EXTREME ULTRAVIOLET POLARIZATION OPTICS FOR POLARIMETRY OF STRUCTURED HIGH HARMONICS

by

Nathaniel Morgan Westlake
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Golden, Colorado
Date ________________________

Signed: ______________________

Nathaniel Morgan Westlake

Signed: ______________________

Dr. Charles Durfee
Thesis Advisor

Golden, Colorado
Date ________________________

Signed: ______________________

Dr. Frederic Sarazin
Professor and Department Head
Department of Physics
High Harmonic Generation (HHG) is a leading way to generate table-top coherent extreme ultraviolet (EUV) and attosecond pulses. Polarization characterization in this spectral range is important to understand the phenomena of HHG and quantify the light used for experiments. Furthermore, polarization control allows for polarization-dependent experiments in the EUV. This thesis proposes and demonstrates the usage of reflection-based polarization optics to act as a polarizer and a quarter wave retarder for 44 nm light, the 9th harmonic of 400nm. The design process for these polarization optics, so-called k-mirrors, in the EUV wavelength range is explained with possible design extensions for other wavelengths.

These polarization optics in EUV are extended with full-beam polarimetry of harmonic light generated with structured illumination. Characterization of the fundamental light’s structured illumination and polarization profile combined with similar characterization of the harmonics can reveal insights into the HHG process. This thesis outlines steps to adapt Stokes polarimetry to perform measurements on EUV, allowing the characterization of novel generation schemes.
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LIST OF SYMBOLS

Electric Field ................................................................. \( \mathbf{E} \)
Frequency in rad/sec ........................................................ \( \omega \)
Index of refraction ............................................................. \( n \)
Magnetic Field ................................................................. \( \mathbf{B} \)
Mueller matrix for arbitrary waveplate ................................ \( M_{WP} \)
Mueller matrix for half waveplate ......................................... \( M_{HWP} \)
Mueller matrix for polarizer ................................................ \( M_{POL} \)
Mueller matrix for quarter waveplate ................................... \( M_{QWP} \)
Mueller matrix for rotator .................................................. \( M_{ROT} \)
Polarization field ............................................................... \( \mathbf{P} \)
Reflectance for P-type polarization ....................................... \( r_P \)
Reflectance for S-type polarization ....................................... \( r_S \)
Relative phase shift from waveplate .................................... \( \phi \)
Speed of light ................................................................... \( c \)
Stokes vector .................................................................... \( \mathbf{S} \)
Transmittance of polarizer along \( x \)-axis ............................... \( p_x \)
Transmittance of polarizer along \( y \)-axis ............................... \( p_y \)
Wave vector .................................................................... \( \mathbf{k} \)
LIST OF ABBREVIATIONS

Boron-Carbide ......................................................... B4C
Colorado School of Mines ............................................. CSM
Degrees of Freedom .................................................... DOF
Extreme Ultraviolet .................................................... EUV or XUV
Hermite-Gaussian ...................................................... HG
High Harmonic Generation .......................................... HHG
Infrared ................................................................. IR
Laguerre-Gaussian ...................................................... LG
Stereolithography ....................................................... SLA
Ultra High Vacuum ..................................................... UHV
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For those that shall follow after.
CHAPTER 1
INTRODUCTION

High Harmonic generation (HHG) is the leading way to generate table-top coherent extreme ultraviolet (EUV) and attosecond pulses. HHG is a particular interest in industrial and scientific applications, including lithography for microchip manufacturing, measuring magnetic circular dichroism [1] and attosecond pulse generation for measuring ultrafast phenomena. Using a single pump beam will only produce linearly polarized harmonics using a linearly polarized fundamental. Due to the process of HHG in gas, retrieving circularly polarized harmonics out of the gas jet from a single pump beam is extremely difficult. The EUV light from HHG is typically only generated with linear polarizations with a Gaussian-profile pump; however, different spatial and polarization profiles will result in changes in the harmonic-generated light. To measure the polarization profile of the harmonic generated light, we need to be able to use polarization optics in the extreme ultraviolet wavelength range. While transmissive crystal-based polarizers and waveplates work well in the visible range, none of these materials work in the EUV range. This thesis shows the usage of mirror systems that operate in the EUV range as a polarizer and waveplate within a EUV wavelength range. The Fresnel equations show that it is possible to utilize reflection phase shift differences between S- and P- polarizations to control the polarization. To accumulate phase shifts sufficient to achieve quarter-wave retardation, we use 3 reflections in a “K-mirrors” arrangement that transmits the beam inline with the incident direction.

There is already work in this area for EUV polarization control using reflective optics. Smith et al [2] showed that a bi-layer 4-reflector setup using Molybdenum and Boron Carbide performs well around 18-27 nm, significantly shorter than the wavelength we intend to use. The benefits of our generation scheme and wave retarder setup are the high flux of EUV light and the monochromatic 9th harmonic from our fundamental 400nm pulse.

Outside of reflective wave retarders, there are several other existing options for producing polarization-controlled HHG sources. These include clever harmonic generation schemes to generate specially polarized harmonics and thin film multilayer materials to alter polarization states. Circularly polarized harmonics are of particular interest due to the difficulty in producing them and to their utility in measuring magnetic and chiral properties. Only an ellipticity of 0.4 was observed experimentally, Using a typical single-source scheme and N\textsubscript{2} as the HHG medium [3].

Another method to control the polarization state in EUV is through transmissive multilayer films to produce a wave retarder for EUV, as demonstrated by Yamamoto et al and Schmidt et al [4, 5]. While
they perform well in converting to partially circularly polarized light, up to 75%, the the transmission through the wave plate is around 5%. The low transmission gives reflective optics a leg up over the specialty waveplates as they can achieve higher transmission.

Various generation schemes have also been shown to generate highly elliptical polarization using crossed beam mixing (Haung et al [6] & Hickstein et al [7]). The crossed beams both use opposite circular polarizations and are crossed at the position of the gas to create a region of linear polarization constructively. This method allows the mixed beams to generate angularly separated harmonics with RCP and LCP due to the photon selection process in HHG.

The structure of this thesis aims to walk through the process of building up the understanding of Stokes polarimetry and the added complexity with each additional consideration. Chapter 2 aims to build an understanding of the basics of Stokes polarimetry and high harmonics, then Chapter 3 describes the process of performing this as a full-beam measurement on the 400nm fundamental driving beam. Chapter 4 walks through the design of an EUV wave retarder and polarizer for use in the full-beam EUV polarimetry in Chapter 5.
CHAPTER 2
BACKGROUND INFORMATION

In this chapter, I will provide background information for understanding the working phenomena and underlying principles used in this project. The topics covered will be the polarization of light, their formulations, polarization optics, classical Stokes measurements, and the properties of polarized light reflected at an interface.

2.1 Polarization of Light

When thinking about waves, typically sound, water, or other pressure waves, polarization is not considered because these are longitudinal waves. Sound waves create a wave of high and low-pressure regions in the same direction of the propagation of the wave, and thus the energy travels in the same direction as the disturbance. On the other hand, light, an electromagnetic wave, does not have a longitudinal component of electric or magnetic field along the direction of travel. Instead, light travels in a direction orthogonal to the “disturbance” (a changing magnetic or electric field), making light a transverse wave. Following the Maxwell equations with the electromagnetic wave equation, we can show that light is a transverse wave.

Following the substitution of Faraday’s law into the Maxwell-Ampere, we can get the result of the electromagnetic wave equations for electric and magnetic fields:

\[
\nabla^2 E = \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} \tag{2.1}
\]

\[
\nabla^2 B = \frac{1}{c^2} \frac{\partial^2 B}{\partial t^2} \tag{2.2}
\]

where \(E\) and \(B\) are the electric and magnetic field vectors, \(c\) is the speed of light in vacuum and \(t\) is time. The most basic solution to these equations is the plane wave:

\[
E(t) = E_0 e^{i(k \cdot x - \omega t)} \tag{2.3}
\]

\[
B(t) = B_0 e^{i(k \cdot x - \omega t)} \tag{2.4}
\]

where \(k\) is the wave vector and \(\omega\) is the frequency. We can see light as a transverse wave by substituting these solutions into the Faraday equation.
\[ \nabla \times \mathbf{E} = \frac{\partial \mathbf{B}}{\partial t} \quad \text{(Faraday Equation in Vacuum)} \quad (2.5) \]

Adding in the plane wave solutions,

\[ \nabla \times (\mathbf{E}_0 e^{i(k \cdot x - \omega t)}) = \frac{\partial}{\partial t} (\mathbf{B}_0 e^{i(k \cdot x - \omega t)}) \]

Substituting back for \( \mathbf{E}(t) \) and \( \mathbf{B}(t) \),

\[ i \mathbf{k} \times \mathbf{E} = i \omega \mathbf{B} \]

which simplifies to

\[ \mathbf{B} = \frac{\mathbf{k}}{\omega} \times \mathbf{E} \quad (2.6) \]

The cross-product in Equation 2.6 dictates that the direction of propagation (\( \mathbf{k} \)) is perpendicular to both the electric and magnetic field components of the wave, making light a transverse wave. Because most formulations of electromagnetic waves use electric properties, the polarization of light is defined as the direction of the electric field component.

