 9.1](#), the ionization probability depends on the time delay between the pump and the probe pulses. The probability is higher when the two electrons are located near each other, as depicted in [Figure 9.2](#). Experimentally, one can measure the doubly ionized helium ion (He^{2+}) signal as function of time delay between the probe pulse and the pump pulse. The He^{2+} yield as a function of the time delay would inform us how the electron separation changes with time. Hu and Collins's simulation shows that the separation changes periodically with a 2 fs cycle, which can be observed with the attosecond pulses. The cycle is much larger than the ~ 200 as corresponding to the energy difference between the ground state and the first excited state ($1s2p$) because other excited states are involved.

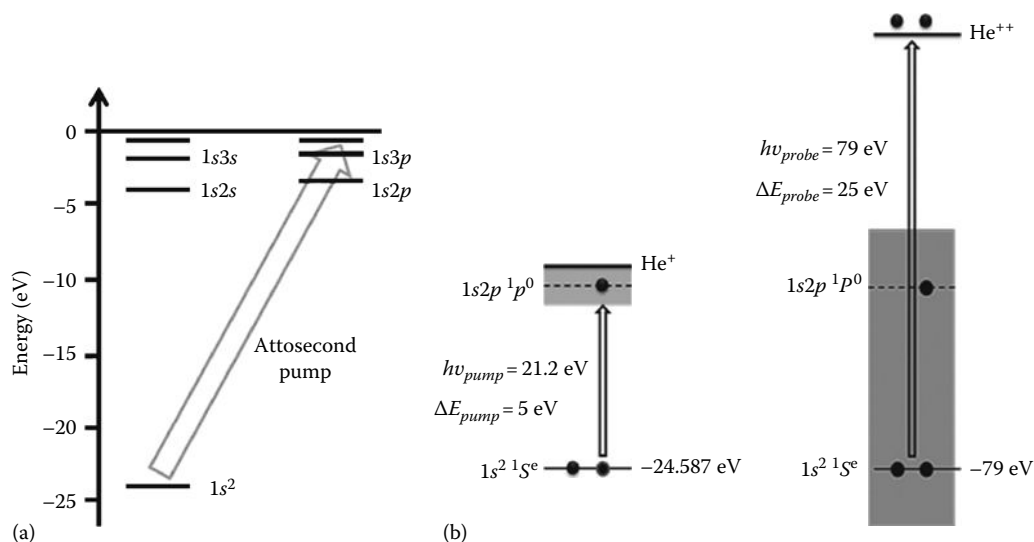


Figure 9.1 Attosecond pump–probe scheme for studying electron dynamics in helium. The energy diagram of singly excited helium is shown on the left.

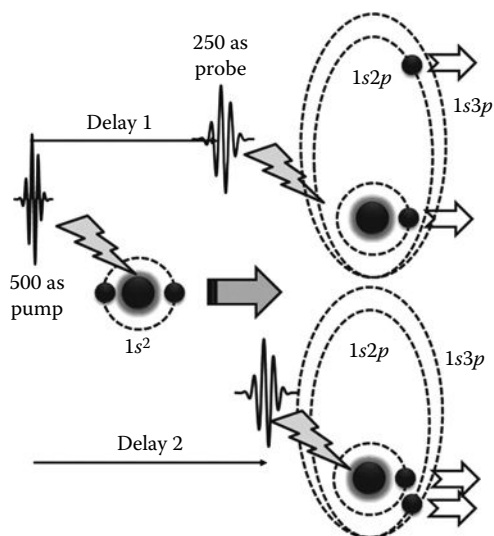


Figure 9.2 Probing the attosecond dynamics in helium. The double ionization probability is higher when the two electrons are close to each other.

Suppose the electric fields of the two attosecond pulses are oriented horizontally as depicted in Figure 9.3a. If the first electron is released vertically in the upward direction ($\theta_1 = 90^\circ$), then the second electron should leave the atom in the downward direction ($\theta_2 \approx 270^\circ$), as a result of the conservation of linear momentum. Figure 9.3b shows the cross sections of the two-electron ejection process for different time delays in attosecond pump–probes. The cross section depends on the second

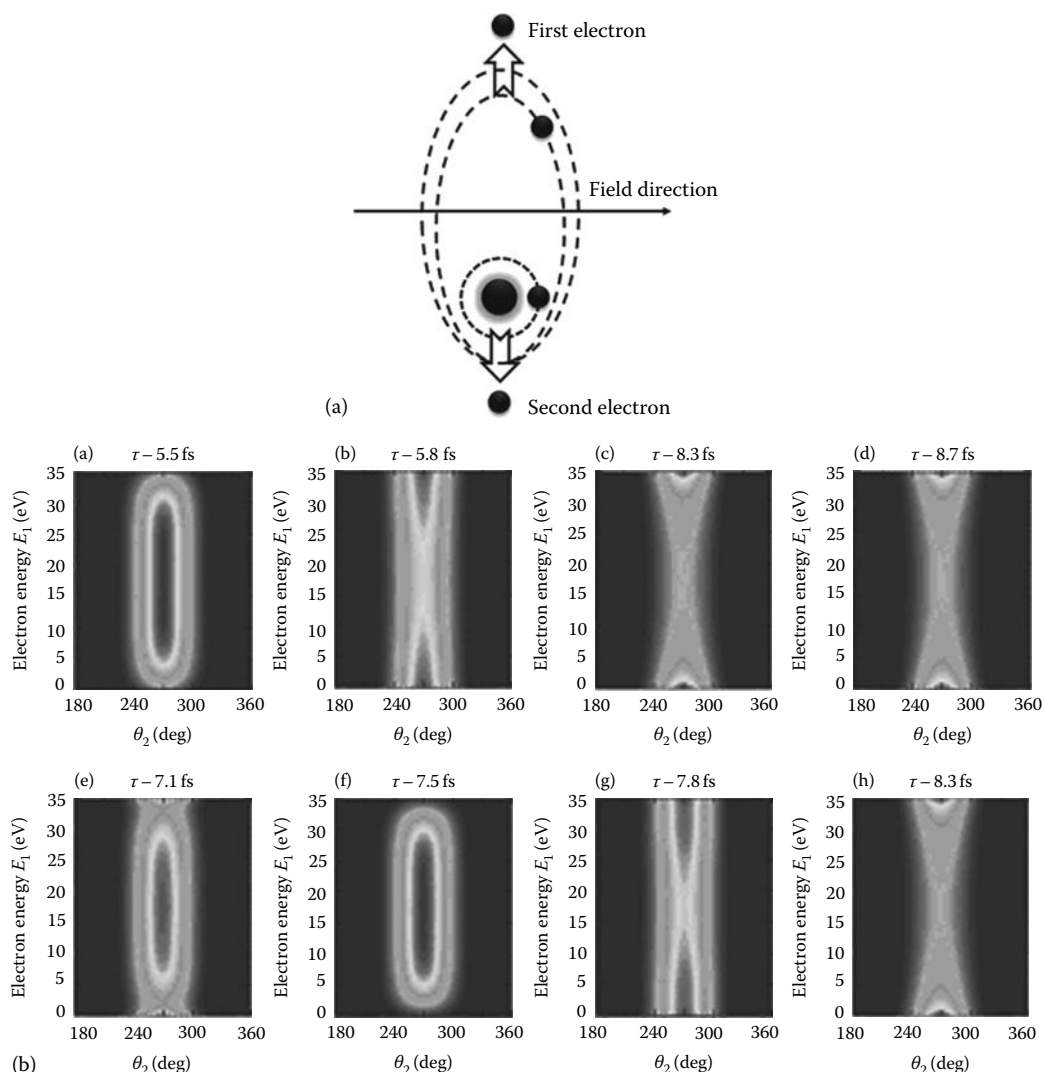


Figure 9.3 (a) Directions of attosecond field and electrons. (b) The contour plots for cross sections of helium double ionization under the attosecond pump-probe scheme. (Reprinted with permission from S.X. Hu and L.A. Collins, *Phys. Rev. Lett.*, 96, 073004, 2006. Copyright 2006 by the American Physical Society.)

electron energy and its ejection angle when the first electron ejection angle is perpendicular to the attosecond pulse-polarization axis. In this process, the two electrons share the total energy of the pump and probe photons. Different features of energy sharing between the two back-to-back ejected electrons are clearly seen at different delay time.

Measuring either the energy sharing of the two ionized electrons or the double ionization probability of the mixed states as a function of the time delay determines the separation of the two electrons in space at the instant of the probe. Consequently, following one of these quantities in time displays the internal motion of the excited electron.

9.1.2 Requirement on the Attosecond Pulse Energy

To perform such an attosecond pump–probe experiment, one would need a very strong attosecond pulse field (6×10^{14} W/cm²) centered at 21.2 eV. Otherwise, the 50% population on the $1s2p$ state cannot be reached. If that is the case, the double ionization signal would mostly come from the ground state, which would make it difficult to observe anything from the mixed states.

Such a measurement has yet to be demonstrated because the required pump intensity is not easy to reach. As discussed in Chapter 1, the intensity of a Gaussian beam and Gaussian pulse can be calculated by

$$I_0 = \frac{1.88\varepsilon}{\pi w^2 \tau}, \quad (9.1)$$

where

ε is the pulse energy

w is the radius of the focal spot

τ is the pulse duration

When a 500 as pulse is focused to $w = 5 \mu\text{m}$, the required pulse energy is 130 nJ. Experimentalists are still working on generating such energetic EUV pulses. Alternatively, reducing the focal spot size to $w = 1 \mu\text{m}$ is possible. In that case, the required energy is 5 nJ. It should be feasible to produce 5 nJ, <500 as isolated attosecond pulse centered around 20 eV in Xeon gas using the double optical gating method. The pulse spectrum should not reach the ionization potential of the ground state (24.6 eV), otherwise it would produce lots of free electrons from the ground states. Extending the spectrum to low photon energy is preferred.

In many cases, intense attosecond XUV probe pulses are also required. For example, if the probe in the experiments depicted in Figure 9.2 is not strong enough, then the images shown in Figure 9.3 will be difficult to obtain because of the low electron counts.

Many attosecond physics experiments conducted so far used attosecond XUV pulses and femtosecond near infrared (NIR) pulses as the pump and probe, very similar to the scheme used in the attosecond streak camera. This chapter gives two examples of attosecond applications in optics and another example in atomic physics studies.

9.2 Direct Measurement of the Temporal Oscillation of Light

Maxwell's classical theory reveals that electromagnetic waves propagate with the speed that is the same as that of light. He postulated that light is an electromagnetic wave in a certain wavelength range which our eye can see. For example, visible light covers the 400–700 nm range. His wave theory has been successfully applied to solve the problems of propagation of light, such as diffraction and interference. Quantum theory is needed when light–atom interaction is involved, such as in light emission and absorption. The attosecond optical pulses provided a unique tool for

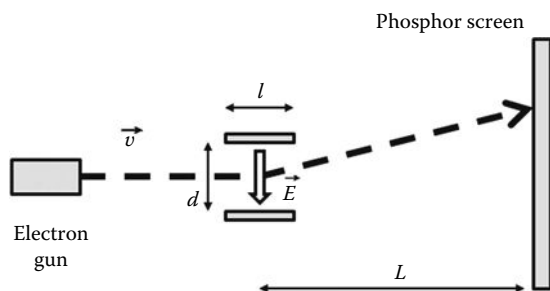


Figure 9.4 The principle behind analog oscilloscopes.

observing the very fast oscillation of the light wave in the way like we see the time variation of slowly varying electric signals with oscilloscopes.

9.2.1 Direct Measurement of Low-Frequency Electric Field

In the framework of classical electrodynamics, the electric field, $\vec{E}$, and the magnetic field, $\vec{B}$, are defined by the force a point charge q experiences,

$$\vec{F} = q(\vec{E} + \vec{v} \times \vec{B}), \quad (9.2)$$

where $\vec{v}$ is the velocity of the charge. To measure the field strength and direction directly, one has to measure the force vector. This can be done easily when the field changes slowly with time. In fact, it is the basic principle of analog oscilloscopes, as shown in Figure 9.4.

In such a device, electrons emitted from an electron gun are deflected by the electric field created by applying the voltage to be measured on a pair of parallel metal plates. For the given velocity of the electron from an electron gun, $\vec{v}$, the spacing, d , and the length of the plates, l , as well as the distance of the plates to the phosphor screen, L , the voltage (and thus the field strength between the two metal plates) can be determined by the position of the electron on the screen. Another pair of deflection plates with a field perpendicular to the previously mentioned pair is applied with a voltage that changes linearly with time to scan the electrons in the horizontal direction (the time axis). The combination of the two pairs of plates plots the voltage oscillation on the screen. The response time of the deflection plates are related to the transit time of the electron passing through the field created by the plates, as well as the capacitance of the plates. Limited by the bandwidth of the deflection plates, this scheme works for up to ~ 10 GHz in frequency, which is far below the frequency of the light field (100 THz). Thus, it is too slow to see the electric field oscillation of light.

9.2.2 Direct Measurement of Light-Field Oscillation

All light detectors before the year 2001 measured the number of electrons generated by light that is proportional to the light intensity. These detectors

are based on the quantum theory of the light. When the intensity is low, the number of electrons is proportional to the number of photons irradiated on the detector's surface. Intensity is a physical quantity that averaged over one wave period, thus one cannot directly see light-field oscillation within an optical cycle using these types of detectors.

9.2.2.1 Definition of Electric Field

In classical electrodynamics, the electric field is defined as the force exerted on a point charge of unit value, i.e.,

$$\vec{E} = \frac{\vec{F}}{q}. \quad (9.3)$$

Therefore, the direct measurement of the light field must rely on the measurement of this force.

It is known that light wave contains both electric and magnetic fields. At a given spatial point, the electric and magnetic field of a linearly polarized monochromatic laser with angular frequency ω can be expressed as

$$\vec{E}(t) = \vec{E}_0 \cos(\omega t) \quad (9.4)$$

and

$$\vec{B}(t) = \vec{B}_0 \cos(\omega t), \quad (9.5)$$

respectively. In the expressions, E_0 is the amplitude of the electric field while the amplitude of the magnetic field is $B_0 = E_0/c$ (c is the speed of light in vacuum). When an electron with velocity $\vec{v}$ is placed in the laser field, the force is also a time-dependent quantity

$$\vec{F}(t) = -e(\vec{E}_0 + \vec{v} \times \vec{B}_0) \cos(\omega t). \quad (9.6)$$

The magnitude of force from the electric and magnetic field is eE_0 and eE_0v/c , respectively. When the speed of the electron is much lower than the speed of light, the contribution from the electric field is much larger than that from the magnetic field. That is, in the nonrelativistic case, the light field can be determined by just measuring the electric field force

$$F(t) \approx -eE_0 \cos(\omega t). \quad (9.7)$$

For visible light, $\omega/2\pi \approx 10^{14}$ Hz, i.e., the electromagnetic field of visible light performs 10^{14} oscillations per second. To measure the force directly, the point charge must be placed in the light field with a precision on the order of a fraction of an optical cycle T_0 (2.6 fs for lasers centered at 800 nm). This became possible with the availability of the attosecond pulses in 2001. The attosecond XUV pulse can deliver probing electrons in the light field with attosecond precision through photoemission in gases. When the electrons are positioned within a time interval $\delta t_{probe} \ll T_0$, the temporal variation of the force can be “frozen.”

9.2.2.2 Definition of Force

In Newton's mechanics, force is defined in terms of acceleration it gives to a body with a certain mass. Therefore, force can be determined by measuring the acceleration. In [Figure 9.4](#), the vertical shift of the electron

on the screen is related to the acceleration of the electron in the deflection field. The acceleration, a , of an electron is its momentum change rate

$$a = m_e \frac{dv}{dt} = \frac{dp}{dt}. \quad (9.8)$$

It turns out that in the measurement of the electric force of light field, it is easier to determine the acceleration by measuring the momentum change.

Suppose at time t_0 , an electron is placed at the location r in a pulsed linearly polarized laser field propagating along the z direction. Experimentally, the final momentum of the electron is always measured when the laser pulse vanishes ($t = \infty$). The change of the electrons' momentum along the direction of the electric field is given by

$$\begin{aligned} \Delta p(r, t_0) &= p(r, t)|_{t=\infty} - p_0 = -e \int_{t_0}^{t=\infty} \mathcal{E}(r, t') dt' \\ &= e[\mathcal{A}(r, t = \infty) - \mathcal{A}(r, t_0)] = -e\mathcal{A}(r, t_0). \end{aligned} \quad (9.9)$$

where $p(r, t)$ and p_0 are the momentum at time t and t_0 , respectively.

Equation 9.9 can be rewritten as

$$\mathcal{A}(r, t_0) = -\frac{1}{e} \Delta p(r, t_0). \quad (9.10)$$

Varying the timing of such an electron probe, t_0 , across the laser pulse provides complete information on the vector potential light wave. Once $\mathcal{A}(r, t)$ is known, one can obtain $\mathcal{E}(r, t) = -\partial \mathcal{A}(r, t) / \partial t$.

