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DEVELOPMENT OF A TABLETOP COHERENT SOFT X-RAY SOURCE

by

HANFU KONG
B.S. University of Central Florida, 2018

A thesis submitted in partial fulfillment of the requirements
for the degree of Master of Science
in the College of Optics and Photonics
at the University of Central Florida
Orlando, Florida

Spring Term
2018

Major Professor: Zenghu Chang
ABSTRACT

The goal of this thesis is to design a tabletop coherent soft X-ray source for attosecond high resolution imaging. We collect signals from gas cells with different length and lens with different focal length. A spectrometer with a grating and a CCD camera is applied to observe and measure the spectrum of the X-ray attosecond pulses.

This thesis first introduces the theory background of ultrafast lasers, then mainly explains high harmonic generation, which is the key method for attosecond pulses generation, subsequently presents the experiment system and analyzes the results from the experiment, also compares different combinations of parameters of the devices.
ACKNOWLEDGMENTS

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1 INTRODUCTION

Since Maiman invented the first laser in 1960, research and application on laser have attracted great attention from physics and optics researchers. For decades, the field of laser has been grown tremendously, and thus caused great impact on our life. The characteristics of laser due to the extremely high photon degeneracy make them widely used in various fields, such as transmission of information using lasers with optical fibers, fine-grained micro-nano components processing, vision correction surgery and cancer treatment, even laser pointers used in our daily life. With the expansion of laser applications, the demand for specific-performance lasers is increasing.

1.1 Ultrafast lasers

Ultrafast lasers are the lasers with ultrashort pulses whose time durations are of the order of picosecond (1ps=10^{-12}s) or less.

In 1962, Q-switch technology was invented, and the laser pulse duration was shortened to nanoseconds. Then in 1964, the first mode-locked He-Ne with 2.5ns pulse duration came out, and later dye lasers also using mode-locked technology generates much shorter duration pulses. More recently, these lasers were replaced by all-solid-state diode-pumped Ti:Sapphire oscillator [1]. Since this material shows a high damage threshold and excellent thermal conductivity, also has a high fluence saturation and also the gain bandwidth is very large from 650nm to 1100nm, it can be considered as one of the best amplifying media at 800nm [2].
Being compact, robust and durable, this technique brought new life to existing exciting applications and has created new directions for photonics research. Applying Ti:Sapphire material together with Kerr lens passive mode-locking achieves ultrashort pulses at high repetition rates. And in 1985, D. Strickland et.al introduced a new technology for ultra-intense pulses generation, Chirped Pulse Amplification (CPA), which proved to be an important breakthrough in this area [3]. Based on the idea that generating an initial ultrashort pulse with low energy, stretching it in temporal domain through spectral phase modulation, then amplifying it with keeping the intensity low enough that can avoid nonlinear effects, CPA technique allows lasers working at high fluences and having good energy extractions while due to the low intensity, nonlinear effects are reduced and destruction of the instruments can be avoided. Figure 1.1 provides an illustration of a normal Chirped Pulse Amplifier system: firstly, a pulse comes out from a short pulse oscillator, then get stretched by a spectral phase modulator used as a stretcher, and then get amplified by an amplifier, finally compressed by another spectral phase modulator which is opposite to the former one, and becomes a short pulse.

However, even applying CPA technology, the shortest pulse duration at mJ level from the Ti:Sapphire laser is still broader than 10 fs (10^{-15} s) [4]. The problem is that through amplifying, the gain region is narrow, which restricts the spectral bandwidth, so the temporal duration cannot be further narrowed. Since one optical cycle is 2.6 fs at 800 nm, to obtain shorter pulse duration at few cycle regime, new techniques were invented such as Optical
Parametric Chirped Pulse Amplification (OPCPA) [5], gas-filled hollow core fiber [6] and filamentation [7].

Figure 1.1 Illustration of the CPA system: Firstly, the pulse is stretched by a spectral phase modulator, then it is amplified and finally compressed by another spectral phase modulator opposite to the initial one. Reprinted from [8]

1.2 High Harmonic Generation

When it come to inside the molecules, atoms move on the femtosecond timescale, and for electrons with much less mass, they change their distribution due to interatomic forces on the attosecond timescale, which is $10^{-18}$ s. Laser pulse duration applying technologies mentioned above can only reach sub-10 fs regime. To further suppress the pulse duration, the central wavelength of the laser must be shifted to extreme ultraviolet (XUV). This could be achieved by High Harmonic Generation (HHG). The first HHG was observed in 1977, pulses
from intense CO\textsubscript{2} laser interacted with plasma generated from some kinds of solid targets \cite{9}. Then in 1987, McPherson and colleagues observed HHG in gases \cite{10}, which was mainly applied in today’s researches. The temporal duration of the high-order harmonics can reach in attosecond region, and thus it can be used as a versatile tool such as observing atomic-scale electron dynamics in real-time, also work for spectroscopy. Figure 1.2 gives a spectrum from hard x-rays to near infrared (NIR). The harmonic orders generated from 800 nm light are shown in the orange box. We can see that the third order harmonic is 267 nm and reaches the ultraviolet region, and higher order harmonics from 27 to 801 nm are in the soft X-rays region. Most commonly used high harmonics are in the XUV and soft X-rays regions.

![Figure 1.2 Spectrum from X-rays to NIR. The top section shows the visible light region in the spectrum. The orange box at the bottom provides locations of several harmonics in the spectrum of 800 nm light. Reprinted from [11]](image-url)
1.3 Coherent Diffractive Imaging and Ptychography

Coherent Diffractive Imaging (CDI) is an emerging technique that achieves diffraction-limited resolution. Since the original demonstration, CDI arouses widely concern and show great potential in material science and biological applications. These are due to the characteristics that CDI works without either optical imaging systems or reference structures for holography [12] or uniformly redundant arrays [13]. And thus, CDI is considered as an ideal technique for XUV and X-ray spectral range imaging, where conventional imaging methods cannot be used to obtain the results.

The method of CDI can be explained as follow. When a coherent wave illuminates a sample, it will generate a diffraction pattern based on the Fourier transform of the sample structure. A detector can be used to collect the magnitude of the Fourier transform, which equals to the square root of the diffraction intensity, while the phase information cannot be collected in this way, which causes the phase information lost. However, we can use an iterative algorithm to retrieve the phase information from accurately measured diffraction intensity, and then correctly reconstruct the sample structure.

Ptychography, which is also called scanning probe CDI, is an important type of CDI methods [14]. Ptychography is a new phase retrieval method, which provides relative phase shift as well as attenuation related to the sample. The basic theory is using a structured illumination probe to scan the sample, then collect a sequence of diffraction patterns from the overlapping regions between adjacent illuminated areas. Ptychography takes the overlapping
region as a real space constraint, which is different from conventional CDI, and allows reconstruction of extended objects. Figure 1.3 depicts a ptychography system. A gas-discharge plasma works as a light source, the XUV beam generated from the source passes through a 500 μm aperture (A), gets spatially filtered and becomes much smaller in spatial range. Then it propagates on a multilayer Bragg mirror (B), which acts as a spectral filter, and the 17.3 nm wavelength light gets reflected to a 10 μm aperture (C). A sample (D) is placed behind the aperture and is movable in 3D based on a mobile platform. The residue beam propagates through the sample and generate diffraction patterns, finally use an XUV sensitive camera (E) to collect the patterns.

