Polarization Dependence of High Order Harmonic Generation from Solids in Reflection and Transmission Geometries

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POLARIZATION DEPENDENCE OF HIGH ORDER HARMONIC GENERATION FROM SOLIDS IN REFLECTION AND TRANSMISSION GEOMETRIES

by

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University of Central Florida, 2020

A thesis submitted in partial fulfillment of the requirements for the Honors in the Major Program in Physics in the College of Sciences and in The Burnett Honors College at the University of Central Florida Orlando, Florida

Spring Term
2020
ABSTRACT

High harmonic generation (HHG) is a process that occurs when an intense laser interacts with a material and generates new frequencies of light. HHG has many practical applications, namely as a spectroscopy technique and source for high frequency light and attosecond pulses. While HHG has been done extensively in gases, HHG in solids is a relatively new field. Solids are appealing as an HHG medium as they require much simpler equipment and are subsequently much more compact, and thus may have a variety of applications previously inaccessible to gas-phase HHG. However, the generation mechanism of HHG in solids has not been fully characterized yet, as the processes behind HHG in gases and solids are not synonymous. Here, we study the influence of polarization, symmetry, and setup geometry on HHG in solids. We study the propagation effects in a transmission geometry setup and use Jones calculus to counteract the polarization change from propagation. We compare these results to a reflection geometry setup, which naturally does not have propagation effects, to determine the validity of the polarization correction technique. We also look at the electric field symmetry dependence on HHG through the manipulation of the laser electric field with a two-color interferometer. The impact of symmetry dependence and propagation effects both contribute to a better understanding of the HHG process in solids.
Acknowledgements

Above all, I would like to thank my thesis committee chair and advisor, Dr. Michael Chini. Without your feedback, time, and availability to answer questions, I would not have the understanding and skills that I do today.

Thank you as well to Dr. Stephen Kuebler for his support as a member of my thesis committee.

I would also like to thank the LUMAS research group for their assistance and guidance throughout my time in the group, particularly Shima Gholam Mirzaeimoghadar. Thank you for teaching me how to approach experimental techniques and troubleshooting experiments with me.

Thank you also to my family for the years of support, and to my friends for the board games and stories. Thank you for giving me many, many reasons to smile.

Finally, this work is supported by the Air Force Office of Scientific Research under award no. FA9550-16-1-0149, the National Science Foundation under grant nos. 1806135 and 1809181.
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CHAPTER 1: INTRODUCTION

High harmonic generation (HHG) is a nonlinear process that occurs when an intense laser interacts with a material and generates new frequencies of light, which are harmonics of the driving laser frequency. Understanding the factors that may influence HHG is important for application of HHG and control of HHG signal. In this thesis, we study the influence of polarization, symmetry, and setup geometry on HHG in order to better understand the HHG mechanism in solids.

Chapter 1 describes the history and significance of HHG, and discusses the more recent development of HHG in solids. Chapter 2 details the current explanations of the HHG process in both gases and solids, then discusses the nature of even and odd harmonics as well as the dependence on both symmetry and polarization. Chapter 3 summarizes equipment and experimental methods used. Chapter 4 discusses a transmission geometry setup, which involves the use of Jones calculus to counteract propagation effects and shows the results of HHG in this setup. Chapter 5 describes and shows HHG results for a reflection geometry setup. Chapter 6 compares the reflection and transmission geometry measurements and discusses the implications of each setup as they relate to each other. Chapter 8 studies the impact of symmetry through polarization using a two-color interferometer. Chapter 9 concludes and summarizes the work described in this thesis, and references are listed in Chapter 10.

This work was done in the LUMAS group in the Physics Department of the University of Central Florida. All work presented in this thesis was collaborative between
myself, Shima Gholam Mirzaeimoghadar, Troie Journigan, Zain Khan, Mamta Singh, John Beetar, and Dr. Michael Chini. At the time of writing, the results of the Jones calculus work have been accepted to the UCF Undergraduate Research Journal for publication.

1.1. History of High Harmonic Generation

High harmonic generation to the 11th order was first seen in 1977 in a plasma generated from solid Al1. Perturbation theory predicted that the harmonic signal would be proportional to $I^q$, where $q$ is the harmonic order and $I$ is the driving laser intensity. However, when HHG in noble gases greatly increased the harmonic order2, 3, a better theoretical understanding was needed to explain the nonperturbative nature that was seen in these high order harmonics2, 4. Instead of an $I^q$ dependence, high-order harmonics exhibit a region of constant intensity with respect to the harmonic order (the plateau region) followed by a sharp drop in intensity (the cutoff). This theoretical development resulted in the 3-step model, which is described in Section 2.1. Many experiments have contributed to the current understanding of the generation mechanisms behind HHG, which has involved, for example, studying the harmonic dependence on atomic density5 or the impact of the dipole moment6-8. At the same time, HHG has been developed for use in various applications, which are discussed in Section 1.2.

Gas-phase HHG experiments need equipment to control the gas (such as vacuum chambers or gas cells) and high intensity lasers, which limits gas-phase HHG to a laboratory setting. These limitations motivated the development of HHG in crystals9, which is currently an area of interest. Solid-state HHG has several benefits over gas-
phase, such as compact setups, minimal equipment requirements, and increased conversion efficiency\textsuperscript{10, 11}. However, HHG in solids required a different theoretical explanation that what had been previously accepted, as the generation mechanism in gases could not be directly applied to solids. The band model (described in Section 2.1) was developed as a bridge between the more familiar three-step model and the new solid-state theory based on semiconductor Block equations and experimental results.