![Figure 2.1 Electromagnetic plane wave showing the transverse nature of the electric and magnetic fields and direction of propagation, \( \mathbf{v} \).](from Wikimedia Commons by SuperManu)

As will be shown in this thesis, light as a transverse wave gives rise to complex properties that can be used to control light, measure material properties, and measure the light itself.

### 2.1.1 Polarization in the Jones and Stokes Formulations

Polarization in an EM wave can be analyzed and understood through Jones vectors, which contain magnitude and phase information about the electric field components in the \( x \) and \( y \) plane. Magnitude differences and phase offsets between \( x \) and \( y \) can significantly change the polarization state to create elliptical and circular polarization. For example, taking equal intensities of \( x \) and \( y \) polarizations in phase, we can constructively create +45° polarized light. Taking similar intensities, 180° out of phase, we can
similarly make -45° polarized light. Taking it a step further and adding a quarter-wave phase shift (90°)
between the x and y polarizations can create circularly-polarized light where the direction of the electric
field will rotate for one point in space. Since there are several different possible orientation states, we will
refer to each as Linear 0° (x), Linear 90° (y), Linear +45°, Linear -45°, Counterclockwise circular (Right
Circular), and Clockwise circular (Left Circular) polarization states. The Jones vectors can represent these
various states as seen in Table 2.1 [8].

Table 2.1 Various polarization states represented in the Jones Vector and Stokes Vector Forms

<table>
<thead>
<tr>
<th>Form</th>
<th>Linear 0°</th>
<th>Linear 90°</th>
<th>Linear +45°</th>
<th>Linear -45°</th>
<th>Circular CCW</th>
<th>Circular CW</th>
<th>Unpolarized</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jones</td>
<td>( E_0 \begin{pmatrix} 1 \ 0 \end{pmatrix} )</td>
<td>( E_0 \begin{pmatrix} 0 \ 1 \end{pmatrix} )</td>
<td>( \frac{E_0}{\sqrt{2}} \begin{pmatrix} 1 \ 1 \end{pmatrix} )</td>
<td>( \frac{E_0}{\sqrt{2}} \begin{pmatrix} 1 \ -1 \end{pmatrix} )</td>
<td>( \frac{E_0}{\sqrt{2}} \begin{pmatrix} 1 \ i \end{pmatrix} )</td>
<td>( \frac{E_0}{\sqrt{2}} \begin{pmatrix} 1 \ -i \end{pmatrix} )</td>
<td>???</td>
</tr>
<tr>
<td>Stokes</td>
<td>( I_0 \begin{pmatrix} 1 \ 0 \end{pmatrix} )</td>
<td>( I_0 \begin{pmatrix} -1 \ 0 \end{pmatrix} )</td>
<td>( I_0 \begin{pmatrix} 0 \ 1 \end{pmatrix} )</td>
<td>( I_0 \begin{pmatrix} 1 \ 0 \end{pmatrix} )</td>
<td>( I_0 \begin{pmatrix} 1 \ 0 \end{pmatrix} )</td>
<td>( I_0 \begin{pmatrix} 0 \ 1 \end{pmatrix} )</td>
<td>( I_0 \begin{pmatrix} 0 \ 0 \end{pmatrix} )</td>
</tr>
</tbody>
</table>

An alternate form of representing polarization information is with the Stokes vectors, also shown in
Table 2.1. The Stokes vectors work with intensity instead of electric field, making them easier for intensity
measurements. Intensity measurements alone have no information about the relative phase between x and
y polarizations; because of this missing information, there are six degenerate polarization states to
consider. For example, an intensity measurement can get equal intensities for x and y polarization but will
not decipher whether the light is ± 45° or R/L circularly polarized. To get from Jones to Stokes, one uses
the relation:

\[
S = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} = \begin{pmatrix} E_{0x}^2 + E_{0y}^2 \\ E_{0x}^2 - E_{0y}^2 \\ 2E_{0x}E_{0y}\cos\delta \\ 2E_{0x}E_{0y}\sin\delta \end{pmatrix}
\]

(2.7)

Where \( \delta = \delta_y - \delta_x \) represents the phase shift between axes and the absolute value of each \( S \) gives the
intensity of the light in each polarization state, with:

- \( S_0 \) representing overall intensity
- \( S_1 \) representing linear 0° (+1) and 90° (-1) polarization
- \( S_2 \) representing linear +45° (+1) and -45° (-1) polarization
- \( S_3 \) representing right circular (+1) and left circular (-1) polarization
A secondary benefit of the Stokes formulation is the ability to quantify the amount of polarized light. For a fully polarized beam, the intensity in all the orthogonal channels should be the same as the overall intensity, following the relation:

\[
S_0^2 \geq S_1^2 + S_2^2 + S_3^2
\]  

(2.8)

However, when \( S_0 \) is greater than the sum of the polarization channels added in quadrature, there is a portion of unpolarized light. The DOP (Degree of Polarization) is defined by the following:

\[
P = \frac{I_{pol}}{I_{tot}} = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0}, 0 \leq P \leq 1
\]  

(2.9)

### 2.1.2 Polarization Optics in Mueller Matrices

Polarization is a commonly used method to control light, from LCDs to ultrafast regenerative amplifiers. In visible and infrared (IR) parts of the spectrum, where there are several birefringent crystals with good transmission, standard polarization optics work in transmission with the light traveling through the material, which changes the polarization of the light passing through.

Polarization optics can be modeled and applied to Stokes vectors using Mueller matrices. For a guide on Mueller matrices and general Polarization formulations, optics and control, see the SPIE Field Guide to Polarization [8]. Mueller matrices can model a single polarizing element or a sequence of elements. The general form of Mueller matrices is the following:

\[
\begin{pmatrix}
    S'_0 \\
    S'_1 \\
    S'_2 \\
    S'_3
\end{pmatrix} =
\begin{pmatrix}
    m_{00} & m_{01} & m_{02} & m_{03} \\
    m_{10} & m_{11} & m_{12} & m_{13} \\
    m_{20} & m_{21} & m_{22} & m_{23} \\
    m_{30} & m_{31} & m_{32} & m_{33}
\end{pmatrix}
\begin{pmatrix}
    S_0 \\
    S_1 \\
    S_2 \\
    S_3
\end{pmatrix}
\]  

(2.10)

Polarizers are designed to pass a well-polarized linear polarization while attenuating or separating the orthogonal polarization. Polarizers can discriminate polarizations with reflection or absorption of the unwanted polarization state with different designs, including [9]:

- **Absorptive**: This is one of the most common types of polarizers, seen in many consumer-based products. Most absorptive polarizers use a special doped plastic sheet stretched to align the polymer chains along one axis. These sheet polarizers are found in LCDs and polarized sunglasses.

- **Birefringence-based**: These polarizers use clear birefringent crystals to create a reflection near the Brewster angle for one polarization of light over another. These are often made into polarizing beam splitters,
• Thin-film: (TFPs) are often used in transmission, with specialty coatings laid many times on one face of the glass to reflect nearly all of the S-type polarization and transmit the P-type polarization by having many layers of index change to reflect the light effectively.

• Wire-grid polarizers: These polarizers use machined (sub-wavelength) metal lines to attenuate one direction of polarization by allowing the electric field along the wires to drive a current in the wires, but not in the perpendicular direction to the wires, a general layout is shown in Figure 2.2.