9.2.2.3 The Retarded Frame

In the laboratory frame, the electric field of a pulse laser field is expressed as

$$\mathcal{E}(r, t) = E_0 \left(r, t - \frac{z}{v_g} \right) \cos(\omega_0 t - kz), \quad (9.11)$$

where

- E_0 is the field envelope
- ω_0 is the carrier frequency
- and k is the wave vector

The time in a retarded frame, t_{ret} , is related to the real time, t , by

$$t_{ret} = t - \frac{z}{v_g}, \quad (9.12)$$

where v_g is the group velocity of the pulse. Obviously, the retarded form moves in space with the group velocity of the pulse.

In this frame, the laser field is expressed by

$$\mathcal{E}(r, t_{ret}) = E_0(r, t_{ret}) \cos \left[\omega_0 \left(t - \frac{z}{\omega_0/k} \right) \right]. \quad (9.13)$$

When the phase velocity ω_0/k equals the group velocity, then

$$\mathcal{E}(r, t_{ret}) = E_0(r, t_{ret}) \cos(\omega_0 t_{ret}). \quad (9.14)$$

In this case, the field in the retarded frame does not change with the position z . If the attosecond XUV beam that generates the probing electron co-propagates with the laser beam, and if the two beams have the same group velocity, then the electrons releasing time, t_0 , is locked to the peak of the laser pulse no matter at which location the electron is emitted. This allows the usage of a gas target size much larger than the laser wavelength for converting the attosecond XUV pulse into probing electrons.

In a focused laser beam, the phase and group velocities are different in the confocal region, which is the origin of the Gouy phase shift. Therefore, the size of the gas target should be much smaller than the Rayleigh range

$$z_R = \frac{\pi w_0^2}{\lambda_0}, \quad (9.15)$$

where

w_0 is the radius of the focal spot

λ_0 is the center wavelength of the laser pulse

9.2.2.4 Measurement Demonstration

The first experiment that directly mapping out the NIR laser field oscillation was done by Goulielmakis et al. The setup they used is almost identical to the attosecond streak camera discussed in Chapter 8 for characterizing isolated attosecond pulses. The 250 as XUV pulses centered at 93 eV were generated in neon gas by the amplitude gating method described in Chapter 8. The driving laser is CE phase stabilized. It produces sub-5 fs pulses with 0.4 mJ energy centered at 750 nm. The same laser field is to be directly measured. They ignored the transverse variation of the field strength by assuming the laser field $\varepsilon(r, t) = \varepsilon(0, t)$.

The XUV pulse co-propagates with the laser pulse collinearly to a second Neon target placed in the focus of a spherical, two-component, Mo/Si multilayer mirror. The focal lengths of the two mirrors are identical, which is 120 mm. The XUV beam is focused by the inner mirror to the second Neon gas target whereas the NIR beam is focused by the outer annular mirror to the same target with a $\sim 50 \mu\text{m}$ size diameter. The time delay between the laser pulse and the XUV probe was varied in steps of 200 as to scan cross the whole pulse. The time step is more than 10 times smaller than the laser-wave cycle (2.5 fs).

The electron spectrometer used in the experiments is a time-of-flight one described in Chapter 7. It measures the electrons emitted in a small solid angle. In this case, it is more convenient to use energy instead of momentum. The field-induced variation of the final energy spectrum of the probe electrons versus delay between the XUV burst and the laser pulse is shown in Figure 9.5. $\varepsilon(t)$ can be obtained using the FROG-CRAB method discussed in Chapter 7, or alternatively, be directly obtained using Equation 9.10, which is the method used by the Goulielmakis et al. The result is shown in Figure 9.6.

The laser pulse was also been measured by a second harmonic autocorrelator described in Chapter 2. It gives a pulse duration of 4.3 fs. However, it cannot see the oscillation directly. Nevertheless, the pulse duration agrees with the direct method. The direct method also gives the

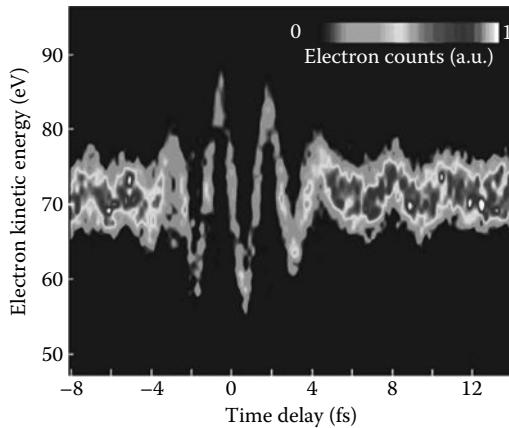


Figure 9.5 The streaked electron spectrum as a function of delay between the XUV and the laser pulses. (From E. Goulielmakis, M. Uiberacker, R. Kienberger, A. Baltuska, V. Yakovlev, A. Scrinzi, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, Direct measurement of light waves, *Science*, 305, 1267, 2004. Reprinted with permission of AAAS.)

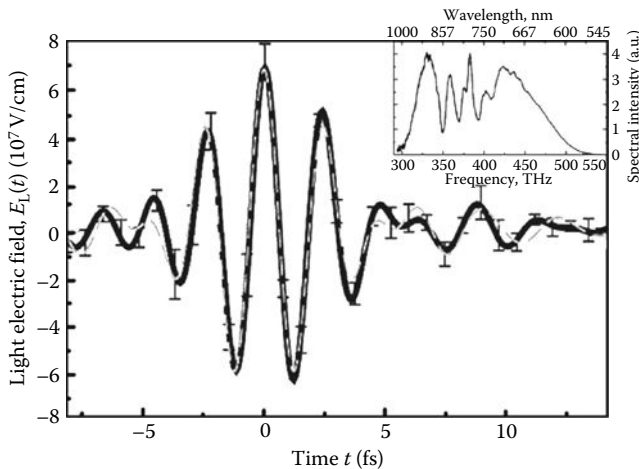


Figure 9.6 Electric field of the few-cycle laser pulse measured by attosecond streaking. The optical spectrum of the laser pulse is shown in the insert. (From E. Goulielmakis, M. Uiberacker, R. Kienberger, A. Baltuska, V. Yakovlev, A. Scrinzi, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, Direct measurement of light waves, *Science*, 305, 1267, 2004. Reprinted with permission of AAAS.)

peak field strength from the maximum energy shift, which is $\sim 7 \times 10^7$ V/cm. With the temporal evolution, strength, and direction of $\mathcal{E}(t)$ measured, a complete characterization of a light pulse in terms of its classical electric field was performed, which is important for measuring laser pulses approaching a single cycle where the conventional optical methods such as FROG become less accurate.

9.3 Direct Measurement of Spatial Variation of Field in Bessel Beams*

9.3.1 Bessel Beam

In Chapter 7, we showed that spatial distribution at the focal point of a laser beam truncated by an annular aperture is a Bessel function. The distribution does not change much in the propagation direction just before and after the focus. In fact, this is a special case of solutions to the electromagnetic wave equation that are invariant in the plane transverse to propagation. Such solutions, known as Bessel beams, have been called “diffraction-free.”

For an input beam with finite transverse size, the transverse field profile invariance of the focused beam allows for useful applications, such as optical tweezers, electron acceleration, and harmonic generation. Although obtaining an image of the intensity distribution is trivial, it is not easy to measure the direction and strength of the electric field as a function of the transverse coordinate directly. Interestingly, attosecond pulses make it possible to directly observe the electric field in the transverse plane of a Bessel optical beam.

9.3.1.1 Electric Field of an Ideal Bessel Beam

In cylindrical coordinates, the electric field of a pulsed, linearly polarized Bessel beam with center frequency ω_0 that propagates in the z direction is expressed as

$$\varepsilon(r, z, t) = E_0 f(t) J_0(k_r r) \cos(k_z z - \omega_0 t), \quad (9.16)$$

where

E_0 is the amplitude at the center of the beam

$f(t)$ is the envelope function that specifies the shape of the pulse

J_0 is the zeroth-order Bessel function of the first kind

k_r and k_z are the propagation constant components in the radial, r , and z directions, respectively, such that $\sqrt{k_r^2 + k_z^2} = k = \omega_0/c$.

Experimentally, the zeroth-order Bessel beam can be produced by sending a collimated laser beam to a ring slit. Approximations to Bessel beams are made in practice by focusing a Gaussian beam with an axicon lens to generate a Bessel–Gauss beam.

At a given time, t , and position, z , the electric field distribution is

$$\varepsilon(r) \propto J_0(r). \quad (9.17)$$

Generally, all beams with Bessel-like features arising from Equation 9.16 exhibit radial intensity distributions with concentric fringes. The field direction changes periodically along the transverse direction, which is expressed by the sign change of the zero-order Bessel function of the first kind, $J_0(r)$,

* The text of this section is adapted from the publication: Gilbertson, S., Feng, X., Khan, S., Chini, M., Wang, H., Mashiko, H., and Chang, Z., Direct measurement of an electric field in femtosecond Bessel-Gaussian beams, *Opt. Lett.* 34, 2390 (2009). With permission of Optical Society of America.

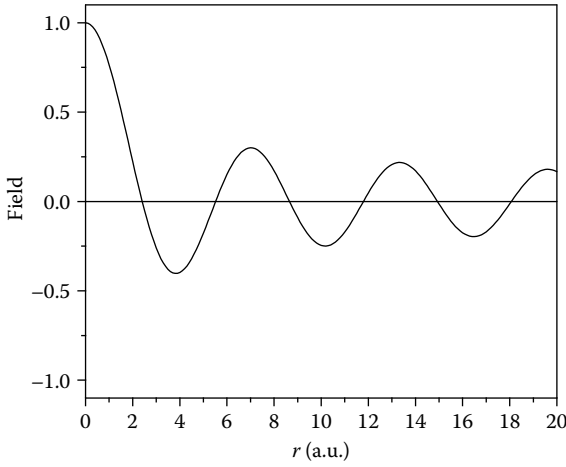


Figure 9.7 Electric-field distribution in the radial direction.

as shown in Figure 9.7. For linearly polarized field, the direction of the field can be specified by the sign of a scalar function, like in Equations 9.16 and 9.17. Here, we are interested in the field direction amplitude change caused by the Bessel function term, not by the cosine term.

9.3.1.2 Field in Experimental Setup

As discussed in Chapter 7, in the attosecond streaking experiments, the field near the focus can be expressed as

$$E_{GB}(r, z) = E_0 e^{i(kz + \frac{k}{2z}r^2)} \left(-i \frac{1}{\lambda_0 z} \right) \int_{\rho_i}^{\rho_o} d\rho \rho J_0 \left(k\rho \frac{r}{z} \right) e^{-ik\frac{\rho^2}{2q(z)}}, \quad (9.18)$$

where λ_0 is the center wavelength. The quantity

$$\frac{1}{\tilde{q}(z)} \equiv \frac{1}{z} - i \frac{\lambda_0}{\pi w_G^2}, \quad (9.19)$$

where

w_G is the radius of the input Gaussian laser beam

ρ_i and ρ_o are the inner and outer radius of an annular aperture, respectively

If $\rho_o - \rho_i \ll w_G$ is satisfied,

$$E_{GB}(r, z) \approx \left(-\frac{i}{2\pi} \right) E_0 e^{i(kz + \frac{k}{2z}r^2)} e^{-i\frac{k}{2q(z)}(\frac{\rho_o + \rho_i}{2})^2} \frac{k}{2z} \\ \times \left[\rho_o^2 \frac{2J_1\left(\frac{k\rho_o r}{z}\right)}{\frac{k\rho_o r}{z}} - \rho_i^2 \frac{2J_1\left(\frac{k\rho_i r}{z}\right)}{\frac{k\rho_i r}{z}} \right]. \quad (9.20)$$

For $r = 0$, $J_0(k\rho[r/z]) = 1$, we have

$$\begin{aligned} E_{GB}(0, z) &= E_0 e^{i(kz + \frac{k}{2z}r^2)} \left(-i \frac{1}{\lambda z} \right) \int_{\rho_i}^{\rho_o} d\rho \rho e^{-ik\frac{\rho^2}{2q(z)}} \\ &= E_0 e^{i(kz + \frac{k}{2z}r^2)} \left(-i \frac{1}{\lambda z} \right) \frac{2\tilde{q}(z)}{k} \left(e^{-ik\frac{\rho_o^2}{2q(z)}} - e^{-ik\frac{\rho_i^2}{2q(z)}} \right). \end{aligned} \quad (9.21)$$

In general, it turns out that an approximation of the field expression near the focus is

$$\begin{aligned} E_{GB}(r, z) &\approx -i \frac{E_0}{2\pi} e^{i(kz + \frac{k}{2z}r^2)} \frac{\tilde{q}(z)}{z} \left[e^{-ik\frac{\rho_o^2}{2q(z)}} - e^{-ik\frac{\rho_i^2}{2q(z)}} \right] \\ &\quad \times \left[\rho_o^2 \frac{2J_1\left(\frac{k\rho_o r}{z}\right)}{\frac{k\rho_o r}{z}} - \rho_i^2 \frac{2J_1\left(\frac{k\rho_i r}{z}\right)}{\frac{k\rho_i r}{z}} \right]. \end{aligned} \quad (9.22)$$

As an example, we choose $\lambda_0 = 750$ nm, $w_G = 15$ mm, $f = 250$ mm. The transverse field distributions for several combinations of ρ_i and ρ_o are shown in Figures 9.8 and 9.9. In attosecond streak camera experiments, the XUV spot needs to be overlapped with the center spot of the Bessel beam. It is desirable to have a large center spot so that the overlapping can be accomplished easily. It can be seen from Figure 9.8 that the width of the center spot is larger when ρ_o is reduced, which is due to the diffraction effect. When both ρ_i and ρ_o are decreased, the center spot is even larger, as shown in Figure 9.9.

9.3.2 Measurement Scheme

The electric-field vector changes direction periodically along the transverse direction, which is expressed by the sign change of the Bessel function between adjacent intensity rings. In principle, the change of field direction can be measured by interferometric methods since the

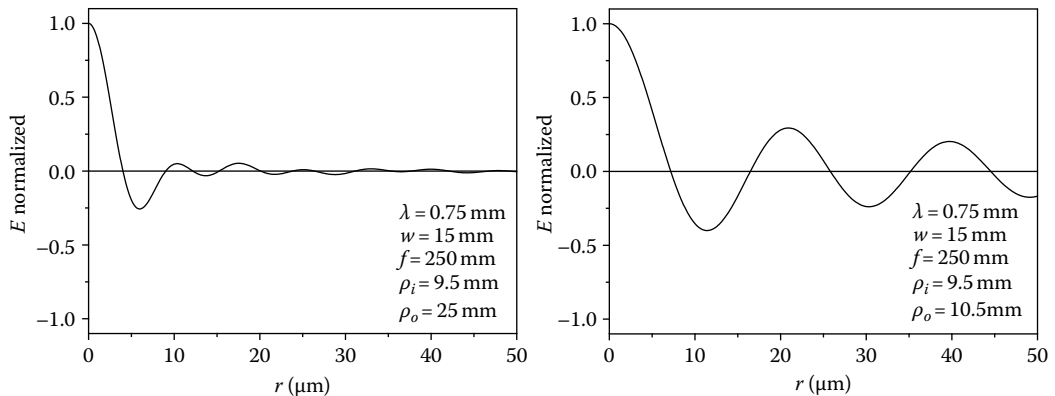


Figure 9.8 Electric-field distributions in the radial direction. The width of the center spot is large for smaller ρ_o .

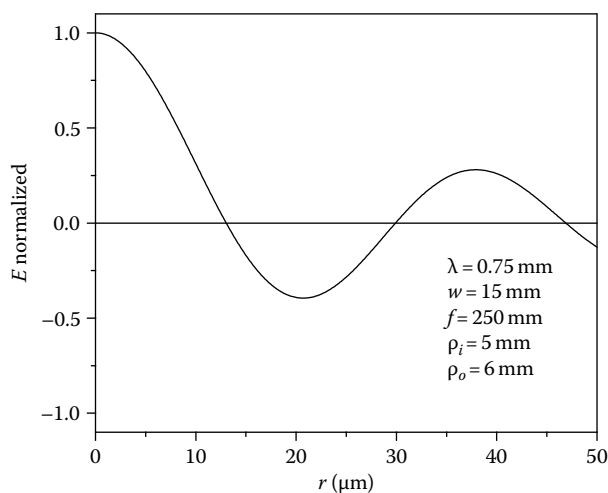


Figure 9.9 Electric-field distributions in the radial direction with a large center spot.

direction reversal can be treated as a π phase shift of the light wave. However, such a phase shift also exists in scalar-field Bessel beams (such as sound waves); thus, the measurement is indirect. The change of field direction with r has never been observed in the past for light beams using the first principle of classical electrodynamics, which defines the electric field as the force exerted on a point charge of unit value. Although measurements have been conducted in the microwave region, there are no such measurements with optical frequencies.