Figure 1.3 Schematic of ptychography system. XUV beam generated from the interaction between driving lasers and noble gas goes through a 500 μm aperture (A) and gets spatially filtered, then reflected by a Bragg grating (B) which acts as a spectral filter to select 17.3 nm wavelength. After passing a 10 μm aperture (C), the beam propagates through the sample (D) which is movable in 3D, and an XUV sensitive camera (E) is applied to detect the diffraction patterns of the beam. Reprinted from [15]

Scanning of X-ray ptychography can be relatively fast. In 2D projection, the scanning
speed can reach 25000 resolution elements per second (resel/s) for 40 nm resolution [16], and in 3D the speed can reach 5800 resel/s for 15 nm resolution [17].
2 ULTRAFAST LASERS

2.1 Wave propagation and Gaussian beam

We come from the equation that

\[ E(x, y, z) = \psi(x, y, z)e^{-jkz} \tag{2.1.1} \]

\( e^{-jkz} \) states that the beam should propagate close to a uniform plane wave, and the factor \( \psi \) shows how much the beam deviates from a plane wave which is what we want to know.

\[ \nabla^2 E + \frac{\omega^2}{c^2} n^2 E = 0 \tag{2.1.2} \]

or writing as

\[ \nabla_t^2 E + \frac{\partial^2 E}{\partial z^2} + \frac{\omega^2}{c^2} n^2 E = 0 \tag{2.1.3} \]

\[ k = \frac{\omega}{c} n \]

We assume the index of refraction is uniform.

Take (2.1.1) into (2.1.2) and use the derivatives of \( E \), we can calculate that

\[ \nabla_t^2 \psi - j2k \frac{\partial \psi}{\partial z} + \frac{\partial^2 \psi}{\partial z^2} = 0 \tag{2.1.4} \]

Since \( k \) is a relative large number, we can neglect the last term, results in

\[ \nabla_t^2 \psi - j2k \frac{\partial \psi}{\partial z} = 0 \tag{2.1.5} \]

and this equation is the central equation for Gaussian beams. It is called the paraxial wave equation.

To reduce arithmetic, we take (2.1.5) into cylindrical coordinate

\[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \psi}{\partial r} \right) - j2k \frac{\partial \psi}{\partial z} = 0 \tag{2.1.6} \]
and define

\[ \psi_0 = \exp\{-j[P(z) + \frac{kr^2}{2q(z)}]\} \quad (2.1.7) \]

which is the wave function for the fundamental lowest-order TEM\(_{0,0}\) mode.

Take (2.1.7) into (2.1.6)

\[ \left\{ \frac{k^2}{q^2(z)}(q'(z) - 1) \right\} r^2 - 2k \left[ P'(z) + \frac{j}{q(z)} \right] r^0 \psi_0 = 0 \quad (2.1.8) \]

To make (2.1.7) a solution, the factors of each power of \( r \) should be zero, which means

\[ q'(z) = 1 \quad \text{(2.1.9)} \]

\[ P'(z) = -\frac{j}{q(z)} \]

From (a), we get

\[ q(z) = q_0 + z \quad \text{(2.1.10)} \]

Assume that \( q(z) \) is complex. We may find a place as \( z=0 \) to eliminate the real part of \( q_0 \), so that \( q_0 \) is imaginary, take it as \( q_0=jz_0 \), then

\[ q(z) = jz_0 + z \quad \text{(2.1.11)} \]

At \( z=0 \),

\[ q(z) = jz_0 \quad \text{(2.1.12)} \]

Substitute (2.1.11) into (2.1.7) at \( z=0 \),

\[ \psi_0(z = 0) = \exp(-\frac{kr^2}{2z_0}) \exp[-jP(z = 0)] \quad \text{(2.1.13)} \]

Since the first exponential term is real, the beam amplitude drops quickly with \( r \), from the peak value of 1 at \( r=0 \) to 0.368 at \( r=(2z_0/k)^{1/2} \) as shown in figure 2.1. The last \( r \) is the scale length of the beam.

\[ w_0^2 = \frac{2z_0}{k} = \frac{\lambda_0z_0}{n\pi} \quad \text{(2.1.14)} \]

This means the amplitude of the beam changes as \( \exp(r^2/w_0^2) \) at \( z=0 \).
Figure 2.1 Variation of the field in the transverse plane. Reprinted from [18]

In the former equations, the q factor shows as the inversion, so we calculate the inverse of q

$$\frac{1}{q(z)} = \frac{1}{z + jz_0} = \frac{z}{z^2 + z_0^2} - j \frac{z_0}{z^2 + z_0^2} = \frac{1}{R(z)} - j \frac{\lambda_0}{\pi \omega^2(z)} \quad (2.1.15)$$

Take (2.1.15) back into (2.1.7) we get

$$\psi_0 = \left\{ \exp \left[ -\frac{kz_0 r^2}{2(z^2 + z_0^2)} \right] \right\} \left\{ \exp \left[ -\frac{j k z r^2}{2(z^2 + z_0^2)} \right] \right\} \left\{ \exp \left[ -jP(z) \right] \right\} \quad (2.1.16)$$

Same with (2.1.13), the factor multiplying $r^2$ in the first exponential factor is the scale length, define as the spot size of the beam

$$w^2(z) = \frac{2(z^2 + z_0^2)}{kz_0} = \frac{2z_0}{k} \left[ 1 + \left( \frac{z}{z_0} \right)^2 \right] \quad (2.1.17)$$

Using $w_0$ to express $w(z)$, we get

$$w^2(z) = w_0^2 \left[ 1 + \left( \frac{\lambda_0 z}{\pi \omega_0^2} \right)^2 \right] \quad (2.1.18)$$

And $R(z)$ is shown as

$$R(z) = \frac{z^2 + z_0^2}{z} = z \left[ 1 + \left( \frac{z}{z_0} \right)^2 \right] = z \left[ 1 + \left( \frac{\pi \omega_0^2}{\lambda_0} \right)^2 \right] \quad (2.1.19)$$

Then we go back to calculate $P(z)$. 

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.1.png}
\caption{Variation of the field in the transverse plane. Reprinted from [18]}
\end{figure}
\[ P'(z) = \frac{-j}{q(z)} = \frac{-j}{z + jz_0} \]  
\[ jP(z) = \int_0^z \frac{dz'}{z' + jz_0} = \ln(z' + jz_0)|_0^z = \ln(z + jz_0) - \ln(jz_0) \]  
\[ jP(z) = \ln[1 - j\left(\frac{z}{z_0}\right)] \]  
\[ 1 - j\left(\frac{z}{z_0}\right) = \left[1 + \left(\frac{z}{z_0}\right)^2\right]^{1/2} - \exp[-j\tan^{-1}\left(\frac{z}{z_0}\right)] \]  

Since

\[ jP(z) = \ln[1 + \left(\frac{z}{z_0}\right)^2]^{1/2} - j\tan^{-1}\left(\frac{z}{z_0}\right) \]

we have

\[ jP(z) = \ln[1 + \left(\frac{z}{z_0}\right)^2]^{1/2} - j\tan^{-1}\left(\frac{z}{z_0}\right) \]

and \( \exp[-jP(z)] \) is

\[ e^{-jP(z)} = \frac{1}{\left[1 + (z/z_0)^2\right]^{1/2}} e^{j\tan^{-1}\left(\frac{z}{z_0}\right)} \]  

Then we can write down the complete equation for the fundamental or the lowest order TEM\(_{0,0} \) mode as

\[ \frac{E(x,y,z)}{E_0} = \left\{ \frac{w_0}{w(z)} \exp\left[-\frac{r^2}{w^2(z)}\right] \right\} \times \exp\left[-j\left[kz - \tan^{-1}\left(\frac{z}{z_0}\right)\right]\right] \times \exp\left[-j\frac{kr^2}{2R(z)}\right] \]  

The first term on the right is the amplitude factor, the second shows the longitudinal phase, and the last term shows the radial phase.