![Figure 2.2 Diagram showing simple wire-grid polarizer passing a single linear polarization (from Wikimedia Commons)](image)

Using the Mueller method to find the direct effect of light passing through a polarizer, we can find the form for an ideal polarizer aligned to the \(x\)-axis (0°). This matrix is the following:

\[
M_{\text{POL}} = \frac{1}{2} \begin{pmatrix}
1 & 1 & 0 & 0 \\
1 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix}
\] (2.11)

Applying this horizontal polarizer matrix to a Stokes vector state will only leave power in the positive \(S_1\) and the \(S_0\) channels, meaning that the only remaining state of light is linear 0° polarization. Similarly, an ideal polarizer aligned to the \(y\)-axis (90°), would be the following:

\[
M_{\text{POL}} = \frac{1}{2} \begin{pmatrix}
1 & -1 & 0 & 0 \\
-1 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix}
\] (2.12)

Applying this vertical polarizer matrix to a Stokes vector will only leave power in the negative \(S_1\) and the \(S_0\) channels (overall power and vertical linear). For a non-ideal polarizer or optic with varying transmission between \(x\) and \(y\) axes, the form of the Mueller matrix is the following:
\[ M_{\text{POL}}(p_x, p_y) = \frac{1}{2} \begin{pmatrix} p_x^2 + p_y^2 & p_x^2 - p_y^2 & 0 & 0 \\
 p_x^2 - p_y^2 & p_x^2 + p_y^2 & 0 & 0 \\
 0 & 0 & 2p_xp_y & 0 \\
 0 & 0 & 0 & 2p_xp_y \end{pmatrix} \]  \hspace{1cm} (2.13)

Where \( p_x \) and \( p_y \) are the electric field transmission parameters through the optic. In order to find the state through an arbitraryity rotated polarizer, one can use a coordinate shift. The rotator Mueller matrix can be used to perform a coordinate shift to look at rotated optics by front multiplying by \( M_{\text{ROT}}(-\theta) \) and back multiplying by \( M_{\text{ROT}}(\theta) \).

\[ M_{\text{ROT}}(\theta) = \begin{pmatrix} 1 & 0 & 0 & 0 \\
 0 & \cos 2\theta & \sin 2\theta & 0 \\
 0 & -\sin 2\theta & \cos 2\theta & 0 \\
 0 & 0 & 0 & 1 \end{pmatrix} \]  \hspace{1cm} (2.14)

Where \( \theta \) is the angle of rotation for the matrix. Since the Stokes parameters \( S_1 \) and \( S_2 \) are for states 45° from each other, the angle is \( 2\theta \) instead of a single \( \theta \) such as the case with the Jones vector form with 90° between the two parameters.

While polarizers are a discriminator for a specific linear polarization, waveplates, another extremely common polarization optic, are designed to change the polarization state of the transmitted light through the optic. Wave plates in visible and IR light are almost exclusively made from birefringent materials. Birefringence (also called double refraction), in the case of uniaxial crystals, arises from a crystal asymmetry that produces a difference in refraction index along one crystal axis, referred to as the extraordinary axis. The phase shift along the extraordinary axis can be found with the following:

\[ \Delta \phi = k_0(n_e - n_o)L, \]  \hspace{1cm} (2.15)

where \( k_0 \) is the wave number, \( n_o \) is the ordinary index of refraction, where as \( n_e \) is the extraordinary index of refraction and \( L \) is the thickness of the material. Changing the projection of the crystal extraordinary axis with the wavefront of the light will change the effective \( n_e \) in that polarization orientation, meaning that to design for a specific phase shift, both crystal orientation and material thickness need to be taken into account. Most waveplates are explicitly designed to provide a quarter- or half-wave phase shift. Half waveplates (HWP) have a relative phase shift of \( \pi \) and can be used to convert between linear polarization states by that half-cycle shift, meaning that with the correct orientation of the fast and slow axes, you can exchange any arbitrary linear polarization to another. A rotated half-waveplate can be represented with the following Mueller matrix:
where $\theta$ is again the angle of rotation for the waveplate. The Mueller matrix for a half waveplate is incredibly similar to the matrix for the rotator, often called a Faraday rotator, which rotates the polarization of the light. The half waveplate only differs by an extra factor of two on $\theta$, as well as a sign flip on the circular-to-circular term, meaning that it will swap the handedness of the input polarization on the output. The other commonly used waveplate, quarter waveplates (QWP), give a phase shift of $\frac{\pi}{2}$ and can convert linear to circular polarization states and vice-versa. The Mueller matrix for the QWP is the following:

$$M_{\text{QWP}}(\theta) = \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & \cos 2\theta & \sin 2\theta & 0 \\
0 & \sin 2\theta & \cos 2\theta & 0 \\
0 & -\sin 2\theta & \cos 2\theta & 0
\end{pmatrix}$$ (2.17)

For an arbitrary phase-shift waveplate, the Mueller matrix is the following:

$$M_{\text{WP}}(\phi, \theta) = \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & \cos 2\theta + \cos \phi \sin^2 2\theta & (1 - \cos \phi) \sin 2\theta \cos 2\theta & \sin \phi \sin 2\theta \\
0 & (1 - \cos \phi) \sin 2\theta \cos 2\theta & \sin^2 2\theta + \cos \phi \cos^2 2\theta & -\sin \phi \cos 2\theta \\
0 & -\sin \phi \sin 2\theta & \sin \phi \cos 2\theta & \cos \phi
\end{pmatrix}$$ (2.18)

where $\phi$ is the relative phase shift between ordinary and extraordinary axes, while $\theta$ is the angle of orientation.

In the case of birefringent waveplates, the design of each waveplate is highly wavelength-dependent since the specific phase shift is dependent on the wavelength. Furthermore, broadband performance is further hindered by indices within the material change significantly with wavelength, which means that a quarter waveplate at 800nm will not work as a half waveplate at 400nm.

### 2.1.3 Classical Stokes Measurements

There is reason to think that you could measure two independent states with a polarizer in two positions. This could be done with a measurement of the vertically polarized light and the horizontally polarized light separately. However, with $\pm45^\circ$ and circularly-polarized light, the intensity from both the vertical and horizontal channels would be identical, despite the fact that these are very different polarization states. The only way to be able to find the circular and $\pm45^\circ$ polarization from two measurements would be to measure the relative phase of each. Measuring the phase difference between
these beams of light is tricky (especially if you are trying to find it for a whole beam profile) [10]. To get around measuring the phase, more intensity measurements can be made to find the $+45^\circ$ and $-45^\circ$ polarized light and the right and left circularly polarized light.

To find the $S_0$ and $S_1$ measurements, you can add the intensities through a polarizer at $0^\circ$ and $90^\circ$. This addition will give the overall intensity of the light, $S_0$. For the $S_1$ measurement, take the difference between the $0^\circ$ and $90^\circ$ measurements ($I_{0^\circ} - I_{90^\circ}$). This subtraction will give the measurement distinguishing $0^\circ$ and $90^\circ$ polarized light.

Finding the $S_2$ measurement can be as simple as taking an intensity measurement with a polarizer at $45^\circ$ and using the intensities from the previous two measurements. To get the $S_2$ measurement, which measures light independently polarized in the $+45^\circ$ and $-45^\circ$ directions, you need the difference between the $45^\circ$ measurement and the overall intensity. To differentiate the independent $45^\circ$ light from the 50% transmission of $0^\circ$ or $90^\circ$ light through the polarizer in this orientation, a factor of two is added, and the overall intensity can be subtracted.

To find the $S_3$ measurement, a quarter waveplate needs to be used in conjunction with the polarizer to make a circularly-polarized "gate." The quarter wave plate comes first, which can orient the fast axis (axis without quarter wave retardation) to vertically polarized, making the slow axis oriented horizontally. The QWP will then convert the circularly-polarized component of the light to $\pm 45^\circ$. Then, with the polarizer oriented at $45^\circ$, this will act to either transmit or reduce the light through the optics depending on the handedness of the polarization. All linear polarization states will get transmitted at 50% power through this system, which means any difference above or below the 50% threshold is Right or Left circularly polarized light, respectively.

\[
S_0 = I_{0^\circ} + I_{90^\circ} \\
S_1 = I_{0^\circ} - I_{90^\circ} \\
S_2 = 2I_{45^\circ} - S_0 \\
S_3 = 2I_{\text{QWP}} - S_0
\]  

(2.19)

### 2.1.4 Polarized light at an interface

As mentioned earlier, birefringent materials are used for waveplates to control the polarization state of light. In EUV, materials are not very transmissive, so reflection-based phase shifts is a promising approach.

When light hits an interface between materials (change in the index of refraction), the light is reflected or refracted and transmitted. The angle of reflection is equal to the incident angle on the material, while the transmitted light is refracted by Snell’s Law, which dictates the refraction of light where there is a
change in the index of refraction, \( n \), at an interface.

\[
n_I \sin \theta_I = n_T \sin \theta_T
\]  

(2.20)

Where \( n_I \) is the index of the incident material and \( n_T \) is the index of the material passed into, and each \( \theta \) is the incident angle on the interface.

While Snell’s law dictates the refraction angle of the light into the material at the interface, the power transmission and reflection are determined by the Fresnel equations. The Fresnel equations are very dependent on polarization and have different quantities for whether the polarization is parallel (S-type) to the plane of the index change or perpendicular to the index change (P-type). Each polarization state has different coefficients for the transmission and reflection of the light, which gives us the four Fresnel equations:

\[
t_S = \frac{2n_I \cos \theta_I}{n_T \cos \theta_I + n_I \cos \theta_T}
\]

(2.21)

\[
t_P = \frac{2n_I \cos \theta_I}{n_I \cos \theta_I + n_T \cos \theta_T}
\]

(2.22)

\[
r_S = \frac{n_T \cos \theta_I - n_I \cos \theta_T}{n_I \cos \theta_I + n_T \cos \theta_T}
\]

(2.23)

\[
r_P = \frac{n_T \cos \theta_I - n_I \cos \theta_T}{n_T \cos \theta_I - n_I \cos \theta_T}
\]

(2.24)

where each \( r \) is the electric field reflection coefficient, and \( t \) is the field transmission coefficient.