1.2. Significance of High Harmonic Generation

HHG has applications in many different capacities. HHG is favored as a source for femtosecond to attosecond pulses\textsuperscript{12} and has consequently led to several different measurement techniques that take advantage of variations in the polarization, duration, and repetition of generated harmonics\textsuperscript{13}. In addition, high harmonic spectroscopy can be used in time resolved measurements\textsuperscript{14, 15} to study atomic and molecular structure and dynamics\textsuperscript{16-20}.

The development of solid-state HHG allowed for the generation of XUV light within a vacuum chamber and the generation of visible and ultraviolet light in compact, free space setups. These wavelength ranges in addition to the denser nature of solids was found to increase conversion efficiency\textsuperscript{10, 11}, making solid-state HHG a desirable option for high-powered tabletop light sources. In addition, since the generation process itself encodes information about the material, the harmonics can give insight about crystal and molecular structure\textsuperscript{11, 21, 22}. 
CHAPTER 2: FUNDAMENTALS OF HIGH HARMONIC GENERATION

The HHG process can be described semi-classically. While the generation mechanism in solids and gases are different, both mechanisms are discussed here for comparison, before detailing several of the factors influencing harmonic signal.

2.1. Recollisional Model

The HHG process can be described semi-classically in what is commonly referred to as the recollisional model or the three-step model. The gas-phase HHG process starts with a short-pulse laser focused onto a gas target. This begins electron movement that is visualized in Figure 1a. The electron tunnels through the binding energy of the parent atom, typically at the peak of the electric field, and begins to travel away from the parent ion. When the electric field changes direction, the electron is pushed back to the parent ion and emits a photon based on the energy that the electron gained while traveling in the electric field.

In solid-state HHG, the electron cannot travel freely, as it is influenced by the lattice potential even after being excited by the laser. This process thus requires different explanation than the HHG process in gases. HHG in solids can be separated into two generation mechanisms referred to as interband and intraband processes. In the interband process (Figure 1b), the electron is excited into the conduction band by the photon, therefore jumping the band gap of the material and leaving a hole in the valence band. The electron and hole travel along their respective bands, and the electron eventually drops back to recombine with the hole, emitting a photon. The photon energy
is based on the energy difference in the band gap at the location of recombination, which provides an inherent limitation to the energy of the emitted photons\textsuperscript{25}. In the intraband process (Figure 1c), vibrations of the electron after being excited into the conduction band produce enough energy to release a photon without the electron transitioning down to the valence band. These two processes both contribute to measured harmonic signal, where the dominating process depends on a variety of conditions, such as the material, harmonic order, and driving laser\textsuperscript{24-27}.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Depiction of the a) 3 step model in gases, b) the interband process in solids and c) the intraband process in solids.}
\end{figure}
2.2. Even and Odd Harmonics

Even and odd harmonics are one of the most fundamental features of HHG. As the names suggest, odd harmonics have frequencies that are odd multiples of the fundamental laser frequency $\omega$ (such as $3\omega$, $5\omega$, $7\omega$...) while even harmonics are even multiples of the fundamental laser frequency $\omega$ (such as $2\omega$, $4\omega$, $6\omega$...). The symmetry of the HHG system determines which harmonics are generated. As previously discussed, the electron will tunnel from its place in the atom around the peak of the electric field of the laser, which can be described as a sine wave. In a symmetric system, this tunneling occurs at point 1 in Figure 2a. As the electric field changes direction, the electron changes direction and recombines (Figure 2a, point 2). The excitation and recombination process will happen at approximately every peak and trough. Therefore, the electrons will tunnel away from the parent ion at approximately points 1, 3, and so on in Figure 2a, then recombine approximately at points 2, 4 and so on in Figure 2a, releasing a photon when they do so. The resulting pulses from the recombination can be written as a train of Gaussian pulses in time:

$$f(t) = \sum_{n=0}^{\infty} A e^{-a(t - n\frac{T}{2})^2}$$

(1)

where $a$ controls the width of the pulse, and the subtracted term in the exponent (here $\frac{nT}{2}$), determines the separation of pulses. As $T$ refers to the period of the laser, these pulses are defined as being separated by half the period of the laser (Figure 2b). The intensity $A$ of the pulses is considered to be constant for all pulses, as a simplification for
ease of discussion. Taking the Fourier transform of these pulses in time results in a series of Gaussian pulses in frequency:

\[ f(\omega) = \sum_{n=0}^{\infty} A e^{-\frac{(\omega - \frac{n\omega}{2})^2}{4a}}. \]  

(2)

When graphed, as in Figure 2c, this series of pulses in frequency show the harmonics that occur with spacing of \( 2\omega \), or odd harmonics. Therefore, when the system is symmetric, electrons tunnel identically at every trough and peak and there are pulses in time every half-period of the laser, which corresponds to odd harmonics in frequency.