The direct measurement of the field in the Bessel beam means determination of the electric force with a probe charge. However, the temporal response of the required force meter should be on the order of attoseconds because the force changes on a femtosecond scale as expressed by the cosine term in Equation 9.16. An attosecond force meter can be constructed where a probe electron is placed in the laser field within a time interval much less than the light wave period to avoid the effects of the cosine term. The force experienced by the probe charge can be measured by its momentum change, as discussed in [Section 9.2](#).

The NIR-field amplitude and direction change can be measured with attosecond streaking. A localized noble gas target is placed in the Bessel beam and an XUV attosecond pulse synchronized to the laser field that creates the Bessel beam is focused to a spot much smaller than the period in the r direction. The photoelectron generated by the XUV pulse in the gas target is streaked by the electric field of the Bessel beam. When the XUV beam scans across the Bessel beam, the momentum shift should change periodically. The scanning scheme is shown in [Figure 9.10](#). At a fixed delay between the attosecond pulse and the laser field, when the XUV beam is scanned across the Bessel rings, the streaked photoelectron spectrum is a function of the radial position.

According to Newton's second law, the time derivative of the momentum, p , of the probe electron is proportional to the force and thus the

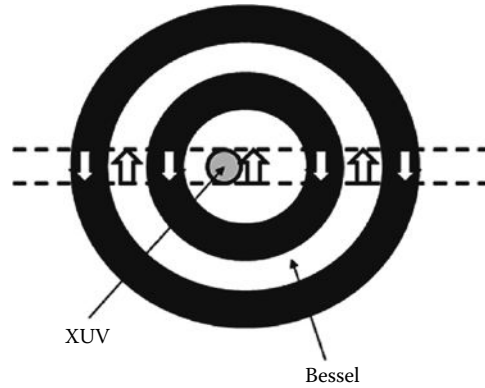


Figure 9.10 XUV focal spot scans horizontally through the region between the two dashed lines in the Bessel beam. The field directions in the Bessel rings are indicated by the arrows.

electric field of the Bessel beam. The measured momentum value of a probing electron placed at point r is

$$p(r)|_{t=\infty} = p_0 - e \int_{t_0}^{\infty} \varepsilon(r, t') dt'$$

$$\approx p_0 + \frac{e}{\omega_0} E_0 J_0(k_r r) f(t_0) \sin(\omega_0 t_0). \quad (9.23)$$

Here, p_0 is its field-free momentum, and t_0 is the time that the probe electron is placed in the field. Equation 9.23 indicates that the electric-field variation in the transverse plane, $E_0 J_0(k_r r)$, can be determined by the measurement of the momentum of an electron placed at various r positions while keeping t_0 constant. This can be accomplished by scanning an XUV beam across the Bessel light beam. The z dependence is dropped in Equation 9.23 when the laser and XUV beam co-propagate.

9.3.2.1 Experimental Demonstration

The setup uses a Mach–Zehnder configuration, which is illustrated in [Figure 9.11](#). The attosecond burst of electrons for probing the electric force is placed in the NIR Bessel beam through photoemission from Argon atoms released by the second gas jet (GJ2) with 276 as XUV pulses.

The laser system is a carrier–envelope phase stabilized chirped pulse amplifier followed by a hollow-core fiber compressor, producing 1 mJ, 8 fs pulses centered at 780 nm at a 1.5 kHz repetition rate. The input near infrared laser is split into two-beams by a beam splitter (BS). The transmitted beam passes through the DOG optics and is focused to an Argon gas target (GJ1) for the attosecond pulse generation. The DOG field is created by using two birefringent quartz plates (QP1 and QP2) and a thin BBO crystal, as discussed in Chapter 8. The XUV attosecond beam is focused by a Mo/Si multilayer mirror to the gas nozzle with 50 μm inner diameter. The energy spectrum of the photoelectron is detected by a TOF spectrometer.

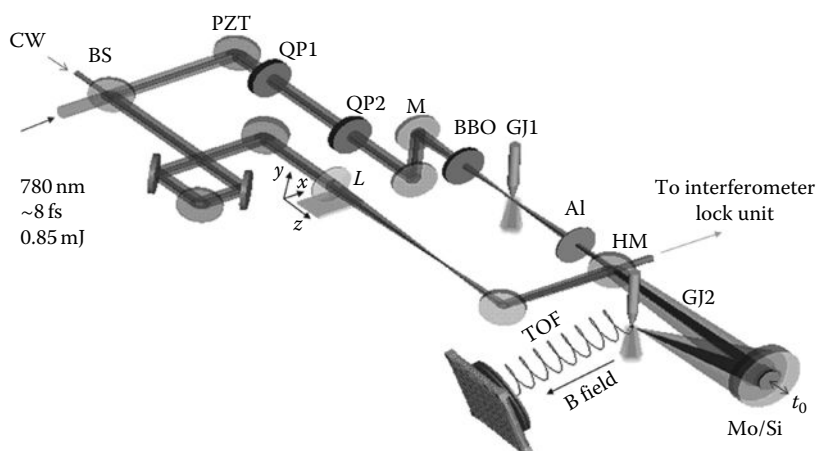


Figure 9.11 Schematic of the attosecond streak camera. BS is a beamsplitter, PZT is a piezoelectric transducer, QP1 and QP2 are quartz plates, M is a spherical mirror, GJ1 and GJ2 are the gas targets, Al is an aluminum filter, HM is the hole drilled mirror, and L is a lens. (Reprinted with permission from S. Gilbertson, X. Feng, S. Khan, M. Chini, H. Wang, H. Mashiko, and Z. Chang, *Opt. Lett.*, 34, 2390, 2009. Copyright 2009 by the American Physical Society.)

Meanwhile, the reflected portion of the beam remains linearly polarized and is used for streaking. It is reflected off a hole-drilled mirror (HM) and is focused by an annular mirror on the gas released by GJ2. The focused NIR beam is a Bessel beam. The XUV and NIR beam after the hole-mirror propagate collinearly. The photoelectrons generated by the XUV pulse are given a momentum kick by the NIR field. The inner Mo/Si mirror is movable with a PZT allowing delay between the XUV and the streaking field to be precisely controlled.

The same setup has been used to characterize the attosecond pulse in situ and for mapping out the field in a Bessel–Gaussian beam. For the former, the XUV spot is placed at the center of the NIR Bessel beam. The attosecond XUV pulses are characterized with the Complete Reconstruction of Attosecond Burst (CRAB) method based on attosecond streaking. By recording the kinetic energy spectrum of the emitted photoelectrons as a function of delay with a position-sensitive time-of-flight (TOF) detector, a streaked spectrogram was created. Using a blind iterative algorithm, PCGPA, the attosecond pulse could be reconstructed from the spectrogram. Figure 9.12a and b shows the measured and reconstructed CRAB traces. The temporal profile shown in Figure 9.12c indicates the pulse duration to be 276 as. The validity of the measurement was confirmed by the excellent frequency marginal comparison shown in Figure 9.12d, which is a comparison of the unstreaked spectrum (dashed line) and the reconstructed spectrum (solid line). The duration of the XUV pulse is one-tenth of the fundamental optical cycle (~ 2.6 fs) and is therefore short enough for generating the probe electrons.

Due to the annular aperture effect from the gold coated annular focusing mirror, the spatial profile of the streaking NIR beam near the focus is

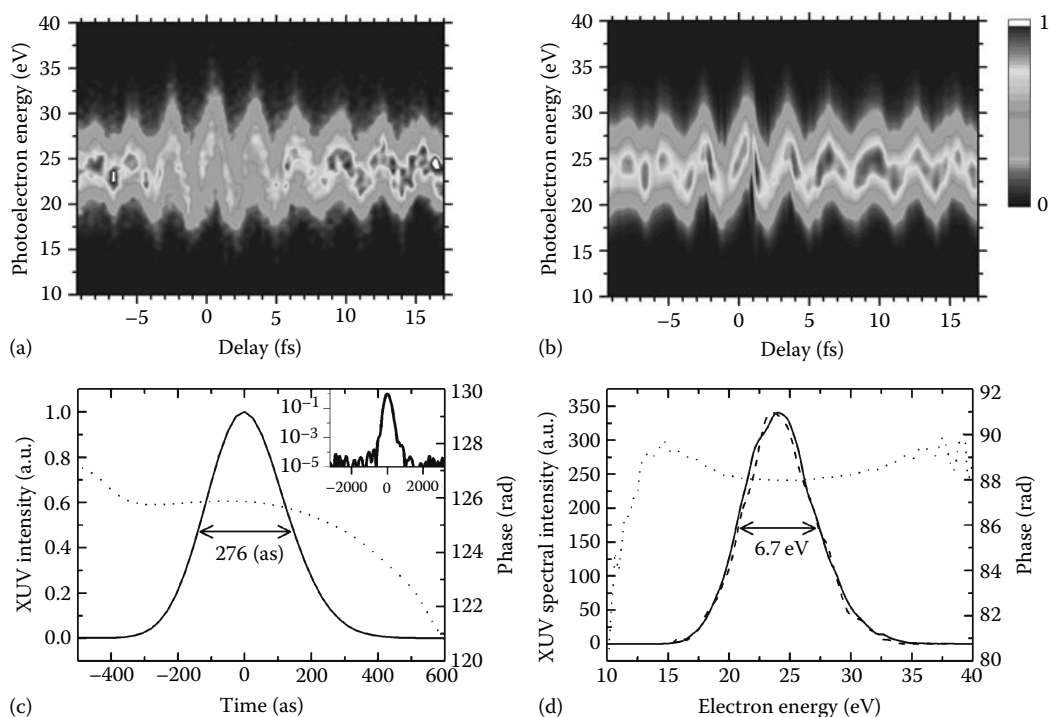


Figure 9.12 Measurement of isolated attosecond pulse generated by double optical gating with CRAB. (a) The measured CRAB trace. (b) The reconstructed CRAB trace. (c) The reconstructed XUV pulse shape (solid line) and phase (dotted line). (d) The reconstructed XUV spectrum (solid line) and phase (dotted line). Also shown is the measured XUV spectrum (dashed line) without the streaking laser for marginal comparison. (Reprinted with permission from S. Gilbertson, X. Feng, S. Khan, M. Chini, H. Wang, H. Mashiko, and Z. Chang, *Opt. Lett.*, 34, 2390, 2009. Copyright 2009 by the American Physical Society.)

similar to a Bessel–Gaussian beam. An image of the focused NIR beam captured with a CCD camera is shown in [Figure 9.13](#). A bright center spot is surrounded by concentric rings, which is a typical Bessel–Gaussian pattern. A lineout across the center of the image indicating the intensity variation is also shown. The diameter of the center is only $5.6\ \mu\text{m}$, which is closed to the estimate presented in [Figure 9.8](#). The XUV beam should be smaller than this value.

Instead of moving the XUV beam, the NIR beam is scanned, which serves the same purpose. A thin lens mounted on a three-dimensional translation stage is added to the streaking arm as shown in [Figure 9.11](#). The XUV spot is fixed in space. The position of the gas jet that produced the probing electron is fixed relative to the XUV beam. By moving the lens position in the transverse direction, the relative position between the XUV and the NIR is shifted, which is equivalent to have the XUV beam scanning across the Bessel–Gaussian beam.

The time delay between the NIR field and the attosecond pulse is fixed, assuring t_0 in Equation 9.23 is constant. [Figure 9.14a](#) shows the streaked electron spectrum plotted as a function of the transverse displacement

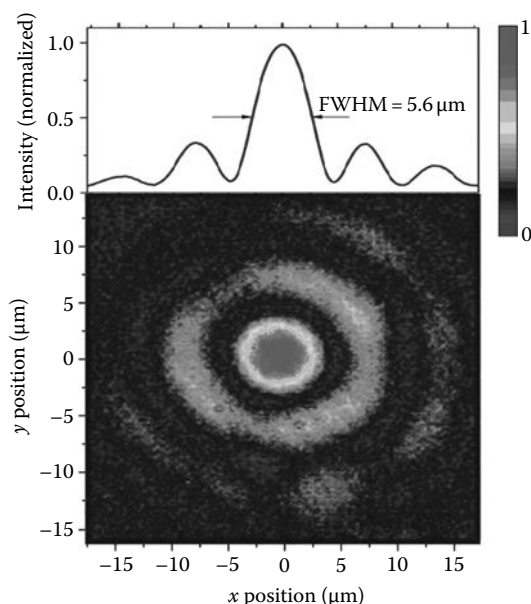


Figure 9.13 (Top panel) The lineout through the center of the image. (Bottom panel) CCD image of the focal spot of the streaking laser beam after reflection by the annular mirror. (Reprinted from S. Gilbertson, X. Feng, S. Khan, M. Chini, H. Wang, H. Mashiko, and Z. Chang, *Opt. Lett.*, 34, 2390, 2009. With permission of Optical Society of America.)

between the focused streaking beam and the XUV beam. As the streaking beam shifts across the XUV, the momentum of the electrons either shifted above or below the field free value, p_0 , thereby accurately mapping the transverse field.

To maximize the streaking, t_0 is set to zero so that both $\sin(\omega_0 t_0) = 1$ and $f(t_0) = 1$. The electric field value at point r can be determined from $(p(r)|_{t=\infty} - p_0)\omega_0/e$. The momentum value is obtained by finding the centroid of the electron kinetic-energy distribution at each spatial point r in Figure 9.14a, as shown by the white line. The resultant electric field is shown in Figure 9.14b. The field value directly gives the direction and magnitude of the electric field. Thus, the spatial distribution of the electric field in the Bessel–Gaussian beam is determined from the first principle of electrodynamics. Obviously, the field direction in the center spot is opposite to that in the first ring. The intensity ratio between the center spot and the first ring obtained from this method as shown in Figure 9.14b agrees well with the results shown in Figure 9.13a that is measured by the CCD.

The XUV spot size on the gas target is an important experimental parameter for attosecond pump–probe experiments. This is because the error of the attosecond reconstruction by CRAB depends on the XUV to NIR size ratio. To avoid NIR laser-intensity variation across the XUV spot, we would like the XUV to be as small as possible. If a transverse scan pattern like Figure 9.13 can be obtained for a setup, then one can be sure that the XUV beam is smaller than the center spot of the Bessel beam.

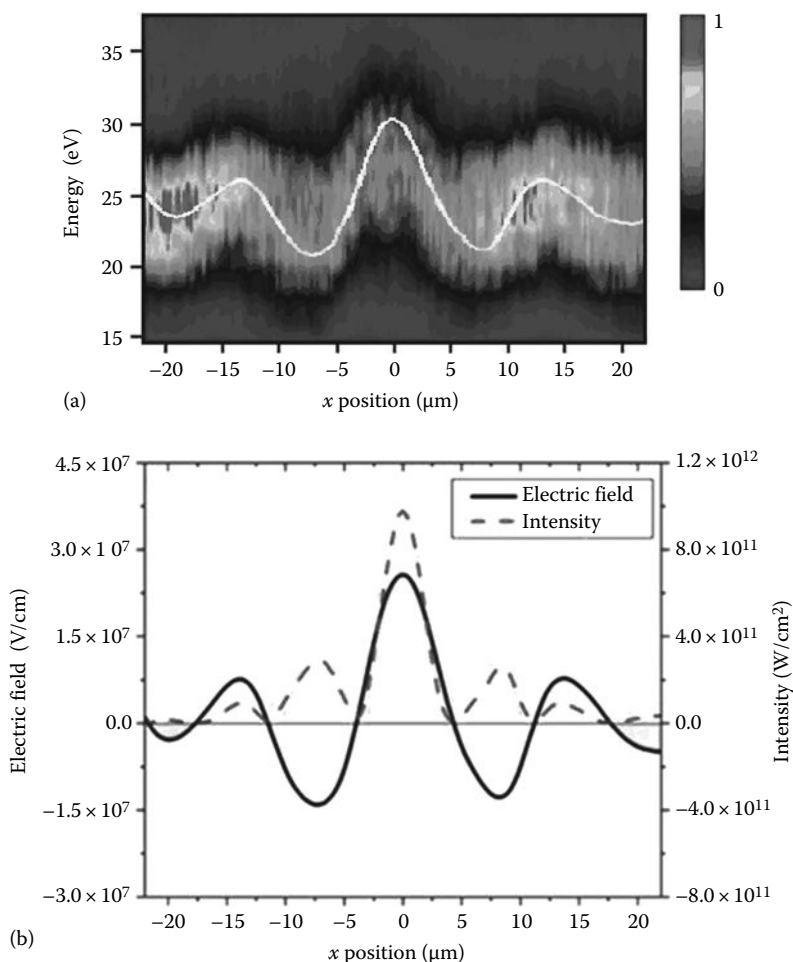


Figure 9.14 (a) The streaked photoelectron spectrum plotted as a function of the radial position of the streaking beam. (b) The electric field and intensity derived from (a). (Reprinted with permission from S. Gilbertson, X. Feng, S. Khan, M. Chini, H. Wang, H. Mashiko, and Z. Chang, *Opt. Lett.*, 34, 2390, 2009. Copyright 2009 by the American Physical Society.)