At \( r=w \), the beam amplitude decreased to \( 1/e \) times of its maximum where \( r=0 \).

Through propagation, the beam spot size is tendency to the dashed lines shown in figure 2.2 according to the equation

\[ w(z \gg z_0) = \frac{w_0z}{z_0} = \frac{\lambda_0z}{\pi n w_0} \]  

so the expansion angle is
\[
\frac{\theta}{2} = \frac{dw}{dz} = \frac{\lambda_0}{\pi w_0} \quad \theta = \frac{2\lambda_0}{\pi w_0}
\] (2.1.28)

This is the minimum angular of a beam with a diameter of \(2w_0\).

Figure 2.2 Spreading of a TEM0,0 mode. Reprinted from [18]

### 2.2 Temporal characterizations of ultrashort pulses

We discussed the spatial characterizations of the laser beams including propagation equations, amplitude variation with propagate distance and the expansion angle in the former part. In this section, we will focus on the temporal properties of the laser beam.

#### 2.2.1 Wavelength Bandwidth

For a transform-limited Gaussian pulse, the frequency bandwidth is

\[
\Delta \omega = \frac{4 \ln 2}{\tau}
\] (2.2.1)

where \(\tau\) is the FWHM.

It is more convenient to get the bandwidth in wavelength through directly measure the laser beam with spectrometers, so we will convert the frequency bandwidth into wavelength.
From
\[
\Delta \omega = \Delta \left( \frac{2\pi c}{\lambda} \right) = \frac{2\pi c}{\lambda_0^2} \Delta \lambda
\]
we have
\[
\Delta \lambda = \frac{2 \ln 2 \lambda_0^2}{\pi c \tau}
\]
where \( \lambda_0 \) represents the center wavelength.

The wavelength bandwidths over pulse widths centered in 800 nm wavelength are listed in table 2.1. And it’s more easily to see the spectral ranges and shapes from figure 2.3.

The 25 fs pulse comes from CPA has a much narrower bandwidth compares to the 5 fs pulse which is generated from hollow-core fibers, whose spectrum region is from 550 to 1050 nm.

Table 2.1 Bandwidth and pulse duration of Transform-Limited Gaussian Pulses

<table>
<thead>
<tr>
<th>( \tau ) (fs)</th>
<th>5</th>
<th>10</th>
<th>15</th>
<th>20</th>
<th>25</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \Delta \lambda ) (nm)</td>
<td>188.3</td>
<td>94.1</td>
<td>62.8</td>
<td>47.1</td>
<td>37.7</td>
</tr>
</tbody>
</table>
2.2.2 Temporal contrast of ultrashort pulses

First we define a temporal quality that the ratio between the intensity at the peak and at a delay $\Delta t$ as temporal contrast, which is a fundamental factor in laser-matter interaction. A 25 fs laser pulse with energy of around 1 mJ focused in a spot size of 1 $\mu$m which is the limit of diffraction at 800 nm results in a peak intensity higher than $10^{18}$ W/cm$^2$. With a temporal contrast of around 6 orders of magnitude, the intensity of the background pedestal is around $10^{12}$ W/cm$^2$, which is high enough for ionizing the atoms before the main ultrashort pulse arrives [20]. If plasma is generated at the same time due to this ionization, it will expand hydrodynamically and thus the subsequent main pulse has no chance to interact with a clean and steep plasma. The deformation on the plasma-vacuum interface has a deep influence on
high harmonic generation. Figure 2.4 depicts the canonical temporal profile of the output pulse from a CPA laser on logarithmic scale. The temporal profile is constituted with four parts in chronological order: a long pedestal (>100 ps), a short pedestal (tens of the pulse duration), pre-pulses and post-pulses. The first long pedestal accounts for the incoherent noise caused by the amplified spontaneous emission (ASE) which is mainly generated in the high gain amplification stages, thus it is called incoherent contrast. The second part, short pedestal, results from imperfect recompression after the stretcher and compressor, is called the coherent contrast. The pre-pulses and post-pulses are replicas of the main pulse generated from reflection or diffusion on optical surfaces and nonlinear effects.

![Figure 2.4 Canonical temporal profile of a CPA laser output pulse. It has long and short pedestals before the main pulse, and pulses on either hand of the main pulse on time scale. Reprinted from [21]](image)

### 2.3 Measurement methods of femtosecond laser pulses

Linear detectors can measure laser pulses of hundreds of fs. When it comes to generate attosecond pulse train, the driving NIR laser pulses can be wide as 100 fs, and for single isolated attosecond pulses, laser pulses are approximately 10 fs, which means the response
speed of linear detectors is too slow. Cross-correction method can be used to measure NIR pulses narrow as 4 fs, which uses isolated attosecond pulses, and is difficult to conduct. Then in 1990s, relatively simple methods based on nonlinear optics came out.

When measuring femtosecond lasers, we need to know the amplitude and the phase. Since we can easily measure the power spectrum by grating spectrometers, the only problem in fully characterizing the lasers is to measure the spectral phase. There are two common techniques for measuring the spectral phase, one is called frequency-resolved optical gating (FROG), and the other is spectral phase interferometry for direct electric-field reconstruction (SPIDER). FROG has also been extended into attosecond region.

2.3.1 FROG technique

FROG is an extension of the thorough autocorrelation technology constructed in the beginning of ultrafast laser era. After measurement, the result shows as a time-frequency spectrogram, and then by using an iterative algorithm we can extract the amplitude and spectral phase.

An autocorrelator can split a laser pulse into two. One pulse is delayed in time, then recombined with the other and get focused together into a second harmonic generation crystal. The second harmonic signal, which is a function of the former time delay, is the autocorrelation trace and can be used to reckon the pulse duration. Autocorrelators can be divided into two types, scanning autocorrelator and single-shot autocorrelator. The scanning
autocorrelator is used to measure reproducible pulses. The time delay is commonly introduced by a Michelson interferometer. In a single-shot autocorrelator, two finite sized (~1cm) laser beams propagate cross each other on a second harmonic crystal with an angle. An imaging detector is used to measure the time delay. CCD is commonly used as the imaging detector. Single-shot autocorrelator is suitable for measuring high-power laser pulses of which the repetition rate is low.

![Diagram of a FROG setup](image)

Figure 2.5 Schematic of a FROG setup. Reprinted from [22]

Figure 2.5 depicts a schematic of a typical FROG setup. When the second order signal which is the frequency-resolved autocorrelation trace generated from an autocorrelator propagates into a spectrometer, a two-dimensional FROG trace is formed, for which the coordinate system is wavelength over delay.