An optic that achieves a phase-shift from a reflection is a Fresnel Rhomb, this optic uses a cut crystal to cause total internal reflection (TIR) inside the crystal to cause a phase shift between the parallel and perpendicular field components. The phase shift with vary based on the incident angle on the TIR interface [11].

Instead of TIR, which requires a transmissive material, we can use materials with a complex refractive index. For waves reflected from an interface with a complex index of refraction, there is a phase shift from the reflection. For polarization perpendicular to the surface, that phase shift is 180°, while parallel to the surface, the phase shift is 0 degrees. For every reflection at an angle, this phase shift can vary between these two limits, meaning that at specific angles, you can get a phase shift along the P-type polarization in the reflection. The ability to "tune" this phase shift with the angle of reflection is the basis for the quarter wave retarder designed in Section 4.1. To get this phase shift out, use the complex refractive index in Equation 2.24 and 2.23. With \( r_S \) and \( r_P \), find the phase of each complex number and find the difference between the two, which gives the relative phase shift between S & P polarizations. With complex indices of refraction, the transmitted angle, \( \theta_T \) is not clear, so this is solved by substituting Snell’s Law, which is
discussed in more detail in Section 4.1.

2.2 High Harmonic Generation

The generation of coherent EUV beams is a complex problem to solve. There are two issues:

- Crystals are not transparent in the EUV range, so typical lasing sources and nonlinear crystals cannot be used
- While it is possible to pump a plasma to get a soft X-ray source, such systems are very complex.

Synchotrons are a possibility to generate EUV light, however are inaccessible for most; so therefore, High Harmonic Generation is the typical method of EUV generation.

2.2.1 Harmonic Generation using non-linear media

The most common method of changing the wavelength of a laser source is second harmonic generation (SHG). This method uses a birefringent crystal with a second-order nonlinearity to phase-match and generate second harmonics. The phenomena of SHG are modeled as part of a second-order process.

When an electric field is applied to a material, it induces a dipole in the material, which often acts linearly to the magnitude of electric field

\[ D = \epsilon E \] \hspace{1cm} (2.25)
\[ P = \chi E \] \hspace{1cm} (2.26)

where \( D \) is the displacement field, \( P \) is the induced dipole in the material, also called the polarization field, and \( \epsilon \) is the permittivity of the material.

However, when reaching high fields, the linear relation breaks down, and we break \( \chi \) into a linear expansion because the driving field is able to reach asymmetries in the material.

\[ P = \chi^{(1)} E + \chi^{(2)} E^2 + \ldots \] \hspace{1cm} (2.27)

where \( \chi \) expands to \( (\chi^{(1)} + \chi^{(2)} E + \ldots) \) and \( \chi^{(2)} \) is the second order non-linearity of the material. If you are able to send high-intensity light into a material with a high second-order susceptibility and phase-match the index of refraction of the fundamental and the second harmonic \((2\omega \text{ or } \lambda/2)\) then you can up-convert the fundamental light efficiently into the second harmonic. While SHG uses a material with an asymmetry (an even order of \( \chi^{(n)} \)) symmetric materials can also generate harmonics; such is the case with the third harmonic generation, a third-order process \((\chi^{(3)})\). Furthermore, in the case of SHG, the material’s asymmetry is the cause of the second harmonic signal, and higher even orders can only be
generated from higher even-order nonlinearities. Similarly, odd-order nonlinearities will generate odd harmonics of the fundamental, which means anisotropic materials will only generate odd-order harmonics. In the case of high harmonics generated in a gas, these nonlinearities are much more complex and dominated by ionizing electrons from the atoms.

### 2.2.2 Phenomena of High Harmonics

Commonly, the phenomena of high harmonic generation are described with a semi-classical model. The three-step model, proposed by Corkum, [12] describes the extremely high electric field being capable of deforming an electron orbital to the point where the electron tunnels out of the orbital in a one-half optical cycle and returns to the atom within the next half-cycle. When the electron returns to the bound state, it releases high-energy photons, where the phase is related to the phase of the fundamental driving light, creating a coherent EUV source. This three-step model is shown in Figure 2.3. EUV are created coherently in the HHG process in short bursts at the frequency of the optical cycle of the driving beam.

For HHG in gas, the odd orders dominate the signal due to the symmetry of the gas prioritizing generation at the odd orders and suppressing the even order-generated light. The coherent light emitted from the process, along with multiple orders of harmonics, interferes in time to create a coherent pulse of EUV light within attosecond time scales.

![Diagram of the three-step model for HHG](image)

Figure 2.3 Diagram of the three-step model for HHG. First, the strong field allows the electrons to tunnel out of the atomic potential well, then accelerates away and back to the atom, recombining and releasing an XUV photon. Diagram from Wikimedia Commons.

Generating circularly polarized harmonics is difficult because of the driving beam’s low recombination efficiency and higher ellipticities. As with circular polarization, a rotating, constant-magnitude electric field will continue to drive the electron in different directions, deterring the ionized electrons in the gas from recombining efficiently.
2.3 Generation Apparatus

To get the best results and a narrow EUV bandwidth, we went with a generation scheme using a 400nm beam focusing into our gas jet to get power centered around the 9th harmonic (44.4 nm), with less power in the 11th and 7th harmonics.

The laser source for the generation is done with an 800nm Ti-Saphire laser, the Solstice Ace, at around 250 mW and 40-50 fs pulses. Along the path from the source, the beam is sent to a deformable mirror (DM) to correct for aberrations and flatten the beam’s wavefront. For our specific generation scheme, the 800nm pulse is frequency doubled to 400nm through a doubling crystal, KTP. The 400nm beam is focused into the HHG chamber with a lens into a gas jet of Argon. After the gas jet, we remove the 400nm from the beam line with an aluminum filter before incidence on the EUV optics. A sample diagram of our system is shown in Figure 2.4.

![Diagram of HHG chamber and EUV beamline and (b) upstream optics to create and optimize 400nm fundamental](image)

The specific parameters for our system dictate that we generate up to the 11th harmonic. With the aluminum filters, anything longer in wavelength than the 7th harmonic is heavily attenuated. The EUV beam we get through the system is primarily composed of the 9th harmonic of 400nm, at 44.4nm (28eV), with small amounts of 7th and 11th harmonics, both around 10% of total beam power.

With the upstream polarization optics, we intend to create a varying polarization profile across the beam, leading to structured light in both intensity and polarization at the focus inside the gas jet, leading
to structured harmonics.

To contribute to the system and improve the system and characterization capabilities of our HHG chamber system, I designed a camera mount to hold the camera and the vacuum bellows, seen in Figure 2.5. This is designed to still allow for minimal tip/tilt and movement control on the camera while being significantly more stable than the previous mount that would flex under vacuum. I also designed a mirror mount system to be able to reflect the HHG spectra from our grating back in line with the camera, seen in Figure 2.6. This allowed our system the capability to spectrally resolve images, which is critical with work using Hermite-Gaussian (HG) HHG.

Figure 2.5 Camera mount for EUV-sensitive camera. This design uses turnbuckles mounted to machined aluminum brackets that attach to the vacuum flanges to hold the camera and provide stability and isolation for the camera.
Figure 2.6 Mirror mount system for HHG spectrometer. Rotated 2x2in gold mirror (center) is held by a 3D printed arm attached to a picomotor mirror mount (left) to reflect HHG spectra from grating (right) to camera. Fits the 2x2in mirror inside of the vacuum flange.
Polarimetry often assumes the beam has a uniform polarization state. In this case, the polarimetry measurements can be done with a single power meter. This chapter aims to give an understanding and background of finding full-beam Stokes measurements before diving into the design and usage of the EUV polarization optics. This chapter follows an example of full beam Stokes polarimetry measurement of a vector beam produced by an S-waveplate, which produces a polarization state whose polarization varies directly with azimuthal angle within the beam. Since the polarization state is a function of position, we aim to do a spatially-resolved characterization of the polarization state. A secondary benefit of this characterization is a look at the performance of the polarizing optics.

This thesis’s main area of interest was to look at the HHG process with structured beam profiles, which includes not only intensity variation across the beam but also polarization and phase variations across the beam. Specifically, we wanted to look at high harmonics generated with vector beams. Vector beams have a first-order Laugurre-Gauss (LG\textsubscript{10}) laser mode with an azimuthal or radial (or mixture) polarization profile around the zero point in the LG mode’s center.