9.4 Controlling Two-Electron Dynamics in Helium Atoms

Atoms and molecules that contain many electrons are the building blocks of matter around us. Understanding the interaction between electrons is one of the basic tasks of atomic physics because the mechanisms have profound impact on the research of many disciplines of sciences as well as for advancing technologies. Due to very fast time scale of the electron-interaction dynamics and lack of tool with the required time resolution, most of the experiments were done in the spectrum (energy) domain in the past except for the Rydberg atoms. However, in such measurements, the effects of electron–electron interactions are averaged over long time

periods. As a result, the dynamics of electron–electron interactions are completely missing. The attosecond technology may provide the tools for probe and control the dynamics of the interactions between the electrons.

Even in the energy-domain experiments, the study of electron–electron interaction is not easy. This is because the motion of individual electrons in atoms is mostly governed by its Coulomb interaction with the nucleus, not with other electrons. In most cases, the effect of the other electrons is relatively weak. Consequently, it is difficult to observe pronounced features that characterize the electron–electron interaction. Fortunately, there are situations where electron–electron correlation plays major roles in atoms and molecules. The simplest case is the doubly excited state of a helium atom where both electrons are excited. Since there are only two electrons, it is relatively easy for the theorists to perform accurate simulations that can be compared with experiments.

Experimentally studying the electron-interaction process in the time domain is extremely challenging. The temporal resolution of the detector must be better than the time scale that the wave packet evolves. The characteristic time scale for electron motion is one atomic unit of time, which is 24 as. A new era in ultrafast science started in 2001 when the duration of coherent light pulses reached attosecond levels for the first time. Unlike femtosecond laser pulses that have been widely used for studying nuclear motions, the advent of the attosecond pulse makes it possible to investigate the much faster electron dynamics in atoms and molecules. As a proof-of-principle experiment, isolated attosecond pulses were used to measure the Auger decay time in Kr atoms, which yield results that agree well with spectroscopy measurements.

In this section, we introduce an experiment done in the authors' lab where double excitation and autoionization in helium atoms were investigated experimentally using isolated attosecond pulses as the pump and using NIR as the probe and control pulse. The interference between an attosecond and a femtosecond electron wave packet was modified by intense near-infrared laser pulses. It was demonstrated that the dynamics of the two electrons in helium can not only be monitored, but also be controlled.

9.4.1 Double Excitation of Helium

Autoionization is one of the processes that are dominated by electron–electron interactions. Being an atom that has only two electrons, helium is a perfect target for studying correlated electron dynamics by examining the autoionization process. In 1963, autoionization in helium with XUV light was experimentally observed by exciting the atoms with synchrotron radiation. In the past few years, it has become a hot research topic for theorists because of the possibility of studying autoionization in the time domain experimentally.

9.4.1.1 *Shell Model*

In the shell model, the motion of individual electrons is considered, whereas the effects of other electrons are treated as screening. The energy-level diagrams obtained using such model for helium are shown in [Figure 9.15](#). Starting with the ground state, absorption of a photon with energies in the

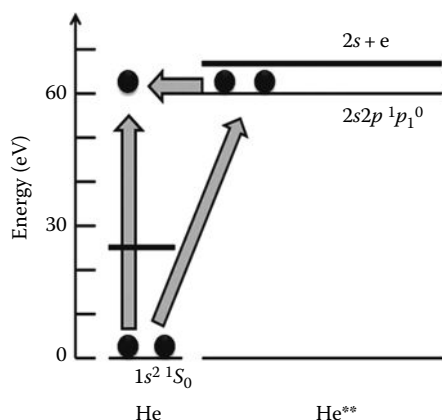


Figure 9.15 Autoionization depicted by the shell model.

60–79 eV range can populate ^1P doubly excited states, such as $2snp$, $2pns$, or $2pnd$. In this designation, for example, for $2snp$, it is implied that one electron is in the $2s$ state and the other in the np state, where $n = 2, 3, 4, \dots$. The lowest allowed transition to a two-electron excitation level, namely, from $1s^2\ ^1S_0$ to $2s2p\ ^1P_1^o$ requires a photon energy of 60.1 eV.

The 60.1 eV photon energy is larger than the ionization potential of the ground state (24.5 eV), thus a single electron can be emitted leaving the other electron in the ground state of He^+ , which is called “direct” ionization. Alternatively, both electrons can be excited to the $2s2p\ ^1P_1^o$ state. This state can then “autoionize” with one electron returning to the ground state of He^+ and the other electron being liberated from the atom. The autoionization is caused by the interaction of the excited electrons with other electrons.

The so-called Fano profiles present as sharp peaks in an XUV-absorption spectrum from helium is the result of the interference between the direct photoionization channel and the autoionization channel, as shown in Figure 9.16. The lifetime of the $2s2p$ doubly excited state, estimated from the width of the Fano spectral profile is ~ 17 fs. The physical processes can also be expressed by the following two equations:

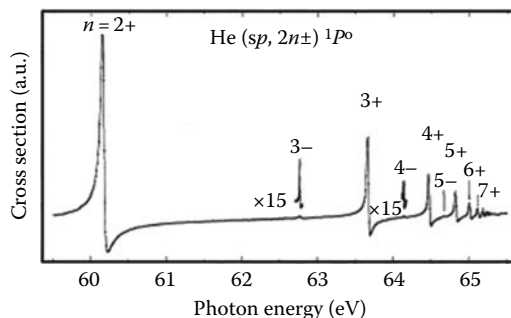


Figure 9.16 Interference peaks due to the interference of electron waves from the autoionization and the direct ionization. (Adapted from deHarak, B.A., Childers, J.G. and Martin, N.L.S., *Phys. Rev. A* 74, 032714, 2006. Copyright 2006 by the American Physical Society.)

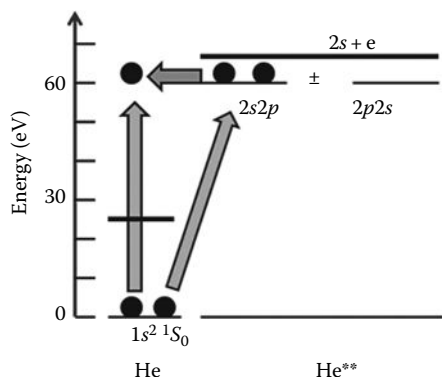
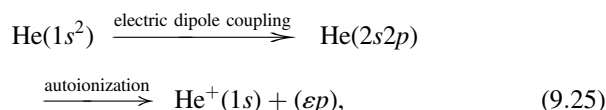


Figure 9.17 Autoionization depicted by mixing states.



and



where εp refers to a continuum state.

9.4.1.2 Coupled Pendulum Model

The shell-model picture is not fully consistent with the experimental results. It was argued by Fano and co-workers that a better description of these doubly excited states be expressed as $2snp + 2pns$, $2snp - 2pns$, and $2pnd$ series, later called “+,” “−,” and pd series, respectively, as shown in Figure 9.17. They predicted that only states in the “+” series are predominantly excited by the photons, which have been confirmed by measurements using synchrotron light.

It can be shown theoretically that the probabilities of dipole excitations from the ground state $1s^2$ to $2snp$ or $2pns$ are comparable. The $2snp$ and $2pns$ are nearly degenerate (separated by only 0.7 meV). In the Perturbation Theory, the wave function of the doubly excited state can be expressed as

$$\psi(2n \pm) = \frac{1}{\sqrt{2}} [u(2snp) \pm u(2pns)], \quad (9.26)$$

where $u(2snp)$ and $u(2pns)$ are the wave functions of the $2snp$ and $2pns$ states, respectively.

The “+” and “−” designation of doubly excited states provides interesting pictures of the correlated electron motion because it describes the joint motion of the two electrons explicitly, which is depicted in Figure 9.18. The pair of electrons in the “+” states approach or recede from the nucleus together. On the contrary, for the “−” states, one electron approaches the nucleus, the other is moving away. The motion

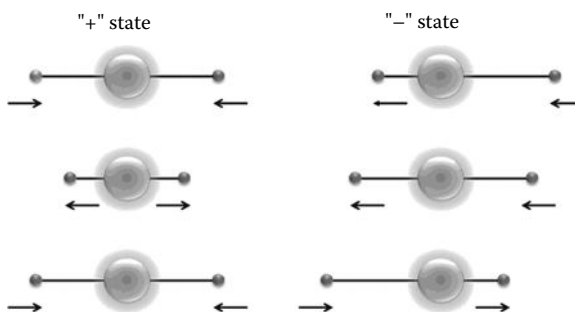


Figure 9.18 Motion of the two electrons in the “+” and “-” states.

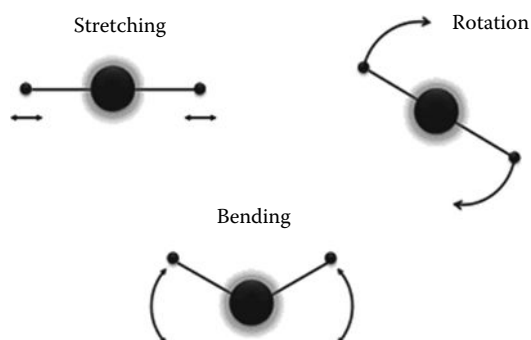


Figure 9.19 The stretching, rotational, and bending modes of doubly excited states.

is similar to the vibration of triatomic molecules if we image the two electrons as two atoms. These different modes of motion between the two electrons lead to drastic difference in photoabsorption probabilities and linewidth. The photoabsorption cross section for the “+” states are much larger than that of the “-” states.

Besides these radial stretches of the two electrons, the two electrons in doubly excited states can also execute bending vibrations and rotations. These modes are depicted in Figure 9.19. They are expected to have periods on the order of sub-fs to a few femtoseconds. To resolve such fast motions, attosecond time resolution is needed in the experiments. The binding energy of the $2s2p$ + state is 5.3 eV. An attosecond pulse centered at photon energy higher than 10.6 eV should be able to ionize both electrons with a single photon. The double ionization probability should depend on the separation of the two electrons. Therefore, measuring the He^{2+} yield as a function a delay between the 60.1 and 10.6 eV attosecond pulses, we can observe the oscillation of the two electrons.

9.4.2 Energy Domain Description of Fano Resonance

In 1961, Fano developed a theory based on configuration interaction to describe the autoionization. Consider an atom with a zero-order approximation Hamiltonian H_0 . Under such approximation, each electron

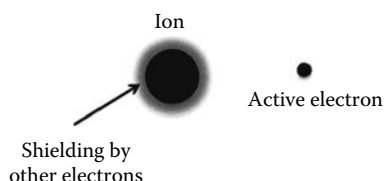


Figure 9.20 Zero-order approximation of a Hamiltonian.

moves independently in the static field of the nuclear and other electrons so that the motion of one electron does not depend on the motion of others, as depicted in Figure 9.20.

9.4.2.1 Zero-Order Approximation

The solutions of the Schrödinger Equation with one active electron are a number of zero-order approximation states, which contain a discrete one, $|\psi_b\rangle$, and a continuum of states $|\psi_\varepsilon\rangle$, i.e.,

$$H_0|\psi_b\rangle = E_b|\psi_b\rangle, \quad (9.27)$$

$$H_0|\psi_\varepsilon\rangle = \varepsilon|\psi_\varepsilon\rangle. \quad (9.28)$$

$|\psi_b\rangle$ and $|\psi_\varepsilon\rangle$ are orthogonal and normalized, i.e., $\langle\psi_\varepsilon|\psi_b\rangle = 0$, $\langle\psi_b|\psi_b\rangle = 1$, $\langle\psi_\varepsilon|\psi_{\varepsilon'}\rangle = \delta(\varepsilon - \varepsilon')$. In principle, there could be more than one discrete state. For simplicity, we assume that there is only one such state.

As an example, for helium atom, the function $r^2|\psi_b(r)|^2$ for $2s^2$ state is shown in Figure 9.21. r is the distance of the electron from the nucleus.

At large r , $\psi_\varepsilon(r) \propto \frac{1}{kr} \sin\left[kr + \frac{1}{k} \ln(2kr)\right]$. Here, k is the wave vector.

The discrete energy level E_b lies within the continuous range of ε . This situation is different from the hydrogen atom case where the continuum states lie above the discrete states. The comparison is shown in Figure 9.22.

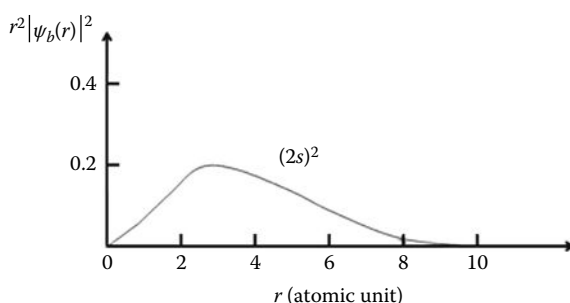


Figure 9.21 Radial electron density function calculated from $|\psi_b\rangle$ for the $(2s)^2$ state of helium. (Adapted with permission from P. Rehms and R.S. Berry, *Phys. Rev. A*, 23, 416, 1981. Copyright 1981 by the American Physical Society.)

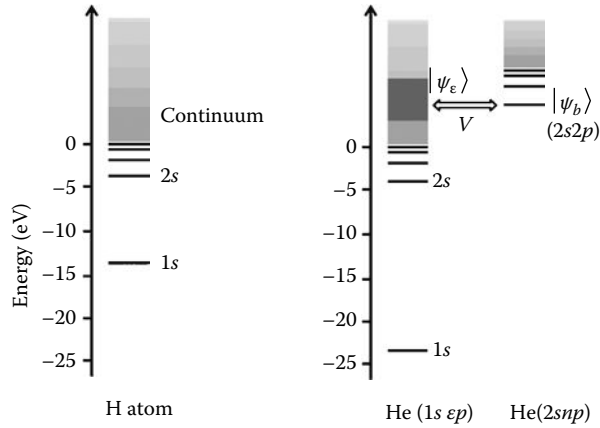


Figure 9.22 Comparison of the energy level diagrams of helium and hydrogen atoms.

9.4.2.2 Configuration Interaction

A more precise Hamiltonian can be written in a general form

$$H = H_0 + V, \quad (9.29)$$

where V is the correlation potential corresponding to the coupling between the discrete state $|\psi_b\rangle$ and the continuous state $|\psi_\varepsilon\rangle$ that is not included in H_0 . The two sets of zero-order solutions expressed by Equations 9.27 and 9.28 are called two configurations. The interaction between them is expressed by V . For helium, V includes the Coulomb interaction of the two electrons, $1/r_{12}$, where r_{12} is the distance between the two electrons. The configuration interaction leads to the decay of the electron from the bound state to the continuum state.

Since

$$\langle\psi_\varepsilon|H_0|\psi_b\rangle = E_b\langle\psi_\varepsilon|\psi_b\rangle = 0, \quad (9.30)$$

we have

$$\langle\psi_\varepsilon|H|\psi_b\rangle = \langle\psi_\varepsilon|H_0 + V|\psi_b\rangle = \langle\psi_\varepsilon|V|\psi_b\rangle = V_\varepsilon. \quad (9.31)$$

Apparently, the matrix element V_ε is a measure of the coupling strength between the two configurations, i.e., between $|\psi_b\rangle$ with a well-defined single value energy (infinitely thin linewidth) to a range of continuum states. V_ε is a smooth function of the continuous energy ε .

There is no interaction within the same configuration, i.e.,

$$\langle\psi_b|V|\psi_b\rangle = 0, \quad (9.32)$$

$$\langle\psi_\varepsilon|V|\psi_\varepsilon\rangle = 0. \quad (9.33)$$

Thus,

$$\langle\psi_b|H|\psi_b\rangle = E_b, \quad (9.34)$$

$$\langle\psi_\varepsilon|H|\psi_{\varepsilon'}\rangle = \varepsilon\delta(\varepsilon - \varepsilon'). \quad (9.35)$$

Using these orthogonal states as bases, an eigenstate of the full Hamiltonian corresponds to an energy E which can be found by solving the equation

$$H|\Psi_E\rangle = E|\Psi_E\rangle. \quad (9.36)$$

The solution can be expressed as

$$|\Psi_E\rangle = a(E)|\psi_b\rangle + \int d\varepsilon' b(\varepsilon', E)|\psi_{\varepsilon'}\rangle, \quad (9.37)$$

where $a(E)$ and $b(\varepsilon', E)$ are functions of E . This equation indicates that when an electron with energy E is detected, it could originate from either the discrete state $|\psi_b\rangle$ or from the continuum states $|\psi_{\varepsilon'}\rangle$. The decay from the discrete $|\psi_b\rangle$ state to the continuum state is called autoionization. The contribution from $|\psi_{\varepsilon'}\rangle$ is the direct ionization. Of course, the contribution from each ionization channel to the measured electron numbers is determined by the coefficients $a(E)$ and $b(\varepsilon', E)$.