This outcome changes when there is asymmetry in the system. There can be many types of broken symmetry, such as the crystal structure and uneven electric field. A more in-depth discussion of the crystal structure will be discussed in Section 2.4. The situation of an uneven incident electric field is used here as a simple case of asymmetry in the system. The process begins in the same way as before, where the electron tunnels at approximately the peak of the laser (Figure 2d, point 1) and recombines later (Figure 2d, point 2). However, in this situation, the negative portion of the electric field may not have enough intensity to cause an electron to tunnel (Figure 2d, point 3). Therefore, there are only pulses emitted once every period of the field, described by

\[ f(t) = \sum_{n=0}^{\infty} A e^{-a(t-nT)^2}. \]  

(3)
where the pulses are emitted every period of the electric field (Figure 2e). This time, a Fourier transform of this results in

\[ f(\omega) = \sum_{n=0}^{\infty} A e^{-\frac{(\omega-n\omega)^2}{4a}} \]  

which has harmonics of spacing \( \omega \), resulting in both even and odd harmonics (Figure 2f).

Figure 2: Visualizing how symmetry affects harmonic signal.
2.3. Creating Uneven Electric Fields with a Two-Color Interferometer

A simple way to create the uneven electric fields discussed in the previous section is to use a two-color interferometer, which relies on the interference between two electric fields of different frequencies. Two sine waves, when added together, may constructively or destructively interfere, depending on the phase delay and the frequency of the two waves. Figure 3 shows the interference from two waves of the same frequency but with different intensities. When in phase, the two waves (shown in Figure 3a as dotted red lines), constructively interfere to increase the intensity of a wave with the same phase as the two interfering waves (shown in Figure 3a as a solid black line). When partially out of phase (Figure 3b), the two waves result in a wave with a change in intensity and an phase shift away from either of the initial waves (Figure 3b).

![Figure 3: Depiction of interference from two identical wavelengths of different intensities. The two interfering waves are shown in red dotted lines, while the combined wave is shown in black.](image)

Summarizing, changing the phase delay between the two interfering waves simply changes the resulting phase and intensity of the combined wave. The resulting wave has
positive and negative components that have an identical magnitude. However, by undergoing the same interference process between two waves of frequencies $\omega$ (the fundamental beam) and $2\omega$ (the second harmonic of the fundamental), we can produce a wave that is no longer symmetric. The symmetry varies with delay between the two contributing waves, as seen Figure 4. The resulting wave can be skewed (Figure 4a) or have very unequal positive and negative contributions (Figure 4b). The intensity of the second harmonic is greatly exaggerated here for clarity, as in practice, a perturbation of a second harmonic that is several orders of magnitude less than the fundamental will produce enough broken symmetry to generate even harmonics $2^1$.

![Figure 4: Calculations displaying potential combinations (solid black line) of the fundamental (dotted red line) and second harmonic beam (dotted blue line). Variations are due to the phase difference between the fundamental and second harmonic beam.](image)

Uneven electric fields can be generated experimentally with a two-color interferometer. An interferometer splits a single light source between two paths. When the light recombines, there is an interreference based on the difference in path length, producing fields similar to what is shown in Figure 3. In a two-color interferometer, a
nonlinear crystal is used to generate a second harmonic from the fundamental. The fundamental and second harmonic are split apart using dichroic mirrors or other filtering techniques. By varying the delay between beam paths, we can achieve the varying levels of symmetry seen in Figure 4. This two-color technique has been used to map the band gap of materials, study the electron behavior, and enhance and control HHG signal.

2.4. Structural Dependence

While uneven electric fields are a simple case for describing symmetry, there are other methods of controlling the symmetry. A prominent example is the configuration of atoms in a crystal. Along different axes of the crystal, the atomic interactions within the structure result in symmetric and asymmetric tunneling forces, depending on the axis of the crystal that the interaction is occurring along.

We use the semiconductor ZnO in our experimental setup, which has a hexagonal wurtzite structure (Figure 5a). Two common cuts of ZnO are a-cut and c-cut, shown in Figure 5b and 3c respectively. When the laser polarization is vertically incident on the a-cut crystal face (Figure 5a, Pol A), the electrons see a pattern of Zn and O atoms above and below with differing bond lengths. This breaks the symmetry of the system, as the force on the electron from Zn and O are different, and even and odd harmonics are generated. However, if the system is rotated such that the polarization is horizontal (Figure 5b, Pol B), the system is symmetric along the laser polarization and only odd harmonics are generated. In contrast, in a c-cut crystal (Figure 5c) all atoms in a layer
are the same, so the system will appear symmetric regardless of the orientation between the polarization and crystal (Figure 5c, Pol A or Pol B). Therefore, a c-cut crystal will produce only odd harmonics regardless of crystal angle with respect to polarization, and will never produce even harmonics just through crystal rotation. The angle dependence, or lack thereof, gives insight to the structure of the crystal, the generation mechanism \(^{34,35}\), and offers a method of controlling harmonic signal \(^{36}\).

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**Figure 5**: a) ZnO crystal, with shaded region showing an b) a-cut and c) c-cut and the potential polarizations incident on the crystal. In all diagrams, the Zn is shown in blue and slightly larger than the green O.

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2.5. Ellipticity Dependence

There has been much past study on the polarization dependence of harmonic signal. Gases have been seen to exhibit a strong dependence on ellipticity of the driving
laser\textsuperscript{37-39}. Indeed, this strong dependence has been used as a gating mechanism for HHG, contributing to the development of isolated attosecond pulses\textsuperscript{40}.