![Figure 3.1 Example of a radially polarized vector beam, with lines showing the direction of polarization across the profile](image)

**3.1 Setup for 400nm Stokes Measurements**

The setup for the classical Stokes measurements follows the description from Section 2.1.3. The measurement was performed with a 400nm diode source to not interfere with the HHG chamber when other experiments were underway.
The light source was a 405nm fiber-coupled laser diode that produced about 10mW of power. Since the output polarization was not pure, the output was sent through a Glan–Thompson polarizer. This amount of unpolarized light was measured by performing a Stokes measurement on the output of the fiber.

After the first polarizer, we placed the polarizing optics to measure the polarization profile. This optic was the 400nm first-order S-waveplate, which is a specially machined zero-order half-waveplate. The S-waveplate is designed to rotate the input polarization through 360° of rotation around a center point. This polarization profile creates a vector beam, such as the one in Figure 3.1. The zero point in the center of the beam results from divergent light of opposite phases around the center destructively interfering. Depending on the input polarization into the S-waveplate, the polarization output can vary between azimuthally and radially polarized around the center zero point. The setup can be seen in Figure 3.2.

To perform the four required measurements to retrieve the Stokes parameters, a location for a quarter waveplate and a polarizer followed. A camera was placed after the polarizer to recover the full beam image. We found that if not aligned correctly, the beam would wander on the camera when rotating the polarizer, so we made fine alignment by hand of the polarizer in front of the camera.

3.2 Results

Using the polarizer in the three required positions and the fourth measurement with the quarter waveplate, the camera saw the expected Hermite-Gaussian (HG_{01} and HG_{10}) modes (albeit rotated around the center) for each of the polarizer measurements, which can be seen in Figure 3.3. We also found a full LG_{10} mode for the measurement with the quarter waveplate, indicating that we will likely get little to no power in the circular polarization mode, which we should expect.
Unfortunately, the polarizer and the neutral density filters on the camera added unwanted structure to the beam, which can be easily reduced by convolving a Gaussian filter with the image data. Using each pixel in each MxN image as a separate intensity measurement, we followed the relations in Equation 2.19, retrieving an MxN×4 3-D array where each MxN “layer” functions as its own Stokes parameter, represented pictorially in Figure 3.4.
Figure 3.4 Images showing full-beam Stokes parameters for 400nm vector beam. Calculated using the full-beam intensity measurements in Figure 3.3. The images were run through a Gaussian filter with $\sigma = 20$.

We can show the polarization ellipses directly on top of the beam profile to better visualize the polarization profile, as seen in Figure 3.5. The beam polarization profile shows a mixture of radially and azimuthally polarized vector beams, with the polarization spiraling outward. As expected, there is a low amount of eccentricity in the polarization; however, this is still non-zero when we would expect no ellipticity in the beam.

Figure 3.5 Representation of the polarization profile of the measured vector beam with polarization ellipses drawn overttop the intensity profile ($S_0$). Polarization ellipses are plotted on a lower-sampled grid for ease of visualization. The color of the ellipses represents the handedness of the circular portion of the polarization; the design for this plot is derived from [10].

The degree of polarization is a good metric to judge measurement confidence to verify the polarization we get from the Stokes measurements. Figure 3.6 shows the DOP profile using the Stokes parameters side-by-side with the logarithmic intensity profile of the beam. This comparison indicates that the DOP
follows the areas of the image with intensity, which is expected as stray background light outside the beam will be unpolarized.

Figure 3.6 Plots of (a) intensity and (b) DOP profiles for vector beam fundamental. Demonstrates the measured DOP will follow the same profile as beam intensity for what we expect to be a well-polarized beam.
CHAPTER 4
DESIGN OF A REFLECTION-BASED WAVE RETARDER

While multi-reflection polarization optics have been designed and used before with high harmonics, with success[2] and [13], our work with this differs with three major aspects:

1. Wavelength. We are using a generating scheme with a 400nm fundamental and generating with Argon which allows us to have a reasonably monochromatic EUV beam at 44.4nm, simplifying the wavelength range for optimal circular conversion.

2. A single layer of material. Our design uses a single layer of B4C, which still achieves high reflectance and phase shift with ease of production and robustness of the mirror.

3. A three-reflector setup. The design of the multi-reflector we used will still reflect the beam back into the optical axis with fewer reflections, thus being able to cut down intensity losses through the reflector.

Working between EUV and visible regimes of light, we work with separate conventions for angles in reflection/transmission. Reflective optics in X-ray/EUV often use the grazing angle (between the beam and the mirror’s surface), while visible optics uses the incident angle (between normal to the mirror and the beam). I will often use the incident angle for the most straightforward translation to the Fresnel equations, even within the EUV wavelength regime.

![Figure 4.1 K-Mirror geometry](image)

Figure 4.1 K-Mirror geometry

Looking at the physical geometry to make this set of equations work, we get a relation between the incident angles on the mirrors:
\[ \theta_2 = 2\theta_1 - \frac{\pi}{2} \]  

(4.1)

Where \( \theta_1 \) is the incident angle on the first and last mirror, and \( \theta_2 \) is the incident angle on the center mirror.

For this analysis, we can look at the reflection power from the Fresnel equations because the transmitted light is not used through each interface. Since we will only be using the reflective coefficients, \( r_S \) and \( r_P \), we can substitute Snell’s Law (Equation 2.20) into Equation 2.23 and 2.24, to remove the angle of transmitted light, \( \theta_T \).

\[
r_P(\theta_I, \lambda) = \frac{n_{T,\lambda} \cos \theta_I - n_{I,\lambda} \sqrt{1 - \frac{n_{I,\lambda}^2 \sin^2 \theta_I}{n_{T,\lambda}^2}}}{n_{T,\lambda} \cos \theta_I + n_{I,\lambda} \sqrt{1 - \frac{n_{I,\lambda}^2 \sin^2 \theta_I}{n_{T,\lambda}^2}}} \]

(4.2)

\[
r_S(\theta_I, \lambda) = \frac{n_{I,\lambda} \cos \theta_I - n_{T,\lambda} \sqrt{1 - \frac{n_{I,\lambda}^2 \sin^2 \theta_I}{n_{T,\lambda}^2}}}{n_{I,\lambda} \cos \theta_I + n_{T,\lambda} \sqrt{1 - \frac{n_{I,\lambda}^2 \sin^2 \theta_I}{n_{T,\lambda}^2}}} \]

(4.3)

4.1 Wave retarder analysis

When designing this wave retarder for our 44.4nm, we want this optic to act as much like a quarter waveplate as possible. To achieve this, we need to account for two design parameters to make this possible:

1. Relative phase shift - this reflector should act as a quarter waveplate, so we need the relative phase shift to be as close to \( \frac{\pi}{2} \) as possible.

2. Contrast ratio - for good full-beam performance and high transmission, there needs to be a low contrast ratio between the S & P polarizations coming off the mirrors. A low contrast ratio will allow good performance as a quarter-waveplate for arbitrary input polarizations.

The original design for this optic was based on a proposed plan for a quarter-wave retarder using Silicon Carbide (SiC) as the material [14]. While SiC is a good material for hardness and ease of depositing on a substrate, we used Boron Carbide (B4C) as a coating material. B4C has been used in this capacity before for a wave retarder. However, B4C was proposed as a cap layer on a molybdenum coating to prevent oxidation and protect the Molybdenum surface. For our purposes, B4C alone gives enough reflectivity in our wavelength range and makes a better mirror coating material due to the hardness and the lack of reactivity. The B4C mirrors are robust and have been easy to clean between uses. B4C has better EUV reflectivity performance than SiC. Although the performance is worse than Molybdenum, B4C’s structural
properties allow for a robust reflector.

As discussed in Section 2.1.4, the light will receive a phase change from a specific angle of reflection, different for each S and P polarization. Since the K-mirror design has three reflections, the total phase change from all reflections through the K-mirror should be designed to total a quarter wave of difference between the S and P reflections. Each mirror needs to be at a high incident angle to get the total phase shift near a quarter wave. Fortunately, high incident angles are also where we can get the highest transmission of EUV through the system.

To get the phase shift from a reflection, we can take the phase of each complex $r_S$ and $r_P$:

$$\phi_S = \text{arctan2}(\text{Re}[r_S(\theta)], \text{Im}[r_S(\theta)])$$
$$\phi_P = \text{arctan2}(\text{Re}[r_P(\theta)], \text{Im}[r_P(\theta)])$$

where arctan2 is the four quadrant arc tangent function and each $\phi$ is the phase shift from the reflection.