Since the conversion efficiency increases with the laser intensity, the second harmonic generation is also seen as an amplitude gating. It can be presumed that the two pulses from the autocorrelator are the unknown pulse that need to be measured and the gating pulse. The
unknown pulse with an electric field $E(t)$ propagates into the nonlinear medium and interacts with a gate $G(t-\tau)$, which is the gating pulse $E(t-\tau)$, and $\tau$ is the delay. The signal generated within this progress can be shown as

$$E_{\text{signal}}(t, \tau) = E(t)E(t - \tau)$$  \hspace{1cm} (2.3.1)

The spectral intensity $I_{\text{FROG}}(\omega)$ for different delays are measured by a spectrometer. The measured spectrogram is shown as

$$I_{\text{FROG}}(\omega, \tau) = \left| \int_{-\infty}^{+\infty} E(t)E(t - \tau)\exp(-i\omega t)dt \right|^2$$  \hspace{1cm} (2.3.2)

Figure 2.6 gives the characterization of a Ti:Sapphire CPA laser pulse measured by a single-shot FROG setup [19]. The autocorrelation trace is obtained by vertical integrating the signal. Specific phase retrieval algorithms are used to reconstruct the pulse shape and phase. The results are shown in figure 2.6(c) and 2.6(d). An advantage of FROG over autocorrelators is that by using FROG we don’t need to make assumptions, thus it’s more accurate.
Figure 2.6 Characterizing the Ti:Sapphire CPA laser pulse measured by FROG: (a) is the measured FROG trace, (b) is the reconstructed trace, (c) is the reconstructed pulse intensity and phase in the time domain and (d) is in the frequency domain. Reprinted from [19]

The theory of the iterative algorithm for extracting the laser field can be conclude as follow. According to equation (2.3.2), through iteration, the value of square root of the measured FROG trace replaces the magnitude of \( I_{\text{FROG}}(\omega,\tau) \). The algorithm uses an estimate for the intensity and phase of \( E(t) \) at each time step, which is determined by the minimization of the difference of \( E(t) \) between the measured and computed FROG trace. The algorithm keeps repeating until it reaches an optimal solution where the error is small enough. And this could take a really long time. In figure 2.6, the result from the iteration is shown in 2.6(b), which is very similar to the collected trace shown in 2.6(a). The validity of the phase retrieval
depends on the FROG error and the frequency marginal comparison. FROG error is defined as the per-pixel root-mean-square error of the FROG trace, and the frequency marginal comparison means comparing the measured and reconstructed intensity spectrum, which is illustrate in figure 2.6(d). Since the reconstructed power spectrum is very similar to the measured one, this phase retrieval is accurate.

2.3.2 SPIDER technique

SPIDER retrieves the spectral phase of an incident pulse through spectral shearing interferometry. It measures the interference between two different pulses centered in the same frequency which are separated in time. This pulse pair is spectrally sheared. Nonlinear effects provide a general approach to totally shearing a pulse pair by a frequency $\Omega$. Basically, this can be done by sum frequency generation between the field we want to shear $\tilde{E}(\omega)$ and a monochromatic frequency $\Omega$, and the result is a sheared field $\tilde{E}(\omega + \Omega)$. This progress is shown in figure 2.7. The monochromatic frequency $\Omega$ can be easily obtain by a strongly chirped ultrashort pulse whose instantaneous frequency, which is approximately linear to time, doesn’t change during nonlinear interaction with the pulse. The magnitude of the spectral shear generated here depends on two parameters: time delay between the two pulses and the chirp amount of the stretched pulses. The shear value is

$$\Omega = -\frac{\tau}{2\phi_2} \quad \text{(2.3.3)}$$

where $\phi_2$ is the second order dispersion of the chirped pulse.
The interference of the two sheared pulses is given by

\[
I_{\text{SPIDER}}(\omega) = |E(\omega)|^2 + |E(\omega + \delta\omega)|^2 + 2|E(\omega)E(\omega + \delta\omega)| \times \cos[\phi(\omega + \delta\omega) - \phi(\omega) + \omega\tau]
\]  

(2.3.4)

which is the SPIDER signal.

Figure 2.7 Using nonlinear interaction with a chirped pulse to generate two sheared copies of the input pulse. Reprinted from [22]

Fourier Transform Spectral Interferometry (FTSI) is used to algebraically extract the phase difference between the two spectrally sheared copies, then apply a Fast Fourier Transform, filter one of the interference terms then another Fast Fourier Transform to get back to the initial situation, result in the performing of the extraction of the phase difference

\[
\phi(\omega + \delta\omega) - \phi(\omega) + \omega\tau
\]  

(2.3.5)

\subsection*{2.4 A typical method to generate femtosecond pulses: mode-locking}

Mode-locking is the most commonly used and effective method to generate ultrashort pulses. For normal lasers, each longitudinal mode has no correlation in phase and is random in time, results in a noisy intensity output, as it shows in figure 2.8(a). The output laser
intensity fluctuates irregularly with time, and the total light intensity is equal to the simple superposition of the strength of each longitudinal mode. While in figure 2.8(b), when the phases get fixed, the longitudinal modes can strengthen and weaken each other and finally generate a single pulse, whose pulse duration is much shorter an amplitude is much larger than the longitudinal modes themselves.

Figure 2.8 (a) Output amplitude over time of a CW laser. The signals have a random distribution of axial-mode amplitude and phases. (b) Output amplitude over time of a mode-locked laser. The phases of the signals are locked, which shows a Fourier-limited pulse. Reprinted from [23]

Mode-locking, which is also known as phase-locking, keeps the phase difference between the longitudinal modes constant. This leads to a high output power. If the phases of 2N+1 longitudinal modes are locked, the modes are coherently superposed to form a periodic pulse sequence and thus the pulse peak intensity is 2N+1 times of the average intensity of a free-running laser.
When $N$ axial-modes are in phase, the output intensity of the laser pulses is shown as the follow equation:

$$I(t) \propto |E(t)|^2 = E_0^2 \cdot \frac{sin^2\left[\frac{1}{2}(2N+1)(\Omega t + \beta)\right]}{sin^2\left[\frac{1}{2}(\Omega t + \beta)\right]}$$

(2.4.1)

The output pulse intensity is described as a periodic pulse train with a period of $T=2\pi/\Omega=2L/c$. That is, the interval between the main pulses is exactly the round-trip time of a pulse propagating inside a cavity, which means the mode-locked laser shows the back and forth motion of a pulse inside the cavity. Figure 2.9 shows three frequency components get in phase at $t=0$ and $t=T$ where high peak pulses are generated, while between the pulses the amplitudes are counteracted.
Active mode-locking, as the name implies, can be achieved by placing a periodic amplitude loss modulator or periodic phase-shift modulator within the cavity and making the modulation duration equal to the round-trip time. The former is called amplitude modulation mode locking and the latter is called frequency or phase modulation mode locking. The theory of active mode-locking is that when a periodic loss modulator is placed in the laser cavity, since the side frequency interval stimulated by the amplitude modulator is the modulation frequency, if we want the phase of the longitudinal mode to be synchronized, we
just need to keep the excitation frequency coincides with the longitudinal mode (as close as possible), then due to injection locking, the phase synchronization is achieved. The key is that the modulation frequency must be equal to the frequency interval of longitudinal mode propagating inside the cavity.

For active mode-locking, the pulse width is fundamentally determined by the modulation frequency, which is limited by the speed of the electronics, and thus cannot be used to produce ultrafast pulses.

Passive mode-locking doesn’t need external adjustment, there is a periodic loss modulator in the cavity, which is formed automatically in the cavity, irrespective of external factors. In passive mode-locking, a strong pulse is absorbed less, which means the loss is low, then eventually oscillation is generated, while a weaker pulse is absorbed more, which means the loss is high, and eventually it is annihilated. For a pulse itself, the rising and falling edges have lower intensity, while the middle has higher intensity. Therefore, when the pulse propagates through the absorber, the front edge is absorbed, the middle portion passes due to absorption saturation (bleaching), and the rear edge is also absorbed in fast saturation absorption or get compressed by other methods in slow saturation absorption, results in the compression of the pulse.