9.4.2.3 Position of Resonance and Modified Bound State

It is shown by Fano that the coefficient

$$a(E) = \frac{\sin \Delta(E)}{\pi V_E}, \quad (9.38)$$

where

$$\Delta(E) = -\tan^{-1} \frac{\pi |V_E|^2}{E - [E_b + F(E)]} \quad (9.39)$$

is related to the phase shift of the bound-state wave function. The energy shift of the resonance

$$F(E) = P \int d\varepsilon' \frac{|V_{\varepsilon'}|^2}{E - \varepsilon'}. \quad (9.40)$$

Here, P means “the Cauchy’s principal part of the integral,” i.e., $F(E) = \lim_{\delta \rightarrow 0} \int_{|E - \varepsilon'| > \delta} d\varepsilon' \left(|V_{\varepsilon'}|^2 / E - \varepsilon' \right)$. $F(E)$ causes an energy-dependent shift of the bound state, which can be approximated by $F(E_b)$. Apparently, the shift depends on the strength of the configuration interaction $|V_{\varepsilon'}|^2$. The shift can be either positive or negative, depends on whether $|V_{\varepsilon'}|^2$ increase or decrease with ε' around E . An external laser field may shift the position of the resonance if it can affect any of the three quantities in the matrix element, $\langle \psi_{\varepsilon} | V | \psi_b \rangle$.

The other coefficient can be expressed as

$$b(\varepsilon', E) = \frac{V_{\varepsilon'}}{E - \varepsilon'} a(E) - \delta(\varepsilon' - E) \cos \Delta(E). \quad (9.41)$$

Substitute these two coefficients into Equation 9.37 and we have

$$|\Psi_E\rangle = a(E)|\psi_b\rangle + a(E) \left[P \int d\varepsilon' \frac{|V_{\varepsilon'}|^2}{E - \varepsilon'} \right] |\psi_{\varepsilon'}\rangle - \cos \Delta(E) |\psi_E\rangle. \quad (9.42)$$

By introducing a modified discrete state

$$|\Phi\rangle = |\psi_b\rangle + P \int d\varepsilon' \frac{V_{\varepsilon'}}{E - \varepsilon'} |\psi_{\varepsilon'}\rangle, \quad (9.43)$$

we have

$$|\Psi_E\rangle = \frac{\sin \Delta(E)}{\pi V_E} |\Phi\rangle - \cos \Delta(E) |\psi_E\rangle. \quad (9.44)$$

This equation provides another description of the measured free electron with energy E , i.e., the autoionization is from the discrete state modified by a mixture of states of the continuum. The direct ionization is still from the states $|\psi_E\rangle$.

9.4.2.4 Resonance Linewidth

By introducing an operator $\tilde{q}$, such that

$$\tilde{q}|\psi_b\rangle = \frac{1}{\pi V_E} |\Phi\rangle, \quad (9.45)$$

we arrive at

$$|\Psi_E\rangle = (\tilde{q} + \epsilon) \sin \Delta |\psi_E\rangle, \quad (9.46)$$

where the normalized energy is

$$\epsilon = \frac{E - [E_b + F(E)]}{\frac{1}{2}\Gamma(E)} = -\cot \Delta(E) \approx -\cot \Delta(E_b), \quad (9.47)$$

in which

$$\Gamma(E) = 2\pi |V_E|^2 \approx \Gamma(E_b) \equiv \Gamma \quad (9.48)$$

indicates the spectral width of the resonance. It can be seen that the resonance linewidth is directly related to the strength of the configuration interaction. A stronger coupling would lead to a faster decay from the bound state to the continuum states, which correspond to a broader resonance. In other words, the bound state decays to a broader energy range of the continuum states. If an external laser field can enhance the configuration interaction, then it would lead to a broadening of the resonance, or shortening of the decay lifetime.

9.4.2.5 Fano Profile and q Parameter

The photoabsorption of an XUV photon by an atom can be described by a dipole transition operator $D = -e\vec{r} \cdot \vec{E}_X(t)$. Here $\vec{E}_X(t)$ is the XUV field. The transition probability from an initial state to one of the eigenstates of H is given by

$$\langle \Psi_E | D | i \rangle = \frac{1}{\pi V_E^*} \sin \Delta(E) \langle \Phi | D | i \rangle - \cos \Delta(E) \langle \psi_E | D | i \rangle \quad (9.49)$$

or

$$\langle \Psi_E | D | i \rangle = \cos \Delta(E) \langle \psi_E | D | i \rangle \left[\tan \Delta(E) \frac{1}{\pi V_E^*} \frac{\langle \Phi | D | i \rangle}{\langle \psi_E | D | i \rangle} - 1 \right]. \quad (9.50)$$

Here, we introduce an important parameter

$$q(E) = \frac{1}{\pi V_E^*} \frac{\langle \Phi | D | i \rangle}{\langle \psi_E | D | i \rangle} \approx q(E_b) \equiv q, \quad (9.51)$$

with which we have

$$\langle \Psi_E | D | i \rangle = -\cos \Delta(E) \langle \psi_E | D | i \rangle \left[\frac{\pi |V_E|^2}{E - [E_b + F(E)]} q + 1 \right] \quad (9.52)$$

or

$$\langle \Psi_E | D | i \rangle = -\cos \Delta(E) \langle \psi_E | D | i \rangle \left[\frac{\Gamma/2}{E - [E_b + F(E)]} q + 1 \right], \quad (9.53)$$

i.e.,

$$\langle \Psi_E | D | i \rangle = -\langle \psi_E | D | i \rangle \cos \Delta(E) \left(\frac{q}{\epsilon} + 1 \right) \quad (9.54)$$

and

$$\frac{|\langle \Psi_E | D | i \rangle|^2}{|\langle \psi_E | D | i \rangle|^2} = \frac{(q + \epsilon)^2}{1 + \epsilon^2}, \quad (9.55)$$

which is the ratio of the transition probabilities from the initial state to the “modified” discrete state $|\Phi\rangle$ and to a band width Γ of an unperturbed continuum state $|\psi_E\rangle$.

For the case of photoemission experiments, Equation 9.50 gives the measured electron number as a function of electron energy, which is the photoelectron spectrum. The ratio of the transition probability $|\langle \Psi_E | D | i \rangle|^2$ to $|\langle \psi_E | D | i \rangle|^2$ is described as the Fano profile,

$$f(E) = \frac{(q + \epsilon)^2}{1 + \epsilon^2}. \quad (9.56)$$

In most cases, $|\langle \psi_E | D | i \rangle|^2$ depends weakly on E , thus measurable quantity $|\langle \Psi_E | D | i \rangle|^2$ gives the Fano profile.

Since

$$f(E) = \left(\frac{q + \epsilon}{1 - i\epsilon} \right) \left(\frac{q + \epsilon}{1 - i\epsilon} \right)^*, \quad (9.57)$$

we can introduce a q -dependent factor,

$$f_F(\epsilon) = \frac{q + \epsilon}{1 - i\epsilon}, \quad (9.58)$$

which describes the amplitude of the Fano profile in the energy domain. It can be decomposed into

$$f_F(E) = (q - i) \frac{\Gamma}{2} f_L(E) + i, \quad (9.59)$$

where $f_L(E) = (2/\Gamma)/(1 - i\epsilon)$ in the first term is the Lorentzian resonance profile, which describes the autoionization from the discrete state. The energy-independent second term on the right-hand side describes the contribution from the direct ionization.

The photoabsorption cross section equals $f_F(\epsilon)f_F^*(\epsilon)$. Taking the non-resonance transition into account, we have

$$\sigma(\hbar\omega_X) = \sigma_0(\hbar\omega_X) \frac{(q + \epsilon)^2}{1 + \epsilon^2}, \quad (9.60)$$

where $\sigma_0(\hbar\omega_X)$ gives the nonresonant cross section.

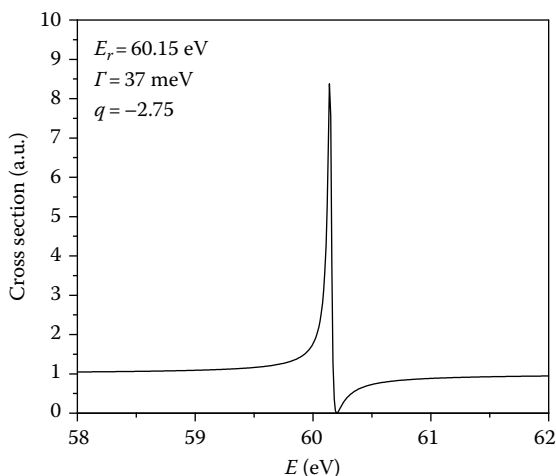


Figure 9.23 Fano profile of helium $2s2p$ state.

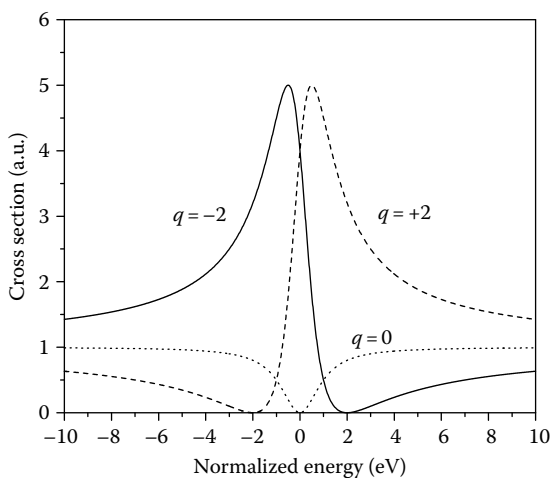


Figure 9.24 Fano profile with different q parameter.

For the $2s2p$ state of helium, the combination of Fano's theory of line profiles with the experimental results of Madden and Codling yielded the discrete state located at $E_r = 60.133 \pm 0.015$ eV, its linewidth $\Gamma = 0.038 \pm 0.004$ eV, and $q = -2.80 \pm 0.25$. The value 60.133 eV corresponds to the position of the resonance and not to the absorption maximum, which is at 60.123 eV. The most accurate experimental values are $E_r = 60.147 \pm 0.001$ eV, $\Gamma = 0.037 \pm 0.001$ eV, and $q = -2.75 \pm 0.01$. The Fano profile of this state is shown in Figure 9.23. Figure 9.24 shows the effects of the q parameter on the profile.

9.4.3 Time-Domain Description of Fano Resonance

Zhao and Lin extended Fano's theory to the time domain. Based on Equation 9.59, they expressed the scattering amplitude as

$$\begin{aligned} F_F(t) &= (q - i) \frac{\Gamma}{2} e^{-\frac{\Gamma}{2}t} e^{+iE_r t} + i\delta(t), \quad t \geq 0 \\ F_F(t) &= 0, \quad t < 0. \end{aligned} \quad (9.61)$$

The first term of Equation 9.61, $(q - i)(\Gamma/2)e^{-(\Gamma/2)t}e^{+iE_r t}$, describes the decay of the bound state (autoionization) and the second term, $i\delta(t)$, describes the direct ionization.

The scattering amplitude in the energy domain is obtained by the Fourier transform of Equation 9.61, i.e.,

$$\begin{aligned} \int_{-\infty}^{+\infty} F_F(t) e^{-iEt} dt &= \int_0^{+\infty} \left[(q - i) \frac{\Gamma}{2} e^{-\frac{\Gamma}{2}t} e^{+iE_r t} + i\delta(t) \right] e^{-iEt} dt \\ &= (q - i) \frac{\Gamma}{2} \frac{1}{\frac{\Gamma}{2} + i(E - E_r)} \int_0^{+\infty} e^{-x} dx + i. \end{aligned} \quad (9.62)$$

Thus, the scattering amplitude in the energy domain is given by

$$f_F(\epsilon) = -\frac{i + q}{1 + i\epsilon} + i = -\frac{q + \epsilon}{1 + i\epsilon}, \quad (9.63)$$

which is the complex conjugate of Equation 9.58. The minus sign in front of $(q + \epsilon)/(1 + i\epsilon)$ does not affect the calculation of the cross section. This confirms that Equation 9.61 is indeed the time-domain correspondence of Equation 9.58.

9.4.3.1 Lorentzian Lineshape

If the direct ionization contribution can be neglected ($q \approx 0$), the profile of the cross section peak is Lorentzian. In the energy domain such a resonance is described by the amplitude

$$f_L(E) = \frac{1}{\frac{\Gamma}{2} - i(E - E_r)} = \frac{2/\Gamma}{1 - i\epsilon}. \quad (9.64)$$

The cross section is given by

$$\sigma_r(E) \propto f_L(E) f_L^*(E) = \frac{1}{1 + \epsilon^2} = \frac{1}{1 + \left[\frac{(E - E_r)}{\Gamma/2} \right]^2}, \quad (9.65)$$

which is the Lorentzian lineshape. The absorption curve of helium without the direct ionization would be Lorentzian as shown in [Figure 9.25](#).

In the time domain, after the Fourier transform, it becomes a decaying amplitude such that

$$F_L(t) = e^{-\frac{\Gamma}{2}t} e^{iE_r t}. \quad (9.66)$$

The population of the resonance state after an excitation pulse is

$$P_r(t) = F_L(t) F_L^*(t) = e^{-\Gamma t}. \quad (9.67)$$

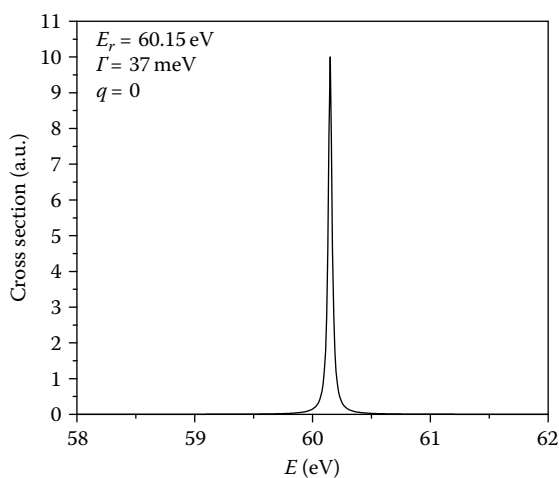


Figure 9.25 Lorentzian lineshape of the $2s2p$ state of helium if there is no direct ionization.

where $1/\Gamma$ is the lifetime of the resonance state. For the $2s2p$ state of helium, $\Gamma = 37$ meV, the lifetime

$$\frac{\hbar}{\Gamma} = 17.8 \text{ fs.} \quad (9.68)$$

The population of the $2s2p$ state due to autoionization is as shown in Figure 9.26.

When the $2s2p$ state is populated through double excitation using attosecond XUV pulses, the starting time of the decay is well defined, which provides the opportunity to measure the decay time directly. In the past, the decay time is derived from the Fano profile measured in the energy domain. For states with even longer lifetime, it could be more precise to do the measurement in the time domain.

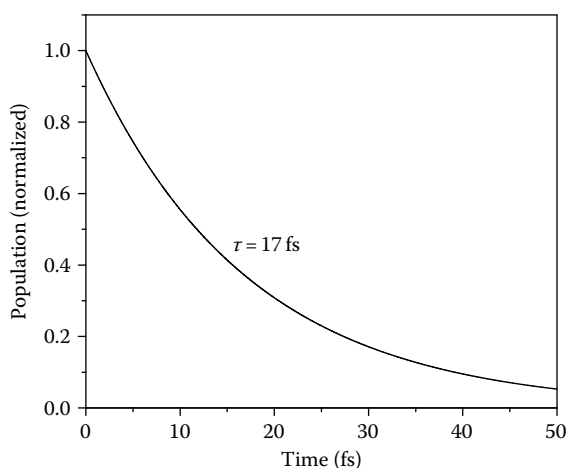


Figure 9.26 The decay of the $2s2p$ state of helium.

9.4.4 Strong-Field Approximation on XUV Photoionization in Laser Fields

So far we discussed the autoionization by XUV field only. Zhao and Lin studied the autoionization process in a combination of XUV and NIR laser field using the strong-field approximation (SFA) model. The attosecond XUV pump and femtosecond NIR probe may allow the process to be studied in the time domain. They assumed that the NIR laser does not participate in the creation and decay of the resonance. We should point that if the NIR laser is strong enough, then it can disturb the decay process. An electron in the excited resonant state can be freed before autoionization by absorbing one of several NIR photons. Within the SFA, only the resonance parameters in the energy domain are used and there is no need to calculate time-resolved electron spectrum by solving the time-dependent two-electron Schrödinger Equation.

9.4.4.1 Direct Ionization from the Ground State

As is discussed in Chapter 5, when all the excited states are not involved, under the SFA the probability amplitude of finding a continuum electron with momentum $\vec{p}$ at time t is given by

$$b_G(\vec{p}, t) = i \int_{-\infty}^t dt' \vec{\epsilon}_X(t') \cdot \vec{d}_c[\vec{p} - \vec{A}(t) + \vec{A}(t')] e^{i \int_{t'}^t \frac{|\vec{p} - \vec{A}(t') + \vec{A}(t'')|^2}{2} dt''} e^{i p t'}, \quad (9.69)$$

where $\vec{d}_c(\vec{p})$ is the dipole-transition matrix element for the ground state to continuum transition.

9.4.4.2 Autoionization from an Excited State

The probability amplitude of the continuum state with momentum $\vec{p}$ from the decay of a resonant state is

$$b_L(\vec{p}, t) = i \int_{-\infty}^t dt' e^{i p t'} \vec{\epsilon}_X(t') \cdot \vec{d}_r[\vec{p} - \vec{A}(t) + \vec{A}(t')] \times \int_{t'}^t dt'' e^{(-iE_r - \frac{\Gamma}{2})(t'' - t')} e^{i \int_{t''}^t \frac{|\vec{p} - \vec{A}(t') + \vec{A}(t'')|^2}{2} dt'''} \quad (9.70)$$

This equation describes the formation and the decay of the resonance in three steps.