Since each reflection has an individual phase shift, we need to look at the total through the system. Our K-mirror reflectors have two reflections at $\theta_1$ and one at $\theta_2$. For the total phase shift from the series of reflections, we can multiply for the total $r_S$ and $r_P$:

$$\phi_{S,TOT} = \text{arctan2}(\text{Re}[r_S(\theta_1)^2 r_S(\theta_2)], \text{Im}[r_S(\theta_1)^2 r_S(\theta_2)])$$
$$\phi_{P,TOT} = \text{arctan2}(\text{Re}[r_P(\theta_1)^2 r_P(\theta_2)], \text{Im}[r_P(\theta_1)^2 r_P(\theta_2)])$$

Much like how a waveplate retards one specific polarization direction, we need the difference to shift one direction relative to the other. The difference between $\phi_S$ and $\phi_P$ will give us the relative phase shift.

$$\phi_{TOT} = \phi_{P,TOT} - \phi_{S,TOT}$$

where $\phi_{TOT}$ is the total relative phase shift through the mirror system. $\phi_{TOT}$ is the parameter we want to pin at $\frac{\pi}{2}$ to be able to convert linear to circular and circular to linear.

For the reflector optics to act as much like a quarter waveplate as possible for the conversion of arbitrary polarizations, we want to minimize the contrast between S and P polarizations as much as possible, meaning we need to take a look at the power transmitted through the K-mirror for both S and P.

Looking at the Fresnel equations, we can get the power from each reflection by taking the absolute value squared of $r_P$ and $r_S$.

$$R_S(\theta, \lambda) = |r_S(\theta, \lambda)|^2$$
$$R_P(\theta, \lambda) = |r_P(\theta, \lambda)|^2$$
For the 3-reflection setup, similar to the phase, each reflection will compound upon the light passed from each prior reflection. We can get the following transmission coefficients for both the S and P polarizations:

\[
T_S = R_S(\theta_1)^2 R_S(\theta_2) = R_S(\theta_1)^2 R_S(2\theta_1 - \pi/2) \quad (4.11)
\]

\[
T_P = R_P(\theta_1)^2 R_P(\theta_2) = R_P(\theta_1)^2 R_P(2\theta_1 - \pi/2) \quad (4.12)
\]

where \( T \) represents the transmission through the K-mirror system in the polarization state.

To start to solve these constraints for the K-mirror, we need to look at the data for \( n \) (index of refraction and imaginary extinction coef) at 44.4nm, which is:

\[
n_{\text{B}_4\text{C}, \text{44.4nm}} = 0.55397 + 0.22374i \quad [15] \quad (4.13)
\]

Using this index of refraction, we can sweep the input angle to find the optimal geometry to act as a quarter wave retarder, which can be seen in Figure 4.2.

Looking at the positioning of the first mirror, solving for the \( \frac{\pi}{2} \) phase shift, we get that the 77.0416° incident angle gives the optimal phase shift and low contrast between S and P polarizations of 1.74:1. To get an idea of the broadband performance at that angle, we can look at the reflectance and phase shift around the 44.4nm wavelength and the 11th and 7th harmonics of 400nm as well.

Figure 4.2 Plot of transmittance and phase shift through B4C K-Mirror as function of input incident angle
4.2 Polarizer

We need a polarizer to tell the performance of the reflection-based $\lambda/4$ wave-retarder. So we can design a polarizer that uses gold, similar to proven designs, [16]. For the 3-reflectors, we need to find an incident angle on the first mirror that will give high contrast with attenuation of P and transmission of S-type polarization. The index of refraction used for Au at 44.4nm is the following:

$$n_{\text{Au}, 44.4\text{nm}} = 0.95226 + 0.53289i[17]$$

Unfortunately, sweeping the first incident angle for the contrast through the K-mirror, we get a trade-off between contrast ratio and signal throughput, as seen in Figure 4.4. To balance the two constraints, we can use a first incident angle at 20 deg, giving us a contrast ratio of 38.23:1 for the 3-reflector setup. While the Au K-mirror does provide a relative phase shift between S & P polarized light, the intensity of the P-polarized light is minimal compared to the S-polarization, so the phase shift is negligible.
Figure 4.4 Plots of the (a) transmittance of S & P polarizations and (b) contrast as a function of first incident angle. The angle chosen was a 70° incident (20° grazing), shown in gray on each plot. These plots show the trade-off between high contrast and overall power throughput for the polarizer design.

Looking at the performance of the Au polarizer around the design wavelength, in Figure 4.5 it’s evident that the polarizer performance is relatively broadband, with overall changes mostly in transmission and not contrast ratio.

Figure 4.5 (a) Transmission through the Gold K-mirror and (b) contrast ratio as a function of wavelength for a first incident angle of 70°.
4.3 Modeling K-mirrors with Mueller matrices

For both the design of the polarizer and the wave retarder, there is a function as a polarizer and a wave retarder, even with the Au polarizer. In order to model the performance of these K-mirrors for an arbitrary input polarization, both the contrast and the phase shift should be taken into account. While the harmonics are centered around 44.4nm, light still should be accounted for in the 7th and 11th harmonics, all values for $p_x, p_y$ and $\phi$ can be found in Table 4.1.

<table>
<thead>
<tr>
<th>K-mirror</th>
<th>B4C (wave retarder)</th>
<th>Au (polarizer)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>7th</td>
<td>9th</td>
</tr>
<tr>
<td>Wavelength</td>
<td>57.14 nm</td>
<td>44.44 nm</td>
</tr>
<tr>
<td>$p_x$ (S-polarized)</td>
<td>0.553113</td>
<td>0.710294</td>
</tr>
<tr>
<td>$p_y$ (P-polarized)</td>
<td>0.213648</td>
<td>0.538005</td>
</tr>
<tr>
<td>$\phi_{TOT}$ (radians)</td>
<td>-1.1415</td>
<td>-1.5708</td>
</tr>
</tbody>
</table>

To analyze the polarization coming off the K-mirror, we can use $p_x, p_y, \phi_{TOT}$ to make an equivalent Mueller matrix. To rotate the wave retarder and polarizer together, we can stack the Mueller matrix to produce an equivalent system with rotators, a waveplate, and a polarizer. The form of this follows:

$$M_{KM} = M_{ROT}(-\theta)M_{WP}(\phi, 0)M_{POL}(p_x, p_y)M_{ROT}(\theta)$$ (4.15)

where $M_{KM}$ is the equivalent matrix for the K-mirror. Combining each K-mirror Mueller matrix for each harmonic (7th, 9th, 11th), one could find the throughput intensity and polarization through our K-mirror systems for arbitrary input polarization.

4.4 Physical Design

Because of the physical size constraints of our system, these K-mirrors need to be as small as possible, vacuum-compatible, adjustable, rotatable, and removed from the beamline under vacuum. We also need enough degrees of freedom to adjust the mirror system to align it back to the optical axis after the reflections. These design parameters necessitate a small footprint tip/tilt mirror mounting solution. For this, we went with Newport MFM-100-LC mounts, which are industrial tip/tilt flexure mounts chosen for the compact design to fit within the dimensions of the rotation mount holding the whole assembly. The parts to hold the mirror mounts were custom 3D printed using a stereolithography (SLA) printer. In our experience, SLA printed parts have good vacuum compatibility and minimal out-gassing issues from the resin curing process.
Figure 4.6 Images of SOLIDWORKS assemblies of (a) B4C K-mirror and (b) Au K-mirror, labeled with grazing angles on each mirror surface

The plastic mount is attached to a rotation stage, a Newport RSP-2T, which allows for axial rotation of the K-mirror, which is especially useful for the polarizer to adjust the polarization transmitted. This rotation mount is mounted to its tip/tilt mount to ensure alignment with the beam line. A mechanical vacuum feed-through is used to rotate the whole body under vacuum. Both mounts are placed on translation stages so each K-mirror can be moved out of the way under vacuum during measurements.

Figure 4.7 Photo of K-mirrors placed inside HHG chamber, Au K-mirror (Left Center), B4C K-mirror (Center) and gas jet (Right Center) are all shown, the fundamental beam comes from the right towards the camera on the left.

4.5 Alignment

The K-mirror assembly with three mirrors has 9 degrees of freedom (DOF). Two for each tip/tilt mirror mount for a total of four, two for the tip/tilt of the assembly with the rotation stage, one for the rotation
of the assembly, and two for the $x/y$ position of the assembly. With so many degrees of freedom, alignment proved to be a challenge. To aid with alignment, we printed an alignment tool to align the rotation stage in the $x/y$ plane and along the $z$ axis so that the beam is normal to the face and centered on the rotation stage.

After aligning the rotation stage, we used an iris immediately after the K-mirror and the position of the beam on the camera to center it, first adjusting the tip/tilt on the flexure mount on the first mirror to pass through the center of the iris, then adjusting the tip/tilt on the second flexure mount to bring the beam to the center of the camera, iterating through this process brings the beam quite close to alignment. Ensuring the beam is in the middle mirror’s center ensures that the incidence angles on the first and third mirrors are close to the intended design angle. This iterative process should bring it close to alignment, which may require further adjustment after rotating the K-mirror 90deg axially. Getting the beam to stay in alignment is quite difficult through the entire 360deg rotation because a small misalignment will compound through the reflections.