However, this is not the case in all situations, especially in solid-state HHG. While the ellipticity can reduce the harmonic signal\textsuperscript{41, 42}, for some materials, an elliptically polarized driving laser can result in an enhancement in the harmonic signal\textsuperscript{43}. This enhancement can vary based on the harmonic order, ellipticity of the input laser, and contribution of the inter- and intraband harmonics\textsuperscript{44, 45}. Additionally, a driving laser with a large ellipticity has been shown to generate circularly polarized harmonics in solids\textsuperscript{46, 47} which has contributed to discussion on the generation mechanism behind HHG\textsuperscript{48}. Circularly polarized harmonics can be used to study molecular structures\textsuperscript{49, 50}, and therefore the mechanisms to generate circular or elliptically polarized harmonics are an area of interest\textsuperscript{51-53}. Due to the diverse effects of polarization of harmonic signal, an understanding of these effects from both an experimental and theoretical point of view is important. The experimental work presented in this thesis contributes to this understanding through documentation of the effects of setup condictions and consequent polarization conditions on the HHG signal.
CHAPTER 3: EXPERIMENTAL PARAMETERS

This work was completed in the LUMAS lab at the University of Central Florida. Our experiment uses a commercial OPA (Light Conversion ORPHEUS-ONE) pumped by a 20 W Yb:KGW (Ytterbium-doped Potassium Gadolinium Tungstate) regenerative amplifier (Light Conversion PHAROS) at repetition rate of 50 kHz. The OPA idler output pulses of about 90 fs. All experiments were completed in the mid-IR range. The beam size was increased with a telescope for a magnification of 2.5x. AR-coated Si lenses were used to focus on the crystals. Harmonics were measured with a UV-enhanced high-resolution spectrometer (Ocean Optics HR2000+ES) after being focused by an aluminum mirror. Ellipticity measurements were taken with a power meter (Newport 843-R) and an linear absorptive polarizer.

We used a 300 µm a-cut ZnO crystal from University Wafer. The harmonic measurement in transmission without any polarization corrections was done at a wavelength of 3.8 µm. The reflection and Jones calculus measurements were done at a wavelength of 3.4 µm, which corresponded to the coatings on the wave plates used in the transmission geometry setup, as the wave plates were low-order and designed for use at 3.5 µm. The wave plates had high transmission (greater than 95% over the laser wavelength range) and used a MgF2 substrate, which is less susceptible to nonlinear propagation than Si substrates.

Calculations for the Jones calculus experiments were done in Python. Although calculations were done with zero as the positive x-axis according to mathematical standard, all numbers and graphs are presented for the reflection and transmission
geometries measured from the c-axis of the crystal, which is conventionally labeled as 0 degrees.

For the two-color interferometer measurements, we used an AGS crystal for the second harmonic generation and were able to generate strong signal without focusing on the back of the AGS crystal. We used an automated translation stage (ThorLabs K-Cube controlled with Kinesis) that could move with micrometer precision. We used dichroic mirrors (Omega Optical) that had a measured reflectivity above 97% from 3000 nm to 3500 nm and an average transmission of above 85% for 1500-2000 nm. Because of these mirrors, we used a fundamental wavelength of 3200 nm. Additionally, we used a c-cut ZnO crystal for these measurements.
CHAPTER 4: TRANSMISSION GEOMETRY

A transmission geometry HHG setup most simply involves a laser passing through the crystal, focusing on the back surface of the crystal, and generating harmonics in the last layers of the crystal. Any harmonics generated in the rest of the thickness of the crystal are absorbed by the crystal, such that the only meaningful contribution to the harmonics after the crystal are ones generated in the last tens of nanometers of the crystal\(^9\).

4.1. Overview

An example of a setup is seen in Figure 6. There is an angle dependence between the polarization of the laser and the crystal axis, requiring that either the crystal be mounted such that rotation is possible or that the laser polarization can be rotated. A transmission geometry setup such as this one is incredibly simple, making this setup very appealing for a variety of experiments.

Figure 6: A simple transmission geometry setup.
Figure 7: Harmonic orientation measurement of a transmission geometry setup with a bulk ZnO crystal. Plotted in a normalized logarithmic intensity scale.

A simple angle dependent measurement of the high order harmonics can be seen in Figure 7. In this scan, we see high order odd and even harmonics along 0 degrees, and only odd harmonics along 90. This matches with the expected symmetry dependence of ZnO discussed in previous sections and with past experimental work\cite{9,22}. However, we also see signals from 60 to 75 degrees that do not match the expected orientation dependence. These features will be discussed further in Chapter 6.

A transmission geometry utilizes a very simple setup. There are minimal optical elements required and transmission HHG can easily occur at normal incidence. One of the largest downsides to a transmission geometry are the propagation effects from the laser traveling in the crystal. Many crystals are birefringent, such that a linear polarization does not remain linear during propagation through the crystal. This occurs because the
polarization components develop a phase delay that is impacted by the thickness of the crystal and the difference between the two indices of refraction of the crystal. As the HHG process occurs in the last layers of the crystal, the polarization changes during propagation through the crystal may impact the harmonic signal in an unknown capacity, and this has been seen to reduce the harmonic intensity in ZnO crystals. Thin film crystals minimize propagation effects, but not all crystal structures are compatible with thin films. Similarly, a reflection geometry has no propagation effects, but has other complications that are not always ideal (discussed further in Section 5.1). One solution to these counteracting these undesirable propagation effects in transmission is a Jones calculus approach to controlling polarization in the crystal.