First, the XUV attosecond pulse creates the population in bound state, E_r , at time t' . The dipole-transition element, $\vec{d}_r(\vec{p})$, is for the transition between the ground state and the excited state.

Second, this bound state decays at t'' to the continuum state with an amplitude, $e^{-\Gamma/2(t'' - t')}$, which describes the autoionization process.

Finally, the continuum electron freed from the bound state propagates from t'' to the final time t . The continuum electron is born at time t'' with a momentum $\vec{p}''$ which is related to the momentum $\vec{p}$ at time t by

$\vec{p}'' = \vec{p} - \vec{A}(t) + \vec{A}(t'')$ as a result of the conservation of the canonical momentum.

The measurement is done at $t = \infty$, thus $\vec{A}(t) = 0$. Suppose there is a delay, τ_d , between the XUV pulse and the laser field. Then,

$$b_L(\vec{p}, \tau_d) = i \int_{-\infty}^{+\infty} dt' e^{i\vec{p}t'} \vec{\mathcal{E}}_x(t' - \tau_d) \cdot \vec{d}_r[\vec{p} + \vec{A}(t')] \times \int_{t'}^{\infty} dt'' e^{(-iE_r - \frac{\Gamma}{2})(t'' - t')} e^{i \int_{t'}^{\infty} \frac{|\vec{p} - \vec{A}(t') + \vec{A}(t'')|^2}{2} dt''}. \quad (9.71)$$

9.4.4.3 Fano Profile

For an isolated Fano resonance, taking into account the contributions from both direct and autoionization, the measured continuum wave amplitude

$$b_L(\vec{p}, t) = i \int_{-\infty}^t dt' e^{i\vec{p}t'} \vec{\mathcal{E}}_x(t') \cdot \int_{t'}^t dt'' \times \left[\vec{d}_r[\vec{p} - \vec{A}(t) + \vec{A}(t')] \frac{\Gamma}{2} (q - i) e^{-\frac{\Gamma}{2}t'} e^{-iE_r t'} + \vec{d}_c[\vec{p} - \vec{A}(t) + \vec{A}(t')] i \delta(t'' - t') \right] e^{i \int_{t'}^t \frac{|\vec{p} - \vec{A}(t') + \vec{A}(t'')|^2}{2} dt''}. \quad (9.72)$$

In the combined XUV and NIR field, the complex amplitude of the state amplitude at the time of the measurement is

$$b_L(\vec{p}, \tau_d) = i \int_{-\infty}^{+\infty} dt' e^{i\vec{p}t'} \vec{\mathcal{E}}_x(t' - \tau_d) \cdot \int_{t'}^{+\infty} dt'' \times \left[\vec{d}_r[\vec{p} + \vec{A}(t')] \frac{\Gamma}{2} (q - i) e^{-\frac{\Gamma}{2}t'} e^{-iE_r t'} + \vec{d}_c[\vec{p} + \vec{A}(t')] i \delta(t'' - t') \right] e^{i \int_{t'}^{+\infty} \frac{|\vec{p} - \vec{A}(t') + \vec{A}(t'')|^2}{2} dt''}. \quad (9.73)$$

9.4.5 TDSE Simulations

9.4.5.1 XUV Photoionization with and without the Laser Field

Time-dependent Schrödinger Equation (TDSE) was solved by Tong and Lin to study doubly excited states of helium in the combined field of an attosecond XUV pulse and an intense NIR laser pulse with peak intensity of the order of $I_0 = 10^{12}$ W/cm². The XUV pulse duration used in the simulation has a full width at half maximum of 200 as, with the central photon energy of 60.0 eV. The NIR laser pulse duration is 5.0 fs centered at 800 nm.

The attosecond XUV pump pulse defines a sharp start time where doubly excited states are formed. The autoionization process can be modified by the intense two-cycle NIR femtosecond laser pulses overlapping

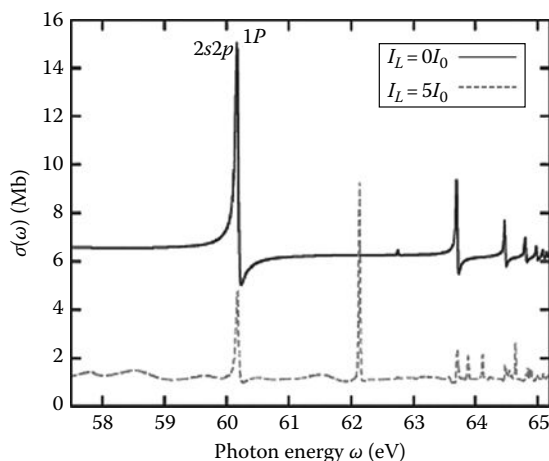


Figure 9.27 The helium photoionization cross section with or without the laser fields. For better visualization, the field-free cross section was upshifted by 5 Mb. The laser intensity is in the units of $I_0 = 10^{12} \text{ W/cm}^2$. (Reprinted with permission from X.M. Tong and C.D. Lin, *Phys. Rev. A*, 71, 033406, 2005. Copyright 2005 by the American Physical Society.)

with the attosecond XUV pulses. The modification can be controlled changing the laser intensity, pulse length, and the time delay between the XUV and NIR pulses. With the many knobs available to tune, it is hoped that the dynamics between two electrons, including the autoionization itself, can be probed and eventually controlled.

When the laser field is turned off, calculated photoionization cross sections are shown in Figure 9.27. It agrees with the measured results shown in Figure 9.16. In this spectrum, the first peak is the $2s2p \ ^1P^o$ state. The pronounced series of peaks are the higher members of the “+” series.

When the laser field is turned on, many additional resonances or doubly excited states are induced when a laser of intensity of $5I_0$ is added on top of the attosecond XUV pulse with no time delay between the two. It is relatively easy to observe new resonances if the photoabsorption of the XUV pulse is performed in the presence of a moderately intense laser pulse.

9.4.5.2 Laser-Intensity Dependence

The dependence of laser-assisted photoelectron spectra on the intensity of the laser, I_L , is shown in Figure 9.28. The laser intensity is varied from 0 to 1, 5, and $10 I_0$. The presence of the $2p^2 \ ^1S$ NIR induced state at 62.2 eV is most prominent. The sharp structure is due to its long lifetime (140 fs). Its Lorentzian shape indicates that there is little interference with the continuum ($q \approx 0$).

Figure 9.28 also shows that the oscillator strength of the $2s2p \ ^1P$ decreases with increasing laser intensity, as its strength is being shared by the other states. The NIR laser intensity is chosen such that direct ionization from the ground state by the NIR laser is negligible. Consequently, the total oscillator strength by the XUV pulse is just redistributed in all the resonances as well as the continuum electrons.

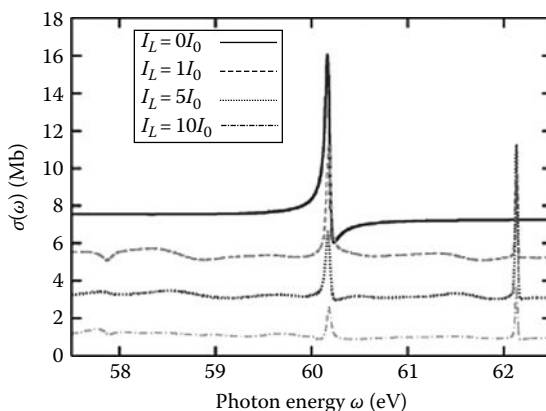


Figure 9.28 Laser-assisted photoionization cross section of He with laser intensities at 0, 1, 5, and $10I_0$, respectively. (Reprinted with permission from X.M. Tong and C.D. Lin, *Phys. Rev. A*, 71, 033406, 2005. Copyright 2005 by the American Physical Society.)

9.4.5.3 TDSE Simulations on Studying Two-Electron Dynamics by Attosecond Pump–Probe

By solving TDSE, Morishita, Watanabe, and Lin showed that when attosecond XUV pulses are used to doubly ionize a two-electron wave packet of helium, the time-resolved correlated motion of the two electrons can be probed by measuring their six-dimensional momentum distributions. For simple wave packets, when the measured momenta are analyzed in appropriate coordinates, the stretching, rotational, and bending vibrational modes of the joint motion in momentum space can be revealed.

Doubly excited states are known to form rotational and vibrational supermultiplets. For example, the $2s^2\ ^1S^e$ state is the ground state of the rovibrational motion, the $2p^2\ ^1D^e$ state is the first rotational excited state, and $2p^2\ ^1S^e$ is the first excited vibrational state. They studied wave packets created by mixing these states. A coherent state made of $2s^2\ ^1S^e + 2p^2\ ^1S^e$, is a bending vibrational wave packet, with an oscillation period of 980 as. A wave packet made of $2s^2\ ^1S^e + 2p^2\ ^1D^e$ is a rotational wave packet, with an oscillation period of 2.0 fs. These oscillations can be resolved in attosecond pump–probe experiments.

9.4.6 Experiments on Autoionization of Helium in NIR Laser Fields

The energy-level diagram of helium including the NIR laser action is shown in left graph of Figure 9.29. The lifetime of the doubly excited state, estimated from the width of the Fano spectral profile, is ~ 17 fs. Observing the autoionization process in time domain is almost impossible using synchrotron light because its pulse duration is on the picosecond level.

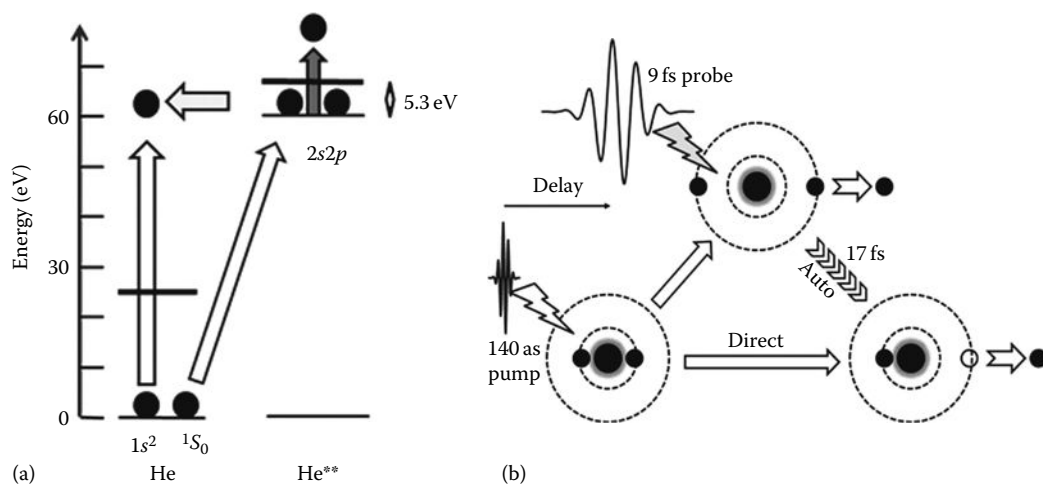


Figure 9.29 (a) Energy diagram of helium. (b) Principle of controlling autoionization of helium. A single isolated XUV pulse starts the process by simultaneously exciting the two electrons to the $2s2p$ state and ionizing one of the electrons from the ground state. An intense near infrared 9 fs laser synchronized with the attosecond pulse ionizes the electrons in the $2s2p$ state before they decay through autoionization. Thus, the interference between the two ionization channels is altered. (Adapted from S. Gilbertson, M. Chini, X. Feng, S. Khan, Y. Wu, and Z. Chang, *Phys. Rev. Lett.*, 105, 263003, 2010. Copyright 2010 by the American Physical society.)

The author's group conducted experiments on autoionization using isolated attosecond pulses as the pump.* The scheme is depicted in the right graph of Figure 9.29. The spectrum of the isolated 140 as duration covers the range from ~ 30 to 70 eV. Unlike a train of attosecond pulses whose spectrum has large amplitude modulations, the spectrum of single isolated attosecond pulses is a smooth continuum around 60.1 eV, which is like the light from the synchrotron, making the observation of Fano profile possible. The duration of the attosecond pulse is much shorter than the lifetime of the $2s2p$ state, 17 fs, which precisely sets the starting time of the autoionization.

The decay of the population in the $2s2p$ state was disturbed by a 9 fs NIR laser pulse centered at 780 nm. In the experiments, the photoelectron spectrum was measured around the Fano peak at 35 eV as a function of delay between the attosecond XUV pulse and the near infrared pulse. The intensity of the laser is on the order of $1 \times 10^{12} \text{ W/cm}^2$, which is intense enough to ionize the electrons in the $2s2p$ state before they completely decay through autoionization. The depletion of the population reduces the contribution from the autoionization to the Fano interference. Consequently, the integrated Fano peak signal is reduced when the two pulses overlap. This allowed the control of the autoionization process and the measurement of the lifetime of the $2s2p$ state.

* From the work: Gilbertson, S., Chini, M., Feng, X., Khan, S., Wu, Y., and Chang, Z., *Phys. Rev. Lett.*, 105, 263003, 2010. Copyright 2010 by the American Physical Society.

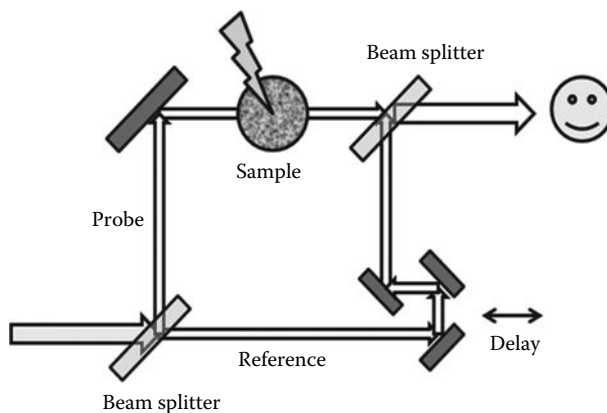


Figure 9.30 A Mach–Zehnder optical interferometer analogy of the Fano profile formation.

The ionization potential of the $2s2p$ state, I_p , is 5.2 eV. Under the experimental condition, the Kyldash parameter is

$$\gamma = \sqrt{\frac{I_p}{2U_p}} \approx 6, \quad (9.74)$$

where U_p is the ponderomotive potential. This implies that the ionization is a multiphoton type, which is highly nonlinear. Thus ionization happened in a time period much shorter than the 17 fs decay time, which is required for this study.

The optical analogy of the Fano profile formation initiated by an attosecond pulse and influenced by an NIR pulse is a Mach–Zehnder interferometer, as illustrated in Figure 9.30. The action of the attosecond pulse to the atomic ground state serves as a BS that sends two waves, double excitation and direct ionization, to two difference interferometer arms. The common ionic final state serves as the second BS that combines the waves from the autoionization and direct ionization arms. In the experiment, the wave from the direct ionization serves as the reference beam. Any changes of the amplitude or phase of the wave from the autoionization channel caused by the NIR laser are equivalent to the change of the complex index of refraction of the optical samples in the other arms.

9.4.6.1 Experimental Setup

The attosecond pump–probe setup is illustrated in Figure 9.31. The input laser, 9 fs, 650 μ J pulse centered at 780 nm with 1.5 kHz repetition rate, was split into two components by an 80% transmitting beamsplitter. The main portion of the beam passed through double optical gating optics in the collinear configuration and was focused to a Neon-filled gas cell. This generated a single isolated XUV pulse with a duration of 140 as. Next, the XUV pulse passed through an aluminum filter to remove the residual infrared photons. The beam then passed through the center of a hole-drilled mirror (HM).

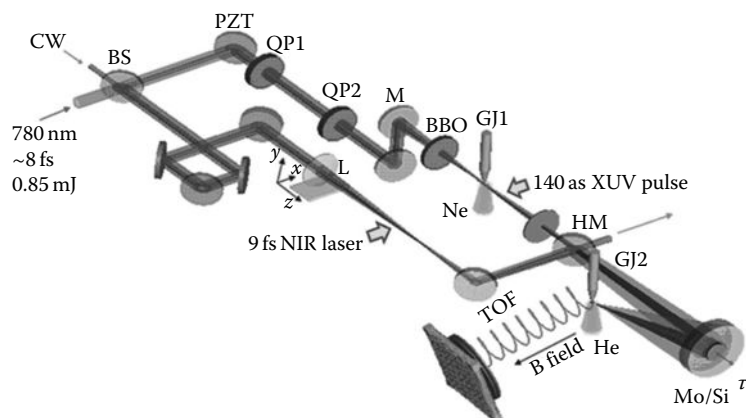


Figure 9.31 Experimental setup for controlling Fano interference in helium.