Balancing these DOF to return the beam onto the optical axis would take an extended amount of time, so we opted for aligning the system to the point that the EUV beam will stay on the camera through the full rotation of the Au K-mirror. Once the beam stayed within the camera’s center, we used code to realign and crop each image on the camera along the same relative position of the beam, outlined in Section 5.2.

4.6 Performance

Using a linearly-polarized fundamental beam sent through a half-wave plate, we can adjust the input polarization through the harmonic process (HHG preserves the linear polarization direction) and, therefore, the K-mirrors. Adjusting the input linear transmission allows us to find experimental values for the polarization contrast of S and P polarizations.

Figure 4.8 HHG chamber setup for analyzing K-mirror polarimetry
The measurements provided close contrast ratios to the expected, with the B4C K-mirror having a measured contrast ratio of 1.84:1 and the Au K-mirror having a contrast ratio of 68.64:1. This came close to the expected 1.74:1 for the B4C K-mirror and higher than the expected 38.23:1 for the Au polarizer.

### 4.6.1 Conversion to Circular Polarization

The wave retarder has an added challenge from typical birefringent wave plates in visible and IR; the wave retarder also acts as a polarizer to a small degree. While the phase shift of $\pi/2$ is the same as a quarter waveplate, the wave retarder will not act strictly as a quarter waveplate when converting incoming polarized light. The contrast ratio means that there is a favored axis of transmission that affects conversion to and from circularly polarized light.

Using the 9th harmonic Mueller matrix for the B4C K-mirror, we can find the optimal angle to convert linear to highly eccentric polarization. To find the optimal angle, we use the following Mueller matrix:

$$M_{B4C}(\theta) = \begin{bmatrix}
0.410943 & 0.106949 \cos(2\theta) & -0.213898 \sin(\theta) \cos(\theta) & 0. \\
0.106949 \cos(2\theta) & 0.410943 \cos^2(2\theta) & -0.205471 \sin(4\theta) & 0.793563 \sin(\theta) \cos(\theta) \\
-0.213898 \sin(\theta) \cos(\theta) & -0.205471 \sin(4\theta) & 0.410943 \sin^2(2\theta) & 0.396782 \cos(2\theta) \\
0. & -0.793563 \sin(\theta) \cos(\theta) & -0.396782 \cos(2\theta) & 0.
\end{bmatrix}$$  \hspace{1cm} (4.16)

where $M_{B4C}$ is the Mueller matrix for the B4C wave retarder at 44.4nm. Typically, a QWP aligned to ±45 deg will exchange 0° linear polarization with circular polarization. However, solving the wave retarder Mueller matrix for maximum conversion from 0° polarized light gives an orientation of ±38° (from the P polarization axis) instead of ±45°. To measure the conversion to circular, we set the input polarization to several angles leading up to the optimal 38°, then rotated the Au K-mirror polarizer through 360° behind the B4C K-mirror. The measurements with the Au K-mirror show the polarization ellipse of the light after
the B4C, a pure circle in this measurement means a higher conversion to circularly polarized harmonics.

![Figure 4.10](image_url)

Figure 4.10 Relative power throughput as a function of Au polarizer angle for various input polarization orientations. This plot shows the high conversion efficiency of the linearly polarized harmonics near the 38° input polarization. The radial scale is arbitrary between plots; this figure is meant to show the purity of the circular polarization state.

The data showing the circular polarization (Figure 4.10 shows conversion to a high ellipticity. However, this optic should be capable of even higher eccentricity than what is shown for the 38° measurement, which the reduced eccentricity is likely due to a sub-perfect alignment of the input polarization with the faces of the B4C k-mirror. This misalignment can be seen with the 0° input measurement not perfectly aligned along the x axis.

The benefit of the B4C wave retarder, from other existing polarization optics, is the high conversion efficiency. We achieved a power transmission through the k-mirror of 37% when converting to circular polarization, which is significantly higher than existing optics. Although this high conversion efficiency is also a result of the specific wavelength we are using. Since most HHG and MCD measurements are performed with higher photon energy light, nearer to many materials’ M-edges, a lower conversion efficiency is expected for a wave retarder designed for those wavelengths.
This section outlines the added challenges from the K-mirror and the EUV wavelength range to retrieve a polarization measurement of the harmonics. These challenges include:

- Image flipping and rotation from an odd number of reflections
- Beam pointing due to slight misalignment with the K-mirrors
- Contrast difference between S and P polarizations on the B4C wave retarder
- Image quality defects due to dust/scratches on the mirror surfaces.

While most of these issues can be solved using clever data processing, several of these issues require more data collected beyond just the four intensity measurements for classical Stokes polarimetry, such as the case in Chapter 3.

5.1 HHG Polarimetry Setup

The HHG chamber setup to perform Stokes polarimetry on the high harmonics is the same as in Section 5.2, with the Al filters after generation, then the B4C k-mirror and the Au k-mirror. The primary difference is the S-waveplate after the half waveplate to generate the vector beam in the fundamental 400nm light. Leaving the HWP in allows the adjustability of the polarization rotation from the vector beam, switching between azimuthal and radial polarizations.

Using the rotation stage on the Au K-mirror to rotate the passed harmonic polarization, we took images of the beam at the camera through the entire rotation in 45° increments. The full rotation images were taken with and without the B4C wave retarder. Additionally, the harmonic beam’s intensity profile was measured without any polarization modification before and after the images with the k-mirrors in place to get an idea of the polarization profile without the distortion from the reflectors.

The first two issues with using the K-mirrors are the beam pointing on the camera from slight misalignment and shifting the beam’s position. The second issue comes with the odd number of reflections on the K-mirrors. The image is flipped along a new axis with each Au K-mirror rotation. Both beam positioning issues are easier to fix within the data processing than with system design.

The reasoning behind taking data through the entire 360° rotation of the Au polarizer is to get measurements of the same polarization state from a different location on the reflectors. From our
experience, the K-mirror reflectors have an affinity to collect dust on the surface, which becomes an issue when the EUV light diffracts from the dust particle and messes with the intensity profile of that measurement. The motivation is that to reduce this effect, we can utilize the small amount of beam pointing in the system to shift the position of dust on the beam image between one measurement and the 180° complement. If we can superimpose these two images, we can average them to minimize the impact of the distortions.

Figure 5.1 Sample raw images from the camera for the polarizer at angles of 90° to -45°, and again with the B4C “waveplate” in place. Different beam positions can be seen through the polarizer’s rotation and reflection across the vertical axis when the B4C K-mirror was added. Images were also taken with the polarizer at 180° complements of each angle, resulting in 8 more images.

5.2 Data Processing

Each raw image was initially processed separately. First was normalizing the intensity to the exposure time on the camera by dividing the intensities by the exposure time (typically 10-15 seconds). Then, we removed background noise by subtracting the modal intensity value from the image. The modal subtraction was accompanied by further subtracting 1% of the image’s peak intensity to ensure further noise removal. This 1% of peak power becomes the new zero intensity point of the image. This background reduction process gave good results, keeping much of the low-intensity harmonic light while removing virtually all the noise from the images. The later DOP analysis in Figure 5.4 shows a good relation between intensity and the degree of polarization in the beam, which indicates a low quantity of background signal.
After the background subtraction, we applied a Gaussian filter with $\sigma = 10$, cleaning up the intensity variations and speckled appearance from dust and scratches on the reflectors. The Gaussian filter worked well to remove most of the issues with beam quality. To be able to find the polarization states from the beam, we need to be able to correct the reflections and crop each image individually to account for the beam wandering on the camera.

The reflections were removed using image transformation tools. For the measurements with only the polarizer, each image was flipped along the axis of measured polarization, undoing the flip from the odd reflection. The image flip aligns the orientation of the beam in the image to the orientation without the k-mirror in place. For the measurements with both the polarizer and the wave retarder, the images were flipped twice, once about the $y$ axis and once about the polarizer direction. To verify that this process worked successfully, we used a stage to clip the EUV beam after the filter and took duplicate measurements with the polarizer and wave retarder. This validation ensured that after the image flipping, the image of the clipped beam was in the same orientation in each image. After the intensity images were all in the same orientation, I worked to crop the images down to the same envelope to contain the beam.

To crop each image down to the same window, I used the primary intensity measurement of the beam with no polarization optics as a reference. First, I cropped this image to a 500x500 area in the center of the image, which contained the high-intensity harmonics and a large enough area around it to see the lower-intensity, higher divergence harmonics. I then used the reference as a “filter” over the position of each 500x500 crop window and found the maximum overlap between the intensity profile of the polarization measurement and the reference profile. The idea here is that the polarization optics cannot image and re-form the beam’s intensity profile; it can only attenuate certain polarizations from the intensity profile, meaning that each polarization measurement will fit neatly within the intensity profile of the beam.