4.2. Jones Calculus

Jones calculus uses 2x2 matrices to describe how an optical component affects the polarization of light, which is represented by a vector. With simple matrix multiplication, an optical system can be reduced to a single matrix that describes the effect of the system on the polarization of the laser. The output polarization is found by multiplying the vector representing of the polarization with the effective matrix.

Jones calculus can be used to predict the polarization change due to the bulk crystal. This method begins by modeling a nonlinear, birefringent crystal as an arbitrary wave plate (verified in Section Jones Calculus results 4.3), which is given by

\[
J_{\text{crystal}} = \begin{bmatrix}
\cos^2 \theta_{\text{crystal}} + e^{i \Delta \phi} \sin^2 \theta_{\text{crystal}} & (1-e^{i \Delta \phi}) \sin \theta_{\text{crystal}} \cos \theta_{\text{crystal}} \\
(1-e^{i \Delta \phi}) \sin \theta_{\text{crystal}} \cos \theta_{\text{crystal}} & \sin^2 \theta_{\text{crystal}} + e^{i \Delta \phi} \cos^2 \theta_{\text{crystal}}
\end{bmatrix}
\]
where $\theta_{\text{crystal}}$ is the angle of the crystal axis to the polarization direction. The difference between the ordinary and extraordinary indices, which causes the polarization phase delay, is contained in the $\Delta \phi$ in Equation (5), where

$$\Delta \phi = \frac{2\pi d}{\lambda}(n_{\text{extraordinary}} - n_{\text{ordinary}}).$$

(6)

This $\Delta \phi$ contains the dependence on the physical features of the crystal and experimental conditions, including the thickness of the crystal, wavelength, and refractive indices.

With this assumption for the HHG crystal in place, the rest of the optical setup can then be built through simple matrix multiplication. In order to get any possible polarization at the exit of the crystal, a half wave plate (HWP) and quarter wave plate (QWP) were added to the system. A HWP rotates linearly polarized light, due to the phase delay of

$$\Delta \phi = \pi \pm 2m\pi$$

(7)

between the ordinary and extraordinary axes of a birefringent material, where $m$ is an arbitrary integer. By plugging this phase delay into Equation (5), the Jones matrix for a HWP is

$$J_{\text{HWP}} = \begin{bmatrix} \cos(2\theta_{\text{HWP}}) & \sin(2\theta_{\text{HWP}}) \\ \sin(2\theta_{\text{HWP}}) & -\cos(2\theta_{\text{HWP}}) \end{bmatrix}.$$  

(8)

A QWP is very similar, but instead converts linear polarization into circular polarization when the phase delay between the ordinary and extraordinary axes of a birefringent material is

$$\Delta \phi = \frac{\pi}{2} \pm 2m\pi.$$  

(9)
This technique can again be used to form the Jones matrix, this time for the QWP

\[
J_{QWP} = \begin{bmatrix}
\cos^2 \theta_{QWP} + i \sin^2 \theta_{QWP} & (1 - i) \sin \theta_{QWP} \cos \theta_{QWP} \\
(1 - i) \sin \theta_{QWP} \cos \theta_{QWP} & \sin^2 \theta_{QWP} + i \cos^2 \theta_{QWP}
\end{bmatrix}
\] (10)

With these three optical elements, all polarizations can be produced at the exit plane of the crystal by changing the angles of the waveplates as the crystal changes angle. This allows us to account for the polarization change caused by the birefringence of the bulk crystal, therefore giving better control over the polarization used to generate harmonics.

4.3. Jones Calculus Results

Before taking harmonic measurements or using the half- or quarter- wave plates, we first verified that Jones calculus could be used to model a ZnO crystal as an arbitrary wave plate while the crystal is under conditions necessary for harmonic generation. After sending vertical polarization through the ZnO crystal, we passed the modified polarization through a rotating, linear polarizer and measured the intensity that remained. We then used Jones calculus to model the same measurement, using the matrices for the arbitrary wave plate and a polarizer. The results can be seen in Figure 8. In these graphs, a range of output power from 1 to 0 corresponds to linear polarization, while any range smaller than this is increasingly circular polarization. We found good agreement between the calculated and experimental polarizations. From the data in Figure 8, we extracted the ellipticity and angle of polarization (Figure 9). Here, we can again see good agreement between total ellipticity change and angle of output polarization. We also see that as the ZnO angle increases to 45 degrees, the ellipticity increases as well to nearly 0.8, before
returning to linear light at a ZnO angle of 90 degrees. This result is important to note, as this suggests 45 degrees has the most correction necessary. The good agreement of these plots confirms the use of an arbitrary wave plate to act as a ZnO crystal when under conditions necessary for HHG.

Figure 8: Power measurement after linear light passes through a ZnO crystal of various angles from both a) Jones calculus calculations and b) a fit of experimental data. a) The calculations for 0 and 90 degrees resulted in the same polarization, and thus these two lines exactly overlap. b) A sample of raw experimental data is shown as points along the 45 degree measurement.

Figure 9: a) Ellipticity and b) angle of polarization measurements extracted from the polarizer scan in Figure 8. The angle of polarization is measured from the major ellipse of the light.
We then added the HWP and QWP in front of the ZnO and calculated the ellipticity at the exit of the ZnO after passing through all three elements. To visualize the result of these calculations, we plot the ellipticity as a function of the HWP and QWP for a given ZnO angle (Figure 10). We find that the HWP repeats every 45 degrees and the QWP repeats every 90 degrees. This behavior is expected based on the symmetry of the wave plates, and thus these plots provide additional confirmation of the calculations.