The reflected portion of the original 9 fs NIR beam passed through an equal optical path length and recombined with the attosecond XUV pulse at the HM. The two beams then reflected at near normal incidence from a two component spherical mirror. The inner mirror, mounted on a PZT, was coated with a Mo/Si multilayer that reflected the XUV photons with about 6% reflectance over the range of 31–70 eV, which covers the 60.1 eV needed for double excitation. The outer mirror was silver coated to reflect the 9 fs NIR pulse. The two beams focused to a second gas jet filled with helium gas that was the target to be studied.

The XUV beam generated a photoelectron burst that was collected by a TOF spectrometer with its axis aligned along the NIR laser and the XUV polarization axis. By varying the delay between the Mo/Si and the Ag mirrors with the PZT, the electron spectrum as a function of delay between the XUV pulse and the NIR pulse was recorded. The energy resolution of the TOF was ~ 0.7 eV at 60 eV. This is much broader than the linewidth of the $2s2p$ Fano profile (34 meV), but still allowed us to clearly identify the Fano peaks at 35 eV.

The isolated attosecond pulses generated with DOG were characterized using the same setup. Neon was used as the detection gas that has the benefits of a stronger signal due to the larger cross section as compared with helium and also because the cross section is flat as compared to other detection gases. The XUV pulse width and phase was determined by using the FROG-CRAB method. Here, the NIR laser pulse was used to streak the photoelectrons by giving them a momentum kick. The energy of the photoelectron spectrum was recorded as a function of delay between the NIR pulse and the XUV pulse as shown in Figure 9.32. A comparison between the actual experimental trace and the reconstructed trace is shown in Figure 9.32a and b, respectively. The reconstructed temporal profile is shown in Figure 9.32c. Here, the pulse duration is 136 as and the pre- and post-pulses have a very low contribution, much less than 0.1%, indicating the attosecond pulse is indeed isolated. Figure 9.32d shows a comparison between the unstreaked XUV spectrum and the Fourier transform of the temporal profile shown in Figure 9.32c. The excellent agreement indicates

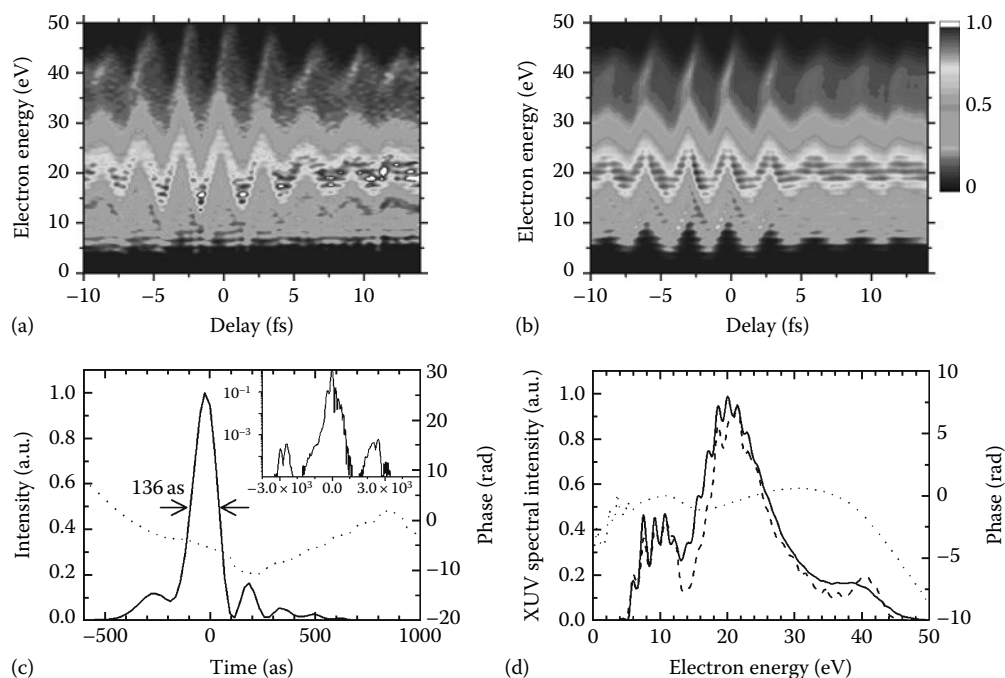


Figure 9.32 Characterization of the isolated attosecond pulses generated with DOG.

an accurate retrieval. Simply changing the detection gas to helium ensured the experimental conditions of the attosecond pulses remained the same.

The 140 as single attosecond pulses were then used to pump the helium atoms. If the NIR laser pulse is combined with the XUV photon pulse before the end of the autoionization lifetime, we can directly probe the electron dynamics. Figure 9.33a shows the photoelectron spectrogram as a function of the delay between the XUV and the NIR pulse. The $2s2p$

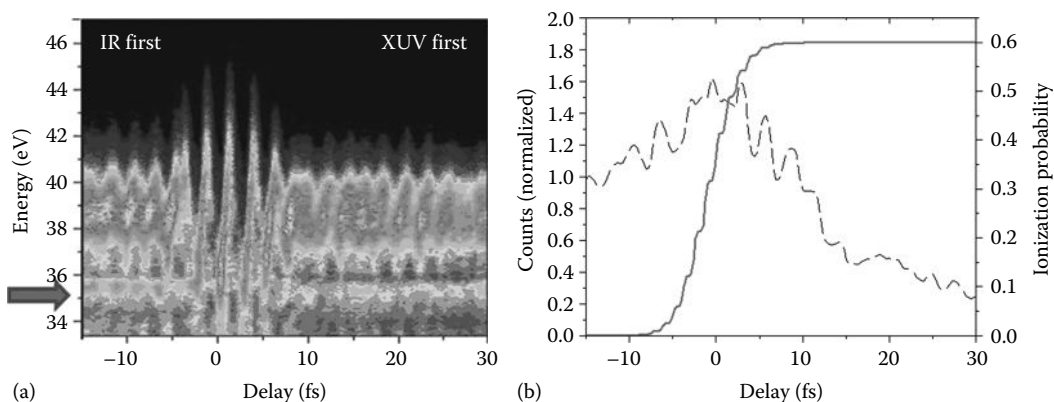


Figure 9.33 (a) The measured photoelectron spectrum. (b) Integrated Fano peak signal and the probability of multiphoton ionization of the $2s2p$ state. The observation direction of is parallel to the NIR laser field vector.

autoionization resonance occurs at 60.1 eV in photon energy or 35.5 eV in photoelectron energy. When the NIR pulse comes first, the resonance is stronger than when it lags behind the XUV pulse. We attribute this to depletion of the autoionization resonance by the NIR pulse due to multiphoton ionization. Since the helium atom is in the $2s2p$ doubly excited state, only 5.2 eV more is required to ionize the atom, leaving the remaining electron in either the $2s$ or $2p$ state.

Based on the magnitude of the momentum shift of the photoelectrons near the high-energy cutoff from the streaking shown in the left graph of Figure 9.33, the intensity of the NIR field was estimated to be $\sim 7 \times 10^{11}$ W/cm². The depletion of the population in the $2s2p$ state at such intensity is calculated using the PPT model which is discussed in Chapter 4. The ionization probability as a function of the delay is shown in the right graph of Figure 9.33. This yielded a final ionization probability of 0.6. Also shown is the total counts integrated over the width of the $2s2p$ resonance as a function of delay. These two results imply that the coupling strength of the two ionization channels, direct and autoionization can be controlled by varying the NIR intensity and delay.

9.4.6.2 Calculations under the Strong-Field Approximation

The formation of the Fano resonance in the presence of a strong NIR laser field can be described by extending a model based on the SFA. In this model, the amplitude of an electron wave in the continuum state with momentum $\vec{p}$ at the observation time $t \rightarrow \infty$ is given in atomic units by

$$b(\vec{p}, \tau_d) = i \int_{-\infty}^{\infty} dt \int_t^{\infty} dt' \vec{\epsilon}_X(t - \tau_d) \vec{d}[\vec{p} - \vec{A}(t)] e^{i\vec{p}t} \\ \times \left[a(t' - t) \frac{\Gamma}{2} (q - i) e^{-(iE_r + \frac{\Gamma}{2})(t' - t)} + i\delta(t' - t) \right] e^{i \int_t^{\infty} \frac{|\vec{p} - \vec{A}(t')|^2}{2} dt'}, \quad (9.75)$$

where τ_d is the delay between the attosecond XUV pulse and the NIR laser pulse. The ionization potential is $I_p = 24.5$ eV and the excitation energy is $E_r = 60.15$ eV. $\Gamma = 37$ meV is the resonance width. The parameter $q = -2.75$.

The depletion of the population in the $2s2p$ state by the NIR laser was taken into account by the amplitude $a(t)$. The amount that the doubly excited state is depleted depends on the delay, since electrons born into the doubly excited state at different delays will interact with a different time range of the NIR pulse. To determine the population of the doubly excited state, the PPT model for multiphoton ionization was used. The ionization probability was calculated at each delay and the amplitude $a(t)$ was modified accordingly in Equation 9.75.

The resulting electron spectrogram, after convolution with the TOF-energy resolution, is shown in the left graph of Figure 9.34 for the XUV pulse reconstructed above in the presence of an NIR field 9 fs in duration and with peak intensity of 7×10^{11} W/cm². Several features can be noted. First, the high-energy portion of the spectrum near 40 eV exhibits the

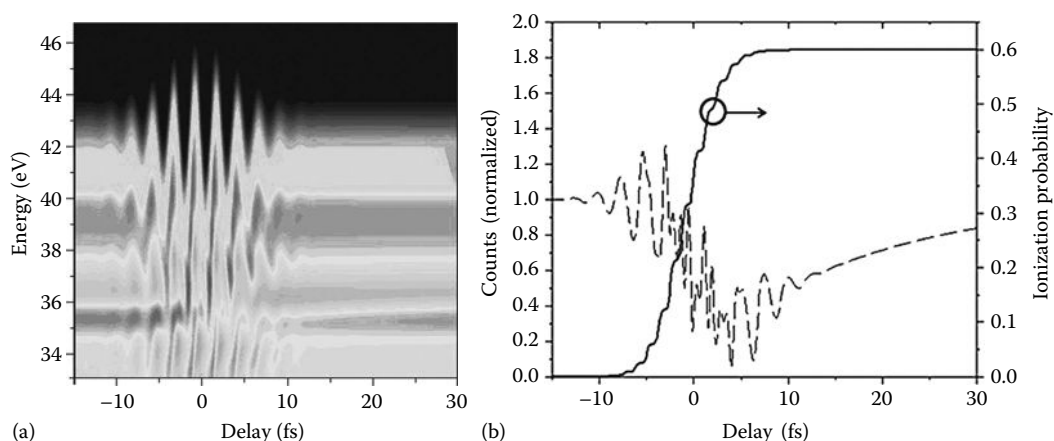


Figure 9.34 (a) The calculated spectrum. (b) Integrated Fano peak signal and the probability of multiphoton ionization of the $2s2p$ state. The observation direction of is parallel to the NIR laser field vector.

streaking effect, indicating the single isolated attosecond pulse. Furthermore, the autoionization line shows a distinct asymmetry about the delay $\tau_d = 0$.

The right graph in Figure 9.34 shows the integrated autoionization line at 35 eV as a function of the delay, along with the ionization probability for comparison. The observation direction is parallel to the NIR laser-field vector. For negative values of delay, for which the NIR laser arrives first, the population decreases quite sharply, and roughly follows the ionization probability. For positive delay, however, the population increases more slowly, and follows the autoionization decay. The periodic modulation of the autoionization line is due to the streaking effect of the directly photoionized electrons which overlap with the autoionization line energy. The decrease of the autoionization signal is correlated with the increase of the probability of the multiphoton ionization, which implies that the contribution to the Fano interference from the autoionization channel can be controlled by the NIR laser. In other words, the effective q parameter can be tuned by the laser. The tuning “knobs” include the laser intensity, pulse duration, and the time delay.

The oscillation in the integrated Fano peak is due to the streaking effects. When the more intense portion of the electron around 39 eV is down shifted by the laser field to 35 eV, the contribution to the interference from the direct ionization channel is increased, which changes the signal at 35 eV. The signal strength depends on both the amplitude and the phase of the down shifted electron wave. This provides a “knob” to control Fano interference on attosecond time scale.

In order to separate the effects of the multiphoton ionization from the streaking effect, the calculation was repeated for an NIR laser polarized perpendicular to the direction of the detected photoelectrons. As has been shown previously, the streaking effect is minimized in this case, due to the diminishing of the $\vec{p} \cdot \vec{A}(t)$ term of the phase modulation $[\vec{p} - \vec{A}(t)]^2$ in the

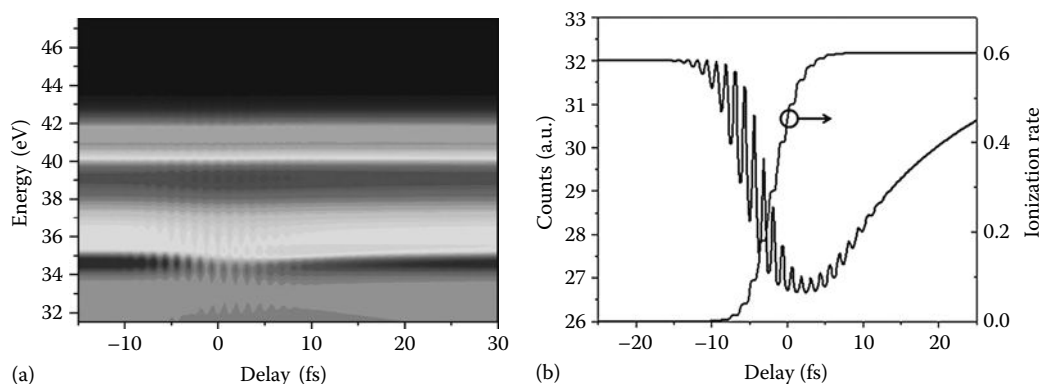


Figure 9.35 (a) The calculated spectrum. (b) Integrated Fano peak signal and the probability of multiphoton ionization of the $2s2p$ state. The observation direction of is perpendicular to the NIR laser field vector.

final term of Equation 9.72. The spectrogram simulated under this configuration with identical XUV and NIR streaking fields to those in Figure 9.34a is shown in Figure 9.35a after convolution with the energy resolution, along with the integrated ionization line at 35 eV in Figure 9.35b. Again, the multiphoton ionization probability of the $2s2p$ state and integrated Fano peak is shown for comparison. Although some modulation is still present from the streaking effect perpendicular to the polarization direction, the effects from the multiphoton ionization on the Fano peak become more pronounced.

After the Fano peak drops to the minimum, it recovers as the delay increases. The recovery curve can be used to measure the autoionization decay time of the $2s2p$ state. Assuming the NIR laser is close to a delta function in time, the multiphoton ionization probability is close to a step function. The detected signal becomes

$$|b(35 \text{ eV}, \tau_d)|^2 \propto \int_0^{\tau_d} e^{-\Gamma t} dt \propto 1 - e^{-\Gamma \tau_d}. \quad (9.76)$$

Thus, the method can be used to observe electron dynamics in atoms.

9.4.6.3 Discussion

The experimental and simulation results demonstrated that contribution from the autoionization to the Fano profile can be controlled by either the laser intensity or the delay between the attosecond excitation pulse and the NIR ionization laser pulse. This allowed us to control the coupling strength of the two channels that interfere, or the so called q parameter, which was almost impossible to do without using attosecond pulses to start the autoionization process. Thus, attosecond pulses make it possible to *control* electron dynamics in atoms. The whole process can be time resolved by streaking the photoelectrons with the NIR laser field, or by measuring the attosecond transient absorption.

Problems

- 9.1 Find the exact energy diagram of helium. Determine the bandwidth of the attosecond pulse that can excite $1s2p$ to $1s4p$ states.
- 9.2 A wave packet is a superposition of the ground state and the first excited state of helium. What is the period of the wave packet motion?
- 9.3 In the pump–probe experiment discussed in [Section 9.1](#), what happens to the measured signal if the pump intensity is much lower than $6 \times 10^{14} \text{ W/cm}^2$.
- 9.4 For the pulses centered at 20 eV, what is the diffraction-limited focal spot size when the f-number of the focusing mirror is 1? Suppose that XUV beam diameter on a spherical focusing mirror is 1 cm, what should the focal length of the mirror be to achieve a focal spot less than $1 \mu\text{m}$?
- 9.5 In the oscilloscope shown in [Figure 9.2](#), the electron energy is 20 keV, $d = 1 \text{ cm}$, $l = 1 \text{ cm}$, and $L = 20 \text{ cm}$. The vertical shift of the electron seen on the screen is 2 cm. What is the voltage applied on the deflection plate? What is the electric-field strength between the two plates?
- 9.6 In Problem 9.5, what is the time for an electron to travel through the deflection plates ($l = 1 \text{ cm}$)? Assuming the travel time is the response time of the measurement, calculate the corresponding frequency in GHz.
- 9.7 Design a device similar to [Figure 9.2](#) that can measure the magnetic-field strength. What limits the response time?
- 9.8 If the deflection plates in [Figure 9.2](#) are replaced by a laser beam, can it measure the electric field oscillation of the laser?
- 9.9 A wave packet is created by mixing the $1s^2$ and the $1s3p$ of helium atom. What is the oscillation period of the wave packet?
- 9.10 A wave packet is created by mixing the $1s2p$ and the $1s3p$ of helium atom. What is the oscillation period of the wave packet?
- 9.11 The intensity of a linearly polarized laser beam is 10^{14} W/cm^2 . The center wavelength is 800 nm. Plot the force experienced by an electron located inside the laser beam as a function of time. As a comparison, plot the force for a He–Ne laser with 100 W/cm^2 . Its center wavelength is at 632.8 nm.
- 9.12 In an attosecond-streak camera, the focal spot size of the laser beam is $30 \mu\text{m}$. The center wavelength is 800 nm. Is a 5 mm long gas target small enough for measuring the laser-field oscillation?
- 9.13 Read the literature and find out the lifetime and q parameters of the $2s3p$, $2s4p$ states of helium. Plot the profiles.