For passive mode-locking, the steepness of the temporal gate is determined by the pulse itself. During each round-trip the pulse gets narrowed until ultimately limited by the available gain bandwidth, and thus passive mode-locking allows ultrafast pulses generation.
SPM (self-phase modulation) is a significant limitation due to the strictly confined optical modes. The generation method of attosecond pulses will be explained in the subsequent chapter.
3 THEORETICAL INTRODUCTIONS TO HIGH HARMONIC GENERATION

3.1 Background of attosecond optics

3.1.1 Atomic unit of time

In molecule scale, the rotational periods are of picoseconds, while the vibrational periods are of femtoseconds, as shown in figure 3.1. These processes are broadly observed and studied by ultrafast lasers operated in IR, visible and UV spectrum. When it comes to much smaller particles, such as electrons, traditional picosecond and femtosecond lasers cannot be applied due to the much shorter motion periods.

Figure 3.2 is a symbolic filmstrip of an electron rotates around a proton. Actually, the atomic unit of time is the characteristic timescale of the motion processes of electrons in microscopic particles. And the observation and study of such processes take place with the help of attosecond pulses.

![Figure 3.1 Timescale in dynamics of atoms and molecules](image)
The applications in molecule scale promoted the development of ultrafast lasers in the NIR and visible region, while the time scale of optical pulses reached the limit in 1987. Though great efforts were made, there were no substantive progress until in 2001, XUV pulses with sub-fs duration were generated by fs NIR lasers interacting with atoms [24]. A transform-limited Gaussian pulse with FWHM of around the atomic unit of time corresponds to approximately 75 eV FWHM power spectrum. This is much broader than visible light range, that means attosecond pulses are in the XUV or X-ray ranges.
Figure 3.3 Power spectrum of an attosecond pulse with the FWHM of around the atomic unit of time.

3.1.2 Mathematica of attosecond Gaussian pulses

A coherent pulse can be expressed by the equation

$$\varepsilon(t) = E(t) \cos[\omega_0 t + \varphi(t)]$$  \hspace{1cm} (3.1.1)

Complex representation which

$$\varepsilon(t) = E(t) \exp\{+i[\omega_0 t + \varphi(t)]\}$$  \hspace{1cm} (3.1.2)

is more convenient. The peak intensity $I_0$ is shown by

$$I_0 = \frac{1}{2} \varepsilon_0 cE_0^2$$  \hspace{1cm} (3.1.3)

And for Gaussian pulses,

$$\varepsilon(t) = E_0 \exp[-at^2] \exp[\omega_0 t + \varphi(t)]$$  \hspace{1cm} (3.1.4)
where $E_0$ is the peak amplitude. The pulse duration, which is the FWHM, is

$$\tau = \frac{2\ln 2}{a} \tag{3.1.5}$$

then

$$\varepsilon(t) = E_0 \exp \left[ -2 \ln 2 \left( \frac{t}{\tau} \right)^2 \right] \exp \{ i[\omega_0 t + \varphi(t)] \} \tag{3.1.6}$$

The intensity profile of the pulse is

$$I(t) = I_0 \exp \left[ -4 \ln 2 \left( \frac{t}{\tau} \right)^2 \right] \tag{3.1.7}$$

In (3.1.5), $a$ is the pulse width parameter. Chirp means the instantaneous frequency of a pulse changes over time. When $\varphi(t) = bt^2$, where $b$ is the chirp parameter, the chirp is linear.

Then the instantaneous frequency can be expressed as

$$\omega(t) = \frac{d}{dt} [\omega_0 t + \varphi(t)] = \omega_0 + 2bt \tag{3.1.8}$$

When $b>0$, the pulse is positively chirped, red components are in the leading edge, which means the instantaneous frequency of the envelope of pulse increases with time, and when $b<0$, the pulse is negatively chirped, blue components are in the leading edge, and the instantaneous frequency decreases with time. Figure 3.4 shows chirp of two Gaussian pulses centered in 8 nm, with a duration of 100 as. (a) Positively chirped ($b=+0.5 \times 10^{-3}$ as$^{-2}$) (b) Negatively chirped ($b=-0.5 \times 10^{-3}$ as$^{-2}$).
Figure 3.4 Depict of chirped pulse. (a) Positively chirped (b) Negatively chirped.

In frequency domain, the spectral amplitude for a linearly chirped Gaussian pulse can be expressed as

\[ U(\omega) = U_0 \exp\left[-2 \ln 2 \frac{(\omega - \omega_0)^2}{\Delta \omega^2}\right] \]  

(3.1.9)

The spectral phase is given by

\[ \varphi(\omega) = -\frac{1}{4} \left(\frac{b}{\alpha^2 + b^2}\right) (\omega - \omega_0)^2 \]  

(3.1.10)

The FWHM of the power spectrum is

\[ \Delta \omega = \frac{4 \ln 2}{\tau} \sqrt{1 + \left(\frac{b}{\alpha}\right)^2} \]  

(3.1.11)

From the uncertainty principle in quantum mechanics, we have

\[ \tau \cdot \Delta \epsilon = 4 \ln 2 \sqrt{1 + \left(\frac{b}{\alpha}\right)^2} \hbar \]  

(3.1.12)

For transform-limited Gaussian pulses, \( \tau[\text{as}] \cdot \Delta \epsilon[\text{eV}] = 1825 \), thus if we want to get pulse shorter than 1 fs, the intensity of the spectral bandwidth of the pulse need to be larger than 1.8 eV.
3.1.3 Harmonic Generation

For 100 as to 1 as pulse duration, the FWHM bandwidth is in the 18 eV to 1800 eV region. This bandwidth is so broad that people haven’t found any laser gain media that could support it till now. To generate attosecond pulses, we can use another way through harmonic generation.

Take second harmonic generation as an example. The electric field amplitude of the generated pulses which has doubled frequency is shown as

\[ E_{2\omega}(t) \propto E_{\omega}^2(t) \]  \hspace{1cm} (3.1.13)

The intensity is shown as

\[ I_{2\omega}(t) \propto I_{\omega}^2(t) \]  \hspace{1cm} (3.1.14)

And for Gaussian pulses, we can write that

\[ e^{-4 \ln 2 \left( \frac{t}{\tau_{2\omega}} \right)^2} = \left[ e^{-4 \ln 2 \left( \frac{t}{\tau_{\omega}} \right)^2} \right]^2 = e^{-8 \ln 2 \left( \frac{t}{\tau_{\omega}} \right)^2} \]  \hspace{1cm} (3.1.15)

Where \( \tau_{\omega} \) is the FWHM of the fundamental pulse, and \( \tau_{2\omega} \) is of the second harmonic pulse.

The relationship between these two factors is

\[ \tau_{2\omega} = \frac{\tau_{\omega}}{\sqrt{2}} \]  \hspace{1cm} (3.1.16)

Similarly, for q order nonlinear process in the perturbative regime,

\[ \tau_{q\omega} = \frac{\tau_{\omega}}{\sqrt{q}} \]  \hspace{1cm} (3.1.17)

Then we can compress a 2 fs pulse to a 200 as pulse through 100th order harmonic generation.
There is a problem while using this method that the conversion efficiency drops dramatically with harmonic order, as shown in figure 3.6. The photon flux could be extremely low for 100th harmonic order, thus it can’t be applied to practical experiments.

Perturbative HHG occurs when the intensity of the pulses is less than $10^{12}$ W/cm$^2$, which is less than the electric field inside the atom, and when the intensity is higher, many commonly used solid materials will be damaged.