Following this process of flipping the images and finding where the beam lies, the images line up to what we would expect. Now that the images are all in the same superposed location, I averaged the redundant images with the same measurement (i.e., $0^\circ$ and $180^\circ$ polarization measurements were averaged).

Now the intensity profiles for polarization at $0^\circ$, $45^\circ$, and $90^\circ$, we can find the $S_1$ and $S_2$ parameters. Finding these Stokes parameters follows Equation 2.19 due to the high performance of the Au polarizer. To get a cleaner intensity profile, we can use the beam images without the polarizer as our $S_0$ parameter.

Getting the $S_3$ parameter proves to be more difficult, primarily because of the contrast on the k-mirror. However, because of the measurements performed with the wave retarder in the beam and the polarizer moved through a full rotation ($0^\circ$ WP and $90^\circ$ WP in Figure 5.1), we can retrieve the contrast ratio and use it to get the profile of $S_3$. Using the intensity differences between the $0^\circ$ with WP and the $0^\circ$ without WP measurements, we can find the contrast along that polarization axis of the k-mirror, in this case, the P
polarization. We can use this transmission ratio ($T_P$) as a weighting function for the circular polarization measurement.

$$S_3 = I_{45^\circ WP} - (T_P I_{0^\circ} + T_S I_{90^\circ})$$  

(5.1)

5.3 Results

Using the process above, with confidence, we were able to retrieve the polarization state for multiple harmonic polarization profiles using different pumping profiles. The stokes parameters for the different polarization profiles are seen in Figure 5.2, with additional profiles in Appendix A.

![Radially Polarized Vector Harmonics](a)

![Azimuthally Polarized Vector Harmonics](b)

Figure 5.2 Full-beam Stokes parameters for (a) radial and (b) azimuthal vector harmonic beam

Using the half waveplate to rotate the polarization profile of the beam, we selected specific positions to generate radial and azimuthal vector beams to generate with. These fundamentals produced the same profile in harmonics, visualized with the plots in Figure 5.3.

I suspect that the issues with the measurement for DOP have to do with the misalignment of the images. The issue could be that when finding the maximum convolution of the beam with each image, there is a match of the highest intensity, which is not necessarily the correct position. Looking at the $S_2$
Figure 5.3 Polarization profile of (a) radial and (b) azimuthal vector harmonic beam. Polarization ellipses are plotted over logarithm of intensity to show the polarization profile extension into the lower intensity portions of the beam.

parameter for the azimuthal beam (in Figure 5.2), no clear lobes are seen in the radial case. This issue carries through the calculations, causing the DOP measurement does not produce a nice singularity in the center, as the radial case does. Instead, the DOP plot has a strange triple lobe pattern and exceeds a DOP of 1 in the upper right of the beam. This leads me to believe that there is significant room for improvement in the beam-pointing correction.

5.4 Discussion

The utility of this polarization characterization of harmonics can immediately be used to characterize polarization profiles of harmonics. The polarization profile measurements can be used to optimize and characterize specialty pumping schemes with varying intensity and polarization profiles. The primary benefit of performing Stokes-type measurements instead of a typical polarimeter without the wave retarder is that the unpolarized light can also be characterized, which contributes to the overall power and would harm the accuracy of measurements with only a polarizer. One use case of characterizing unpolarized light is when looking at reducing specific phase-matching parameters in the gas. With certain pumping and gas profiles, large amounts of dispersed and incoherent EUV light can be produced, which could be characterized by this setup in order to optimize or reduce those characteristics of generation.

This characterization method could be expanded to look specifically at the polarization of power around the LG mode, to characterize the polarization and gain insight into the specific phase matching and generation parameters in the system.
Figure 5.4 degree of polarization for (a) radial and (b) azimuthal vector harmonic beam. Some issues with (b) is a large area of DOP above 1, as well as the lack of a clear singularity, as seen in the intensity profile.
Ultimately, this thesis outlined the methodology and optics for use in characterizing the polarization of HHG EUV. The benefits of this work allow for the characterization of high harmonic light, explicitly being able to interface back and forth between circularly polarized harmonic light using the B4C wave retarder.

The completed work in this thesis included the following:

1. A clear demonstration of Stokes full beam polarimetry
2. A method to design EUV reflective polarization optics for optimization at specific wavelengths and broadband performance over several harmonic orders.
3. Demonstrated the use of the EUV polarization optics to characterize the polarization profile of harmonic light.

Already, Polarization measurements on harmonic light could be extended to current projects with our HHG chamber. The polarimetry be used to validate polarization-dependent information, such as finding the polarization states of retrieved mutually incoherent channels in HHG ptychography measurements, which David Schmidt has been exploring and working with. This work can be extended into other measurements and characterizations, including MCD, polarimetry of novel generation schema, and polarization validation of harmonics.

6.1 Magnetic Circular Dichroism

Magnetic Circular Dichroism is a phenomenon in which magnetic materials have a contrast difference with circularly polarized light when exposed to a saturating magnetic field. Measurement of MCD using reflective polarization control has been used for many different metal types on thin-film samples [18].

Magnetic Circular Dichroism is a straightforward application for circularly-polarized EUV light, which benefits from high-efficiency conversion or generation to circularly-polarized harmonics. While the design wavelength for the B4C K-mirror is not optimal for the wavelengths of the Fe and Co MCD edges, the benefit of using an optic such as this is that it can be designed for specific wavelengths by adjusting the angle on the first mirror.

The sample we obtained is a thin layer of Fe deposited on a Si substrate acquired from collaborators at NIST. While we ultimately could not retrieve a MCD measurement using this sample, we built expertise and experience working with polarimetry and conversion into circular harmonics with the k-mirrors.
Looking at the results from other sources within this wavelength range [7], we should see a 1-2% change in the transmission through the sample at 44.4nm. The main issue we struggled with was system instability on the order of 1-5%, which did not allow us to find the contrast difference on the order of 1%. Another factor harming our measurement is the wavelength, 44.4nm, which means that we are not on the M Iron edge for MCD (at 24nm, 52eV), which gives a higher contrast ratio of 10-20%.

A potential avenue to move forward with MCD measurements is to use 800nm fundamental light to generate harmonics, then use a modified B4C mirror to optimize conversion at the 21st harmonic of 800nm (38nm), which lies close to the M edge of Germanium, allowing us the possibility of a more pronounced contrast difference. The modified k-mirror is as simple as adjusting the grazing angle on the first reflector to around 16deg instead of the 12deg optimized for 44.4nm.

6.2 Higher-order vortex beams

The most straightforward expansion of this work is to follow the same process and measurements as outlined in this thesis and characterize higher-order LG mode harmonics [19]. This is quite as simple as using a higher-order S-waveplate to create higher-order vortex beams for the fundamental. We already have a second and seventh-order S-waveplate for this purpose; the work for this measurement could be as simple as swapping in the S-waveplates before the lens and taking equivalent measurements as before. The differences in this work would come in with the profile of the beams after the polarizer, passing HG\textsubscript{11} modes or other high-order spatial modes.

6.3 Polarimetry of OAM HHG

Another experiment we have been planning and exploring is the generation of harmonics using two crossed beams with +1 and -1 orders of orbital angular momentum (OAM); this will generate harmonics with spatially-separated +1 and -1 OAM orders, similar to work in [7], but expanding beyond just circularly-polarized pumps and moving into using OAM pumps.

Already, polarization characterization of the two 800nm beams is proving beneficial to troubleshoot issues with beam interference and eccentricity at focus. Polarization analysis of the OAM harmonic light could provide insight into issues with a generation or characterize the polarization profile of single-order OAM HHG.
Figure 6.1 Upstream optics for NCP-HHG separating linear polarized fundamental into spatially separated beams of RCP and LCP, brought back together into the focus.
REFERENCES


APPENDIX A
ADDITIONAL CHARACTERIZED HHG BEAM PROFILES

The results shown in Section 5.3 were also found for two additional beam profiles, to verify that the methodology was capable of producing polarization profiles for beams outside of well-optimized vector beams as shown in Chapter 5. The two additional harmonics beams characterized were generated by the following:

1. an offset of the S-waveplate.

2. the same offset of the S-waveplate with added 0.03 waves of trefoil from the aberration control on the deformable mirror.

Figure A.1 Full-beam Stokes parameters for harmonics generated with (a) offset S-waveplate and (b) offset S-waveplate with added aberrations.
Figure A.2 Polarization profile for harmonics generated with (a) offset S-waveplate and (b) offset S-waveplate with added aberrations.
APPENDIX B
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B.1 Wikimedia Commons

Figure 2.1 by SuperManu - Self, based on Image:Onde electromagnetique.png, CC BY-SA 3.0, https://commons.wikimedia.org/w/index.php?curid=2107870

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