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Appendix A: Solutions to Selected Problems

Chapter 1

1.1 $\varepsilon = P_0 \tau$.

1.4 (a) 0.33 mJ; (b) 15.7 GW.

1.6 $P = I_0 \pi w^2$.

1.7 $E = 1.2 \times 10^{-10} \text{ V/cm}$.

1.15 $\tau \Delta \omega_{FWHM} = 5.56$.

1.16 $p/kT = 3.2 \times 10^{16} \text{ atoms/cm}^3$.

1.22 398.

1.25 $2.2 \times 10^{16} \text{ W/cm}^2$.

Chapter 2

2.2 $\frac{2}{\pi} \frac{P}{w_0^2} = 1.6 \times 10^5 \frac{\text{W}}{\text{cm}^3}$, $z_R = 1.57 \text{ mm}$.

(a) $I(r, z) = \frac{2}{\pi} \frac{P}{w_0^2} \frac{1}{1 + \left(\frac{z}{z_R}\right)^2}$. (b) $(r, z) = \frac{2}{\pi} \frac{P}{w_0^2} e^{-\frac{2r^2}{w_0^2}}$.

2.3 $\frac{z_R(400 \text{ nm})}{z_R(800 \text{ nm})} = \frac{0.785 \text{ mm}}{0.394 \text{ mm}} = 2$.

2.4 $\frac{\Theta(400 \text{ nm})}{\Theta(800 \text{ nm})} = \frac{12.7 \text{ mrad}}{25.6 \text{ mrad}} = \frac{1}{2}$.

2.15 (a) 10^7 ; (b) 89 nm; (c) 459 nm; (d) 1.8 μm .

2.21 (b) 1 kW; (d) no, the wavelength is too long.

2.23 a. At 0.8 μm : $\frac{c}{n(\lambda_0)} = 2.050 \times 10^8 \text{ m/s}$.

At 1.6 μm : $\frac{c}{n(\lambda_0)} = 2.056 \times 10^8 \text{ m/s}$.

b. At $\lambda_0 = 0.8 \mu\text{m}$: $\frac{c}{n(\lambda_0) - \lambda_0 \frac{dn}{d\lambda} \Big|_{\lambda_0}} = 2.031 \times 10^8 \text{ m/s}$.

At $\lambda_0 = 1.6 \mu\text{m}$: $\frac{c}{n(\lambda_0) - \lambda_0 \frac{dn}{d\lambda} \Big|_{\lambda_0}} = 2.046 \times 10^8 \text{ m/s}$.

c. At $\lambda_0 = 0.8 \text{ }\mu\text{m}$: $\frac{\lambda_0^3}{2\pi c^2} \frac{d^2 n}{d\lambda^2} \Big|_{\lambda_0} = 46.97 \text{ fs}^2/\mu\text{m}$.

At $\lambda_0 = 1.6 \text{ }\mu\text{m}$: $\frac{\lambda_0^3}{2\pi c^2} \frac{d^2 n}{d\lambda^2} \Big|_{\lambda_0} = 23.5 \text{ fs}^2/\mu\text{m}$.

d. At $\lambda_0 = 0.8 \text{ }\mu\text{m}$: $\tau(z) = \tau_0 \sqrt{1 + \left(\frac{z}{z_D}\right)^2}$, increase.

At $\lambda_0 = 1.6 \text{ }\mu\text{m}$: increase.

2.24 a. $I_{0p} = \frac{4\sqrt{2}}{\sqrt{\pi}} \frac{\mathcal{E}}{\tau \cdot \pi w^2} = 5.7 \times 10^{11} \text{ W/cm}^2$, $n = n_0 + n_{21} I_{0p}$
 $= 1.500057$.

b. On the leading (or rising) edge: increase with time.
 On the falling edge: decrease with time.

c. $b_0 = 0$, $b = \frac{4\pi}{\lambda_0} a_0 n_{21} I_{0p} L = 0.002 \text{ fs}^{-2}$.

d. $B = \frac{2\pi}{\lambda_0} n_{21} I_{0p} L = 0.45 \text{ rad}$.

e. $\Delta f_{in} = \frac{0.44}{\tau} = 1.76 \times 10^{13} \text{ Hz}$, $\Delta f = \Delta f_{in} \sqrt{1 + (2B)^2}$
 $= 2.36 \times 10^{13} \text{ Hz}$.

Chapter 3

3.13 $(0.2 \text{ rad}) \times \frac{180^\circ}{\pi \text{ rad}} = 11.5^\circ$.

3.14 $(0.2 \text{ rad}) \times \frac{1.55 \text{ fs}}{2\pi \text{ rad}} = 50 \text{ as}$.

3.15 $1.01^5 - 1^5 = 0.05 = 5\%$.

Chapter 4

4.7 At $0.8 \text{ }\mu\text{m}$, the values of the chirp at 1×10^{14} and $1 \times 10^{15} \text{ W/cm}^2$ are 30.9 and 3.09 as/eV, respectively. At $1.6 \text{ }\mu\text{m}$, the values of the chirp at 1×10^{14} and $1 \times 10^{15} \text{ W/cm}^2$ are 15.4 and 1.5 as/eV, respectively. Both laser intensity and laser wavelength can serve as experimental knobs to tune the chirp of attosecond pulses.

Chapter 5

5.1 The field amplitude ratio is 4. The intensity ratio is 16.

5.3 The ionization potential $I_p = 24.587 \text{ eV} = 0.9 \text{ a.u.}$, $\alpha = 1.8 \text{ a.u.}$

$$\begin{aligned}
 5.9 \quad E_0 &= \sqrt{\frac{2I}{\epsilon_0 c}} = \sqrt{\frac{2 \times 10^{12}}{8.85 \times 10^{-12} \times 3 \times 10^3}} \\
 &= 2.7 \times 10^7 \text{ V/m. It is 10 times weaker than the field at} \\
 &\quad \times 10^{14} \text{ W/cm}^2.
 \end{aligned}$$

$$\begin{aligned}
 5.10 \quad E &= \frac{e^2}{4\pi\epsilon_0 r^2} = \frac{(1.6 \times 10^{-19})^2}{4\pi \times 8.85 \times 10^{-12} \times (1 \times 10^{-9})^2} \\
 &= 2.3 \times 10^6 \text{ V/m. It is 10 times weaker than the laser field at} \\
 &\quad 10^{12} \text{ W/cm}^2.
 \end{aligned}$$

$$\begin{aligned}
 5.11 \quad t &= \frac{d}{v} = \frac{d}{\sqrt{\frac{2K}{m_e}}} = \frac{10^{-9}}{\sqrt{\frac{2 \times 10 \times 1.6 \times 10^{-19}}{9.1 \times 10^{-31}}}} = 0.53 \times 10^{-15} \text{ s} \\
 &= 0.5 \text{ fs.}
 \end{aligned}$$

Chapter 6

- 6.1** No, the transition from a continuum state to the ground state only emits one XUV photon.
- 6.2** No, they are the fundamental laws of physics, like Newton's law for mechanics.
- 6.17** The reference frame moves in the z direction with a velocity equal to the light speed in vacuum.
- 6.18** It should increase linearly with the driving laser energy.

Chapter 7

- 7.1** The radius of the center spot increases and the amplitude of the first ring decreases as the inner radius of the annular aperture increases.
- 7.2** For $a = 10$ mm, the phase changes from 0 to π rad in 0.4 mm. For $a = 5$ mm, the phase changes from 0 to π rad in 1.2 mm.
- 7.3** The kinetic energy $K = \hbar\omega_x - I_p$. For argon, K extends from 9.2 to 34.2 eV. For neon, it extends from 4.5 to 28.5 eV.

Chapter 8

- 8.3** 25 as. The spectral phase is a straight line.
- 8.7** Suppose that the input pulse is linearly polarized in the vertical (y) direction. The laser field before the first quartz plate can be expressed as

$$\varepsilon_{input}(t) = E_0 e^{-2\ln 2 \left(\frac{t}{\tau_p}\right)^2} \cos \left[\omega_0 \left(t + \frac{nT_0}{2} \right) \right]. \quad (\text{P8.7.1})$$

The time offset $nT_0/2$ in the carrier wave is introduced to simplify the analysis. The plate is a multi-order whole-wave plate, i.e.,

$$T_d = nT_0. \quad (\text{P8.7.2})$$

Quartz is a positive uniaxial crystal, which means that the o-ray pulse leads the e-ray pulses. The optic axis of the crystal is set at 45° to the vertical direction. After the first quartz plate, the o-ray and e-ray pulses are

$$\begin{aligned}\varepsilon_o(t) &= \frac{E_0}{\sqrt{2}} e^{-2\ln 2 \left(\frac{t+T_d/2}{\tau_p} \right)^2} \cos[\omega_0(t + nT_0)] \\ &= \frac{E_0}{\sqrt{2}} e^{-2\ln 2 \left(\frac{t+T_d/2}{\tau_p} \right)^2} \cos[\omega_0(t)],\end{aligned}\quad (\text{P8.7.3})$$

$$\begin{aligned}\varepsilon_e(t) &= \frac{E_0}{\sqrt{2}} e^{-2\ln 2 \left(\frac{t-T_d/2}{\tau_p} \right)^2} \cos[\omega_0(t)] \\ &= \frac{E_0}{\sqrt{2}} e^{-2\ln 2 \left(\frac{t-T_d/2}{\tau_p} \right)^2} \cos[\omega_0(t)].\end{aligned}\quad (\text{P8.7.4})$$

Their polarization directions are orthogonal.

Projecting them into the vertical (y) and horizontal (x) directions, we have

$$\varepsilon_x(t) = \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+T_d/2}{\tau_p} \right)^2} - e^{-2\ln 2 \left(\frac{t-T_d/2}{\tau_p} \right)^2} \right] \cos(\omega_0 t), \quad (\text{P8.7.5})$$

$$\varepsilon_y(t) = \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+T_d/2}{\tau_p} \right)^2} + e^{-2\ln 2 \left(\frac{t-T_d/2}{\tau_p} \right)^2} \right] \cos(\omega_0 t). \quad (\text{P8.7.6})$$

8.8 The laser field before the first quartz plate can be expressed as

$$\varepsilon_{\text{input}}(t) = E_0 e^{-2\ln 2 \left(\frac{t}{\tau_p} \right)^2} \cos \left[\omega_0 \left(t + \frac{\left(n + \frac{1}{2} \right) T_0}{2} \right) \right]. \quad (\text{P8.8.1})$$

The plate is a multi-order half-wave plate, i.e.,

$$T_d = \left(n + \frac{1}{2} \right) T_0. \quad (\text{P8.8.2})$$

After the first quartz plate, the o-ray and e-ray pulses are

$$\begin{aligned}\varepsilon_o(t) &= \frac{E_0}{\sqrt{2}} e^{-2\ln 2 \left(\frac{t+T_p/2}{\tau_p} \right)^2} \cos \left[\omega_0 \left(t + nT_0 + \frac{T_0}{2} \right) \right] \\ &= -\frac{E_0}{\sqrt{2}} e^{-2\ln 2 \left(\frac{t+T_d/2}{\tau_p} \right)^2} \cos[\omega_0(t)],\end{aligned}\quad (\text{P8.8.3})$$

$$\begin{aligned}\varepsilon_e(t) &= \frac{E_0}{\sqrt{2}} e^{-2\ln 2 \left(\frac{t-T_d/2}{\tau_p} \right)^2} \cos[\omega_0(t)] \\ &= \frac{E_0}{\sqrt{2}} e^{-2\ln 2 \left(\frac{t-T_d/2}{\tau_p} \right)^2} \cos[\omega_0(t)].\end{aligned}\quad (\text{P8.8.4})$$

Their polarization directions are orthogonal.

Projecting them into the vertical (y) and horizontal (x) directions, we have

$$\varepsilon_x(t) = \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+T_d/2}{\tau_p} \right)^2} + e^{-2\ln 2 \left(\frac{t-T_d/2}{\tau_p} \right)^2} \right] \cos(\omega_0 t), \quad (\text{P8.8.5})$$

$$\varepsilon_y(t) = \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+T_d/2}{\tau_p} \right)^2} - e^{-2\ln 2 \left(\frac{t-T_d/2}{\tau_p} \right)^2} \right] \cos(\omega_0 t). \quad (\text{P8.8.6})$$

8.9 In the case where the first quartz plate is a multi-order whole-wave plate, the laser field components after the quarter-wave plate are

$$\begin{aligned} \varepsilon_x(t) &= \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+\frac{T_d}{2}+\frac{T_0}{4}}{\tau_p} \right)^2} - e^{-2\ln 2 \left(\frac{t-\frac{T_d}{2}+\frac{T_0}{4}}{\tau_p} \right)^2} \right] \cos\left(\omega_0 t - \frac{T_0}{4}\right) \\ &= \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+\frac{T_d}{2}+\frac{T_0}{4}}{\tau_p} \right)^2} - e^{-2\ln 2 \left(\frac{t-\frac{T_d}{2}+\frac{T_0}{4}}{\tau_p} \right)^2} \right] \sin(\omega_0 t), \end{aligned} \quad (\text{P8.9.1})$$

$$\varepsilon_y(t) = \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+\frac{T_d}{2}}{\tau_p} \right)^2} + e^{-2\ln 2 \left(\frac{t-\frac{T_d}{2}}{\tau_p} \right)^2} \right] \cos(\omega_0 t). \quad (\text{P8.9.2})$$

For such a configuration, $\varepsilon_x(t)$ is the gating field and $\varepsilon_y(t)$ is the driving field for polarization gating.

In the case where the first quartz plate is a multi-order half-wave plate, the laser field components after the quarter-wave plate are

$$\varepsilon_x(t) = \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+\frac{T_d}{2}}{\tau_p} \right)^2} + e^{-2\ln 2 \left(\frac{t-\frac{T_d}{2}}{\tau_p} \right)^2} \right] \cos(\omega_0 t), \quad (\text{P8.9.3})$$

$$\begin{aligned} \varepsilon_y(t) &= \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+\frac{T_d}{2}+\frac{T_0}{4}}{\tau_p} \right)^2} - e^{-2\ln 2 \left(\frac{t-\frac{T_d}{2}+\frac{T_0}{4}}{\tau_p} \right)^2} \right] \cos\left(\omega_0 t + \frac{T_0}{4}\right) \\ &= \frac{E_0}{\sqrt{2}} \left[e^{-2\ln 2 \left(\frac{t+\frac{T_d}{2}+\frac{T_0}{4}}{\tau_p} \right)^2} - e^{-2\ln 2 \left(\frac{t-\frac{T_d}{2}+\frac{T_0}{4}}{\tau_p} \right)^2} \right] \sin(\omega_0 t). \end{aligned} \quad (\text{P8.9.4})$$

Now, $\varepsilon_x(t)$ is the driving field and $\varepsilon_y(t)$ is the gating field for polarization gating. This is desirable when the time-of-flight spectrometer axis is in the horizontal direction. The input beam is vertically polarized so that high reflectance over a broad spectral range on 45° turning mirrors in laser beams can be achieved.

Chapter 9

9.1 $E_{1s4p} - E_{1s2p} = 2.5$ eV, which is the minimum bandwidth required.

$$\mathbf{9.2} \quad \frac{h}{E_{1s2p} - E_{1s^2}} = \frac{6.626 \times 10^{-34}}{21.2 \times 1.6 \times 10^{-19}} = 195 \times 10^{-18} \text{ s} = 195 \text{ as}.$$

9.3 The signal will be dominated by ions from the ionization of the $1s^2$ state, which makes the detection of ions from the mixing state difficult.

9.4 The wavelength of the 20 eV light is $\lambda_X = 0.062 \text{ } \mu\text{m}$, which is the diffraction-limited focal spot size when $f\# = 1$. When the beam diameter $D = 1 \text{ cm}$, the focal length should be $f = (w/\lambda_X)D = (1 \text{ } \mu\text{m}/0.062 \text{ } \mu\text{m})1 \text{ cm} = 16 \text{ cm}$.

$$\mathbf{9.10} \quad \frac{h}{E_{1s3p} - E_{1s2p}} = \frac{6.626 \times 10^{-34}}{1.9 \times 1.6 \times 10^{-19}} = 2.180 \times 10^{-15} \text{ s} = 2.18 \text{ fs}.$$