![Intensity spectrum of harmonics generated in the perturbative regime.](image)

Figure 3.5 Intensity spectrum of harmonics generated in the perturbative regime.

In 1987, McPherson and colleagues discovered a new kind of HHG phenomena [25], that when a linearly polarized driving pulse with a short duration and an intensity of $10^{14}$W/cm$^2$ interacted with noble gases, which have large binding energies, odd harmonics of tens or even hundreds in order were generated. After quickly decay of the intensity of some of the initial harmonics, a plateau formed where the intensity remained approximately
constant over many orders. Finally, the signal cut off at the highest order, as shown in figure 3.7. For initial NIR lasers, photon energy is around 1.5 eV, thus the range of the five harmonics in the plateau is 6 eV, which is broad enough for attosecond pulses.

![Intensity spectrum of harmonics generated in the nonperturbative regime.](image)

Figure 3.6 Intensity spectrum of harmonics generated in the nonperturbative regime.

The maximum photon energy can be calculated by

\[
\hbar \omega_{x,max} = I_P + 3.17U_p \quad (3.1.18)
\]

\[
U_p [eV] = 9.33 \times 10^{14} I_0 \lambda_0^2 \quad (3.1.19)
\]

3.2 High harmonic generation

3.2.1 Three-step model

In 1993, scientists came up with a semi-classical model to describe the mechanism of HHG, which is widely known as the three-step model. This model reveals the dynamics of the bound electrons of noble gas atoms, which are first ionized and tunnel through the atomic
barrier, then propagate in the electric field and may return to the ion, finally recombine with the ion and emit harmonics, as shown in figure 3.8.

Figure 3.7 Three-step model. (a) Tunneling ionization, (b) acceleration in the continuum electric field, and (c) recombination of the electron and the atom.

### 3.2.2 Single atom response

Consider a linearly polarized laser field interacts with a hydrogen atom. The potential with no external laser electric field is depicted in figure 3.9. As the laser field is applied, when the intensity reaches around $10^{14}$ W/cm², the field near the maximum of each oscillation is comparable to the atomic Coulomb field. Then the potential well is transformed into a potential barrier by the superposition of the laser field and the Coulomb field, as depicted in figure 3.10.
Figure 3.8 The potential of a hydrogen atom. The dashed line in the middle indicates the ground state.

Figure 3.9 The Coulomb barrier formation.

The dynamics of the electron conform to the three-step model. The electron in the ground state is possible to tunnel through the barrier, which is the first step. Then the electron
moves in the laser field, which is the second step. The electron first moves away from the
nucleus, and as the laser field changes to the opposite direction, the electron is driven back by
the electric force and gain kinetic energy of tens or even hundreds of electron volts. Finally,
the electron and the initial ion recombine and emit a photon, which is the third step.

However, not all of the ionized electrons would return to the ion. If the initial laser
beam is not linearly polarized, the trajectory of an ionized electron won’t be a complete loop,
and even for a linearly polarized laser beam, the possibility of the electrons to return
determines by the chase of the laser field by the time the tunneling ionization occurs. The
maximum kinetic energy of the ionized electron could be $3.17 \ U_p$, which is the
ponderomotive potential, represents the average energy of a charged particle in an
electromagnetic wave which is given by

\[
U_p = \frac{1}{4} \frac{q^2 |E|^2}{m \omega^2} \approx 9.33 \times 10^{-14} I \lambda^2 \ [\text{eV}]
\]

where $q$ is the charge and $m$ is the mass of the particle, $E$ is the electric field and $\omega$ is the
frequency, $I$ is the intensity, $\lambda$ is the wavelength. Since it’s inversely proportional to the mass,
the ponderomotive potential of an electron is much larger than an ion.

By the time the electron returns and recombines with the ion, it releases both its kinetic
energy and its ionization energy, which form high-order harmonics. Thus the highest-order
harmonic energy can be written as

\[
\hbar \nu_c = I_p + 3.17 U_p
\]

where $I_p$ is the ionization potential. $\nu_c$ is called the cutoff frequency [26].

This three-step process is depicted in figure 3.11 [27]. The electric trajectory is shown
in the top figure with black lines, and the gray line represents the electric field. Whether the trajectory is long or short depends on the comparison of the time that electron released with 0.05 T₀. Larger trajectory is called long trajectory. The electrons with short trajectories recombine with the ions when the laser field is still negative, and the electrons with long trajectories are decelerated before recombination by the positive electric field, thus they both have less energy than the cut off energy.

Figure 3.10 Illustration of HHG using three-step model. Reprinted from [27].

Long trajectory can be depressed through phase matching and spatial filtering, thus only the electrons with short trajectories can reach the detector. The return time of such electrons extends from t = 0.25T₀ to t = 0.7T₀, which is 1.2 fs and 600 as at FWHM. Therefore, short trajectory is possible to be applied to generate attosecond pulses.
3.2.3 Phase matching

We have discussed single atom physics in the former part, while high harmonic generation is a multi-particle process and the total effects need to be concerned. It’s necessary to set the phase-conditions of the atoms to increase the corresponding radiation and thus obtain efficient HHG.

The total phase vector can be written as

\[ k(\omega) = k_f(\omega) + k_{\text{disp}}(\omega) + k_{\text{plas}}(\omega) + k_{\text{geom}}(\omega) \]  (3.2.3)

where \( k_f(\omega) = \frac{2\pi\omega}{c} \) is the wave vector in free space, \( k_{\text{disp}}(\omega) \) is the wave vector shift due to dispersion in a neutral medium, \( k_{\text{plas}}(\omega) \) is due to dispersion in a plasma and \( k_{\text{geom}}(\omega) \) is due to geometric dispersion.

The wave vector mismatch of the qth harmonic order is written as

\[ \Delta k = qk(\omega_0) - k(q\omega_0) \]  (3.2.4)

were k is the wave vector, q is the harmonic order and \( \omega_0 \) is the frequency of the driving laser [28]. The phase-mismatch in free space is 0, so the rest three terms need to be considered.

The wave vector shift of the phase-mismatch due to dispersion in a neutral medium can be written as

\[ \Delta k_{\text{disp}}(\omega) = [n(\omega_0) - n(q\omega_0)] \frac{q\omega_0}{c} \]  (3.2.5)

where n is the refractive index, which mainly contributes to this kind of phase-mismatch. The refractive index is larger than one for a 800 nm driving laser and smaller than one for the XUV radiation, thus this wave vector mismatch is a positive value.

As explained in the former section, only part of the ionized electrons will recombine
with the ion and generate high harmonic radiation. The electrons that remain ionized results in the variation of the refractive index that can be written as

\[ n_{\text{plas}}(\omega) = \sqrt{1 - \left(\frac{\omega_p}{\omega}\right)^2} \]  
\[ \omega_p = \sqrt{\frac{e^2 N_e}{\epsilon_0 m_e}} \]

where \( \omega_p \) is the frequency in the plasma. The wave vector shift can be written as

\[ \Delta k_{\text{plas}}(\omega) = q k_{\text{plas}}(\omega_0) - k_{\text{plas}}(q \omega_f) = \frac{\omega_p^2 (1-q^2)}{2q \omega_0} \]

which is negative when \( q > 1 \).