![Figure 10: Ellipticity calculations for possible configurations of a HWP and QWP for a ZnO angle of a) 0 degrees, b) 15 degrees, and c) 30 degrees. Bright spots represent the high ellipticity of light, while dark spots show low ellipticity (or more linear) light.](image)

As seen in Figure 10, the symmetry of the wave plates leads to multiple configurations of the setup that result in dark minimums. As we want to produce linear polarization after this setup, we were most interested in these minimums, though this plot could easily be used to select any desired polarization, and the symmetry of the wave plates would allow for multiple configurations for any polarization. To extract the possible values of the setup, we found the HWP and QWP combinations that resulted in ellipticity less than 0.0125. We considered this value to be functionally linear polarization, as the
calculations were done on a grid with accuracy of 1 degree to match experimental accuracy, and thus we were not able to achieve an ellipticity of exactly zero for most angles of ZnO. Indeed, a smaller ellipticity cutoff resulted in some ZnO angles not having any possible configurations that fit within the constraint. Table 1 shows some of the possible QWP and HWP angles for a given ZnO angle. From this list of possible configurations, we designed the experimental setup simply by selecting corresponding angles which required the least amount of adjustment, as all angles should work equivalently. A sample of selected angles are boxed and shown in a bolded font.

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A sample selection of angles is shown through bold and boxed numbers.
Using the calculated and selected angles, we set up our experiment and repeated the measurement done for Figure 8, which involved a polarizer and power meter after the setup. We found that we could achieve linear polarization at the output of the crystal (Figure 11). The largest deviations from linear polarization are present in ZnO angles from 30 to 60 degrees and 120 to 150 degrees, which is reasonable considering that those angles had the largest ellipticity change to counteract. Additionally, since our calculations were on a grid, there may have been slight adjustments to the configuration used that could have improve the linearity of the polarization that we did not have access to. This final measurement of linear polarization confirms the use of the Jones calculus as a technique for controlling the polarization at the output of a bulk crystal.

![Figure 11: Polarizer scan after the jones calculus corrections for a ZnO angle from 0 to 180 degrees](image)

To summarize the work done until this point, we have shown the use of Jones calculus to counteract the propagation effects within the bulk ZnO crystal in order to control the polarization at the exit of the crystal, which is the location of harmonic
generation. Thus, we are able to ensure that the harmonics from bulk ZnO will be generated with linear polarization.

We could now move forward with an angle dependent harmonic measurement. The results of this scan are shown in Figure 12. We see unexpected signal at 45 degrees compared to the measurement without polarization corrections before generation (Figure 7). As the difference between the two mentioned measurements are the control of polarization, this suggests that the extra features are polarization dependent. This will be discussed further in Chapter 6.

![Figure 12: Angle dependent scan of bulk ZnO when using the Jones calculus polarization corrections. Plotted in a logarithmic intensity scale.](image)

The low signal-to-noise ratio seen in this measurement may be from the waveplates necessary to correct the polarization. The large retardance (3/4 waves and
3/2 waves) and non-uniform retardance unintentionally lengthen the laser pulses and decreased peak intensity. The harmonics are dependent on the laser intensity, and thus the harmonics become weaker and the signal-to-noise ratio decreases.
CHAPTER 5: REFLECTION GEOMETRY

A reflection geometry for HHG uses the front surface of the crystal to generate the harmonics, thus generating the harmonics without the light having passed through the crystal. This removes any potential for propagation effects that are present in the transmission geometry.

5.1. Overview

A reflection geometry setup is most simply a laser focused on the front surface of the crystal at some incident angle. The harmonics are generated on the surface, then the beam is reflected and sent into a detector. A simplified example of potential setup is seen in Figure 13.

![Figure 13: Simple example of a reflection geometry setup.](image)

HHG in reflection is a versatile method for HHG, as harmonics can be generated in metals and other opaque materials\textsuperscript{55, 56}, not just in transparent crystals. In addition, reflection geometry is a surface interaction and thus is useful in surface measurements\textsuperscript{57}.
There is a dependence on both the angle of incidence as well as the angle between polarization and the crystal axis, which can both give insight about the generation process and role of symmetry. Perhaps most importantly, reflection geometry is free from propagation effects that may be present in transmission geometry, which can alter the harmonic signal.

Reflection geometry does have its own set of challenges. The intensity of reflected signals can be very small, which makes detecting the harmonics challenging. In addition, it has been shown that there may be power loss at shorter wavelengths, increasing the challenge of measuring strong harmonic signal. Thus, the setup design is very important, in order to maximize the amount of signal while also minimizing the induced errors in the experiment. For example, Figure 13 shows a reflection geometry that introduces a small angle, such that HHG no longer occurs at normal incidence, which may alter the signal, and indeed larger angles have been shown to result in a loss of power in the harmonic signal. However, this setup but can reflect the entirety of the beam without cutting the beam spatially and subsequently cutting the power. Other options, such as a dichroic mirror to separate harmonics from the fundamental, may not have good reflection or transmission at all wavelengths and therefore could block some of the harmonic signal. Overall, a reflection geometry setup can become quite complex. Within the harmonic signals themselves, it has been shown that a reflection geometry can introduce nonlinear reflection coefficients that can alter signal, similar to how propagation effects in transmission geometry alter signal.
5.2. Results

Our reflection geometry utilized a D-mirror placed between the focusing lens and the ZnO to reflect the harmonics into the spectrometer setup in a setup similar to Figure 13. The light was focused onto the ZnO crystal front surface and reflected onto the D-mirror by introducing a slight angle into the ZnO mount, though this angle was less than 5 degrees. We found that p-polarization incident on the crystal resulted in the highest intensity harmonics.