The geometric dispersion is caused by the driving laser. The focused Gaussian laser beam contains an additional phase term along \( z \)-direction, which is widely known as Guoy phase. The wave vector for \( z < < z_R \) is written as

\[ k_{\text{geom}}(z) = \frac{d\zeta(z)}{dz} \approx \frac{1}{z_R} \]

Then the wave vector shift is

\[ \Delta k_{\text{geom}} = q k_{\text{geom}}(\omega_0) - k_{\text{geom}}(q \omega_0) = \frac{q-1}{z_R} \]

Thus the total phase mismatch can be expressed by

\[ \Delta k_{\text{total}} = [n(\omega_0) - n(q \omega_0)] \frac{q \omega_0}{c} + \frac{\omega_p^2 (1-q^2)}{2q \omega_0} + \frac{q-1}{z_R} \]

where the first term of the right is positive, the second is negative and the third is positive.

To achieve phase-matching, the phase-mismatch introduced by the electrons need to counteract the phase-mismatch due to dispersion mentioned above [29].
4 TABLETOP COHERENT SOFT X-RAY SYSTEM

This thesis mainly develops a tabletop coherent soft X-ray system as a light source for high resolution X-ray imaging experiments. We did some designs to fit specific requirements of two different experiments, circularly polarized soft X-ray generation for X-ray holography in magnetic media with high spatial and temporal resolution and broadband XUV and soft X-ray ptychography for attosecond high resolution imaging.

4.1 Driving Laser System

Our laser system for circularly polarized soft X-ray generation is a Ti:Sapphire laser capable for generating 800 nm wavelength pulses with duration of 20-30 fs, power of 2W. The laser system for ptychography is a three-stage amplification Ti:Sapphire laser with an OPCPA system, the output 1.7 μm pulses have duration of 10-12 fs with power of 1.8 W.

4.2 Soft X-ray Generation and Measurement System

4.2.1 Circularly Polarized HHG

Figure 4.1 is the setup for circularly polarized soft X-ray generation for X-ray holography in magnetic media with high spatial and temporal resolution. The driving laser propagates through the lens and focused on Barium Borate (BBO), then second harmonic which has reversed polarization is generated and propagate together with fundamental pulses. The Calcite pair is used to optimize the overlap of the two driving pulses, which works together with the λ/4 plate to transfer the two pulses into counter-rotating polarized. The
driving pulses interact with the noble gas flowing through the gas cell. XUV to soft X-ray pulses are generated through this process. The Al filter applied here is to block the NIR pulses and let soft X-ray pulses pass through. Then a spherical grating is used as a spectrometer, and the signals are collected by the CCD.

Figure 4.1 Schematic of the experimental device system for circularly polarized soft X-ray generation for X-ray holography in magnetic media with high resolution.

4.2.2 Broadband soft X-ray Ptychography

Figure 4.2 is the setup for broadband XUV and soft X-ray ptychography for attosecond high resolution imaging. The driving IR laser comes from OPCPA, propagates through a lens focusing system, which is protected by shelters due to the influence on laser quality of air flux, then interacts with noble gas flowing through the gas cell. The generated X-rays propagate together with the initial IR laser. The Ag filter is applied here to block the IR laser and let soft X-ray pulses pass through. The toroidal mirror is used to focus XUV and X-ray which cannot be focused by normal optical focusing systems due to the extreme short wavelength. The gate valve is used to make it possible to replace devices and optimize the beam in the left or right parts of the gate valve separately since this is a vacuum system. In
the sample chamber, the soft X-ray pulses firstly pass a pinhole, then illuminate the sample placed behind the pinhole. The diffraction pattern is collected by a CCD camera which has a short pin on it to block 0-order harmonic.

Two turbos are installed on the gas cell chamber and the sample chamber to create a vacuum. A barometer is installed on the tube connector in front of the gas cell to accurately measure the gas pressure in the chamber. The gas cell and sample are placed on 3-axis picomotor stages, and the toroidal mirror is placed on a 2-axis picomotor mirror mount which make optimization much more accurately. A chiller connected with a chip is mounted on the sample chamber, so that the residual oil in the air inside the chamber will deposit on the cold chip rather than the camera.

Figure 4.2 Schematic of the experimental device system for broadband XUV and X-ray ptychography for attosecond high resolution imaging.

Figure 4.3 Experimental setup through Solidworks.
5 EXPERIMENTS AND RESULTS

First, I will introduce some theories behind the HHG experiments.

The HHG process efficiency mainly depends on these factors: the density of atoms, the length of the nonlinear medium, the absorption of generated harmonics by medium and the phase velocity matching between the fundamental and harmonic fields. Thus the parameters of the gas cell will dramatically affect the efficiency of the HHG process. Gas cells can be divided into three types: gas jet, finite gas cell and infinite gas cell. Gas cells have longer medium lengths and higher atomic densities than gas jet, which would lead to higher conversion efficiencies, while longer medium length will also cause more absorption of harmonics [30]. Figure shows the total photocurrent as a function of laser intensity from these three kinds of gas cells in optimum pressure, which are 43 Torr for the semi-infinite gas cell (SIGC), 7 Torr for the finite gas cell (FGC) and 3.5 Torr for the pulsed valve (PV) which is a gas jet. It can be easily seen that SIGC can generate the highest photocurrent. Although no saturation is observed until the maximum intensity is reached in gas jet, the photocurrent is still much lower than using gas cells. In our experiments, we also tried these HHG sources, through comparing of the results, we get similar conclusions.
Figure 5.1 Schematic of total photocurrent as a function of laser intensity for three different kinds of gas cells. Reprinted from [30].

5.1 Circularly Polarized Soft X-ray Generation for X-ray Holography

5.1.1 Argon Circularly Polarized HHG

Figure 5.2 depict harmonics generated from a 0.3 mm gas jet. It’s easy to see high intensity signals through Argon, even with the gas jet. It shows that the third harmonic is missing in every three harmonics. We can explain this phenomenon through conservation of energy and photon spin angular momentum [31]. For circularly polarized harmonics with frequency $\omega_c$, from energy conservation we have

$$\omega_c = n\omega_1 + l\omega_2$$  \hspace{1cm} (5.1.1)

which means the harmonics are generated from $n$ photons with a frequency of $\omega_1$ and $l$ photons with a frequency of $\omega_2$. To reach photon spin angular momentum conservation we
need to keep \( l = n \pm 1 \), thus

\[
\omega_c = n\omega_1 + (n \pm 1)\omega_2 \tag{5.1.2}
\]

This leads to the phenomenon that adjacent harmonics have counter-rotating circular polarizations. The photon energy difference is proportional to \( \omega_1 - \omega_2 \) for the nearest harmonics of two nearest pairs, and \( \omega_1 + \omega_2 \) for harmonics of adjacent pairs. If we define \( \omega_1 = q\omega_2 \), where \( q \) can be any number, we can write (5.1.2) as

\[
\omega_c = n(q + 1)\omega_2 \pm \omega_2 \tag{5.1.3}
\]

For the simple case of HHG driven by initial pulses and second harmonic with frequency of \( \omega \) and \( 2\omega \), which means \( \omega_1 = 2\omega_2 \), we have

\[
\omega_c = (3n \pm 1)\omega_2 \tag{5.1.4}
\]

which results in this characteristic spectrum that every third harmonic order is missing.

Figure 5.2 Signals using 0.3 mm Jet.

5.1.2 Helium Circularly Polarized HHG

Figure 5.3 shows the experiment device system of circularly polarized HHG through
Helium. The pink pulse shows the reflectivity over photon energy of the narrow band XUV mirror pair. This mirror pair is used to reflect harmonics with photon energy of 58 eV and 60 eV, as shown in the small figure below. The red arrow means this harmonic is right-circularly polarized signal, while the green one means it’s left-circular signal. The complete result is shown in figure 5.11.

Figure 5.3 Circularly polarized HHG system using Helium.

Similar results can also be seen in figure 5.4 [32]. Figure 5.4 (A) describes the circularly polarized harmonics were generated by mixing a NIR field with a wavelength of 780 nm with its second harmonic with a wavelength of 390 nm in a 30 Torr Ar-filled waveguide with a length of 1 cm. And figure 5.4 (B) provides the function of intensity over harmonic orders, in which we can see that the 13th and 16th orders are right circularly polarized (RCP), 14th and 17th orders are left circularly polarized (LCP), while 15th and 18th orders are missing.
Figure 5.4 Characterizing circularly polarized HHG. (A) Counter-rotating beams from a Ti:Sapphire laser, 780nm RCP fundamental pulse and 390 nm LCP second harmonic, focus into an Ar-filled waveguide. The orientation of the circularly polarized HHG field can be rotated by tuning temporal delay $\tau_{RB}$ between the counter-rotating beams. (B) Circularly polarized HHG spectrum observed by an XUV spectrometer. Reprinted from [32]

Figure 5.5 depicts the high-order-harmonic spectrum for $\bar{\omega} = 0.05$ and $F_0 = 0.1$. Here, for a fixed frequency $\bar{\omega}$ and linearly polarized field strength $F_0$, any bicircular driving scheme is characterized by parameter $\alpha$. $\bar{\omega}$ is the mean frequency of the counter-rotating fields, which is defined as

$$\bar{\omega} = \frac{\omega_1 + \omega_2}{2} = \omega_1 + \alpha = \omega_2 - \alpha$$  \hspace{1cm} (5.1.5)

where $\omega_1$ is the frequency of RCP field and $\omega_2$ is of LCP field, and

$$\alpha = \frac{\omega_2 - \omega_1}{2}$$ \hspace{1cm} (5.1.6)

For $\alpha = 0$ in figure 5.5 (a), the driving pulses are linearly polarized, and the high harmonics in the y direction are zero. Thus, the RCP signals are equal to LCP signals, and the expected odd harmonic orders can be clearly observed in the plateau of the spectrum. As shown in (b) to (d), with the increase of the value of $\alpha$, the plateau gets depressed. This effect can be explained as follows. For higher harmonics, higher gain energy is required so the electron needs to be further away from the nucleus in the first step of the three-step modal.
However, as the distance getting further, the $L_z$ term gets larger, which is the factor of angular momentum corresponding to the rotation around the $z$ axis [34], and results in the acceleration of the electron in $y$ direction. This acceleration will cause a deflection and increase $y$ elongation and reduce $x$ elongation. Since the energy gained by the electron through electric field is proportional to only the elongation of $x$ component, it will decrease through this process. And this also gives answer to that counter-rotating drivers are better applied in circularly polarized HHG than corotating drivers. In the corotating scheme, $\alpha$ is defined by the average of the frequencies of the two drivers. This implies the $\alpha$ value is much larger than of the counter-rotating driver and results in a much stronger reduce of the plateau.
Figure 5.5 High-order-harmonic spectrum. The green curve is the RCP spectrum, and the orange curve is the LCP spectrum. (a) Both curves are on top since the signal in y direction vanishes due to $\alpha = 0$. Reprinted from [33]

Figure 5.6 is HHG signals using a 5 mm gas cell. An edge can be seen as the signal cut off, which is the Al edge. As shown in figure 5.7, the Al edge is at around 80 – 150 eV for a thickness of 0.2 μm. For photon energy of 80 eV, the harmonic order is around 50th. Figure 5.8 is HHG signals at lower energy. There is no edge appears here.
Figure 5.6 Signals using 5 mm cell at Al edge

Figure 5.7 Al filter transmission for XUV and soft X ray. Reprinted from CXRO X-Ray Interactions with Matter database.
Figure 5.8 Signals using 5 mm cell at lower energy.

Figure 5.9 is the HHG signals using an 8 mm gas cell. The signals are stronger than using the 5 mm cell.

Figure 5.9 Signals using 8 mm cell.

Figure 5.10 is the pressure scan from 50 Torr to 370 Torr. Firstly, as the gas pressure increase, the intensity of harmonics increases dramatically due to the increase of the atom
density, then reaches the maximum at approximately 230 Torr. The signals are saturate, so we
changed the integration duration from 10 s to 8 s. After reaching the maximum, the intensity
starts to decrease due to large absorption. The signal almost disappears after 370 Torr. Then
we do a position scan at 230 Torr from 6.75 mm to 8.75 mm. The results are shown in figure
5.11. The maximum appears at 8 mm. Figure 5.12 is the result of X-ray intensity with
arbitrary unit as a function of photon energy from 50 eV to 70 eV through Helium at 230
Torr.

Figure 5.10 Pressure scan from 50 Torr to 370 Torr.
Figure 5.11 Position scan at 230 Torr with position from 6.75mm to 8.75 mm.

Figure 5.12 X-ray intensity over photon energy through Helium at 230 Torr.
5.1.3  Neon Circularly Polarized HHG

Figure 5.13 and figure 5.14 are signals through Neon, similar to signals shown before. From figure 5.15 which depicts the pressure scan through Neon from 50 Torr to 290 Torr, we find that the maximum of the harmonic intensity appears at around 170 Torr, which means high harmonics are easier to be absorbed by Neon. The maximum through Neon is lower than through Helium, which shows a lower HHG efficiency.

![Image](image.jpg)

Figure 5.13 Signals using 5 mm cell at Al edge
Figure 5.14 Signals using 8mm cell with 8s integration.

Figure 5.15 Pressure scan from 50 Torr to 290 Torr.

5.2 Broadband XUV and Soft X-ray Ptychography

Figure 5.16 is a schematic of ptychography technique. In this experiment, we use a
pinhole as the order sorting aperture, whose information is observed by SEM shown in figure 5.17.

Figure 5.16 Schematic of Ptychography.

Figure 5.17 Graph of the pinhole information from SEM.

Figure 5.18 shows the IR diffraction pattern and HHG signal through a 50 μm pinhole, while figure 5.19 shows the green He-Ne beam diffraction pattern and HHG signal through a
5 μm pinhole.

Figure 5.18 Graphs from 50 μm pinhole. Left shows IR diffraction pattern log plot and right shows HHG signal in log scale.

Figure 5.19 Graphs from 5 μm pinhole. Left shows green He-Ne beam diffraction and right shows HHG signal in log scale.

Figure 5.20 is a standard CDI experiment steps flowchart.

Our experiments are still in progress.

Figure 5.20 CDI experiment steps.
6 CONCLUSIONS

This thesis starts from the methods of generating ultrashort pulses, explains the HHG process, which is based on the three-step model, and shows the experimental device systems and the results. A tabletop coherent soft X-ray source setup is designed and constructed for two applications: circularly polarized soft X-ray generation for X-ray holography in magnetic media with high resolution and broadband XUV and soft X-ray ptychography for attosecond high resolution imaging. We mainly discuss the results from experiments of the first application. Gas jet, 5 mm gas cell and 8 mm gas cell were used for Argon, Helium and Neon HHG processes. The phenomenon that every third order missing which is related to the circularly polarized HHG process is observed. We find that gas cell has better results than gas jet, and the longer gas cell is better. It’s easy to generate high harmonics through Argon, while it’s hard through Neon. We do pressure scan and find that for Helium, the best gas pressure for HHG process is 230 Torr, and 170 Torr for Neon. We also provide some figures of the IR diffraction pattern and HHG signals through the pinhole using ptychography technique. This experiment is still in progress.
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