We took a rotational measurement of the harmonics, as seen in Figure 14. We see angle dependence in the low order harmonics and no angle dependence in the band gap signal, as expected, but an unexplained broad feature and the band gap signal obscure the 6th through 9th harmonics. The higher order harmonics also unexpectedly display no angle dependence. This is visible in the line plot of Figure 15a, where the 11th, 13th and 15th harmonics stay at a high intensity regardless of angle, while the 4th and 5th harmonics both show a variation as the crystal rotates. The lack of orientation dependence for the higher order harmonics is something currently unexplained and requires further investigation. The 5th harmonic has relatively small orientation dependence, likely because the intensity is so high, but there is a peak around 45 degrees in Figure 15a (also visible as a dark red in Figure 14). In an independent alignment of the setup, we were able to see the 7th harmonic, which was previously obscured by the broad feature. The spectrum of this alignment (Figure 15b) at angles of 0, 45 and 90 degrees shows the same extra feature at 45 degrees. Overall, these measurements show that a reflection geometry HHG does have additional features at 45 degrees for low-order harmonics,
similar to the high-order signals seen in the Jones calculus measurements. This will be discussed further in Chapter 6.

Figure 14: Orientation dependent scan in a reflection geometry setup with ZnO. Plotted in a logarithmic intensity scale

Figure 15: Lineouts of two alignments of the reflection geometry setup. a) There is a lack of angle dependence in the higher order harmonics compared to the lower order. The 5th harmonic, while saturated, displays a relative peak near 45 degrees. b) In an independent measurement, the 7th harmonic (around 500 nm) shows a smaller but still significant signal for 45 degrees, compared to the expected 0- and 90-degree signal.
CHAPTER 6: COMPARISON OF SETUP GEOMETRIES

In this chapter, Figure 7, Figure 12, and Figure 14 from other sections of this thesis have been reproduced in Figure 16 for ease of comparison. Figure 14 has also had the x-axis modified (now between 0 to 90 degrees) to better compare to the other measurements.

In Figure 16, the expected pattern of even and odd harmonics at 0 degrees and 90 degrees is seen for all the measurements. These harmonics agree with past measurements\(^9,22\).

However, as previously stated, there are extra features from 60 to 75 degrees in the ZnO angle dependence without polarization corrections (Figure 16a) and no signals at 45 degrees. Conversely, there are signals centered around 45 degrees in the ZnO angle dependence with Jones calculus corrections to the polarization (Figure 16b). This suggests that the large

![Figure 16: Comparison of orientation spectra from transmission through bulk ZnO, transmission through bulk ZnO with polarization corrections from Jones calculus, and a reflection geometry measurement of ZnO. All plots are with a logarithmic intensity scale.](image)
ellipticity change as the ZnO angle approaches 45 degrees (as shown in Figure 9) may suppress the signal. This is supported by past experiments, where the 11th harmonic has been shown to lose about half of the harmonic intensity when the ellipticity is at 0.4, and decreases further as the ellipticity increases\textsuperscript{42}. Therefore, the relatively large ellipticity around 45 degrees may be enough to stop the generation process. The extra features at 60 to 75 degrees may then be the edge of these harmonics at 45 degrees; able to appear once the ellipticity sufficiently reduces as the ZnO continues to increase. If these are indeed harmonics at 45 degrees, then there must also be some symmetry within the ZnO such that that both even and odd harmonics can be present.

As the reflection and transmission geometry with Jones calculus corrections to the polarization both functionally do not have propagation effects, they were expected to have similar results, and thus provide more certainty to these conclusions regarding the signals at 45 degrees. However, because the reflection geometry scan does not show angle dependence of the high order harmonics, we cannot directly compare the high order harmonics, and so must look at patterns in the low order harmonics with the current data. The signal that we see at 45 degrees in both the 5th harmonic in Figure 16c and the 7th harmonic in Figure 15b support the results seen in Jones calculus, and further suggest that harmonics are visible at 45 degrees when there is linear polarization.

Overall, these results suggest that the strong polarization change at 45 degrees from transmission through bulk ZnO is greatly interrupting the harmonic signal. The nature of the features described here is not yet understood. The angle dependence of ZnO is an active area of research at this time, as the theory currently is not able to completely
reproduce experimentally observed behavior\textsuperscript{42, 61}. More theoretical work, such as with DFT calculations, could be used to determine the origin of these signals at 45 degrees. Similarly, more theoretical work is needed to explain the lack of high-order harmonic angle dependence in a reflection geometry.
CHAPTER 7: TWO COLOR MEASUREMENTS

7.1. Overview

The two-color interferometer, as discussed in Section 2.3, uses the fundamental beam and second harmonic of the fundamental to produce uneven electric fields. In our setup (Figure 17), we used an AGS crystal, which has a relatively high efficiency for the second harmonic. The AGS crystal generates the second harmonic with a polarization perpendicular to the fundamental, therefore we used a waveplate in the fundamental arm to control the relative polarizations, as we had a waveplate suited for the fundamental wavelength on hand. The dichroic mirrors used to separate the beams had a measured reflectivity above 97% from 3000 nm to 3500 nm, making them a good choice for reflecting the fundamental beam away from the second harmonic while transmitting the second harmonic intensity. Because of these coatings, we used a wavelength of 3200 nm for these experiments, which produced a second harmonic at 1600 nm. We spatially aligned the two beams with a camera, then scanned the automatic translational stage to find the temporal overlap. Since we are working with a pulsed laser, we do not have the continuous sine function shown in Section 2.3, but rather must align the setup such that the short pulses overlap in time.

Figure 17: Two-color interferometer setup.
7.2. Results

As previously discussed, different cuts of a ZnO crystal provide different symmetry considerations to an HHG setup. An a-cut crystal can produce even and odd harmonics at different angles of the crystal, while a c-cut crystal will only generate even harmonics when there is asymmetry elsewhere in the system. When using an a-cut crystal, we found that we could see a lingering 4\textsuperscript{th} harmonic despite attempts to minimize even harmonics through rotation. Therefore, for this experiment, using a c-cut crystal eliminates the need to distinguish if even harmonics are from the crystal orientation and structure or from the asymmetric electric field. Figure 18 shows the difference between the harmonics at and away from the overlap position of the interferometer, or when there is introduced asymmetry and when there is not. The even harmonics are completely absent when the interferometer is not overlapped (harmonics 4 and 6 at wavelengths around 775 nm and 550 nm respectively). Also at the overlap, we see an increase in intensity of the odd harmonics, although the saturation is such that only the 7\textsuperscript{th} harmonic (wavelength around 450 nm) is clearly visible in Figure 18.

![Figure 18: Comparison of the harmonics generated at and away from overlap in a c-cut crystal.](image)
We then moved the state with a step size of 0.3 μm and were able to see the 4\textsuperscript{th} harmonic around 800 nm harmonic and a very faint 6\textsuperscript{th} harmonic around 550 nm (Figure 19) appear and disappear during the scan. The 4\textsuperscript{th} harmonic shows clear interference fringes, which are due to the stage movement and the periodic changing of the electric field, thus changing the symmetry of the system. When the two beams are not temporally overlapped, there is no harmonic signal, which we can see as a trailing of the signal on either side of the harmonic.

Figure 19 shows the harmonic measurement when the two arms of the interferometer have parallel polarizations and full intensity of the second harmonic. The intensity of the second harmonic can be weak; indeed, our second harmonic

![Figure 19: Harmonic measurement of a c-cut crystal as the stage moves. The 4\textsuperscript{th} and 6\textsuperscript{th} harmonic are visible with a linear intensity scale.](image-url)
measurement was too weak to be detected by our power meter. However, the intensity of the second harmonic can be varied to have different effects on the HHG signal.\textsuperscript{32, 33} We placed a rotatable polarizer in the second harmonic arm and measured the intensity of the generated harmonics during stage movement. As the polarizer rotated, both the angle of the polarization and the intensity changed. In Figure 20, we can clearly see the breakdown of signal as the polarizer rotates. However, these plots do not distinguish between the influence of relative polarization angles and influence of second harmonic intensity.

![Figure 20: Color plots showing the harmonic signal decrease as the polarizer rotates the second harmonic polarization while also decreasing the signal. Plotted in a logarithmic intensity scale.](image)

To better distinguish the source of the signal change, we integrated over the 4th harmonic for different experimental conditions. Two measurements were taken with different starting orientations of the polarizer. The angles listed in Figure 21 refer to the angle between the polarizations of each arm. In the “strong” signals in Figure 21, the fundamental and second harmonic arms began with parallel polarization. Then as the polarizer rotated, the intensity of the second harmonic decreased as the relative angle of
polarization between the two arms increased. In the “weak” signals, the two arms began with perpendicular polarization. Then as the polarizer rotated, the relative angle between the polarizations decreased while the intensity also decreased. In this way, we were able to measure two different second harmonic intensities for each relative polarization angle.

The “strong” 90 degrees and “weak” 0 degrees were taken when the polarizer was perpendicular to the second harmonic polarization, so none of the second harmonic signal should pass, as we see in Figure 21. We can still see some oscillation in these signals, however, this is minimal and can be attributed to the polarizer not blocking all second harmonic arm signal. While the 45 degrees harmonic signal of both intensities should be the same, they do not have quite the same harmonic yield and there is a peak shift between the two. However, there is still signal for both measurements, which is what would expect.

Figure 21: Integration of the 4th harmonic signal as the polarizer rotates the polarization and decrease the intensity of the second harmonic arm.
The “weak” 90 degrees signal occurs when there is no intensity loss from the polarizer, but the arms have perpendicular polarizations. This is an important finding. Rather than even harmonics strictly generated when there is an uneven electric field produced from interfering two-color fields, the even harmonics can be generated with two-color fields that are perpendicular. Even harmonics therefore arise not necessarily from an ionization asymmetry, but an asymmetry during the electron travel. The electron’s travel outside of the parent ion, commonly called the electron trajectory, is affected by the two-color fields, where the fundamental beam ionizes the electron and the second harmonic is used to modify the electron trajectory. This type of measurement has been used to distinguish and control the electron trajectory in gas-phase HHG measurements\textsuperscript{31, 32}. While gas-phase HHG can exhibit a short and long trajectory for the electrons, long trajectories have never been observed in solid-state HHG. The two-color interferometer built here therefore shows promising results for future investigation into the trajectories in solid-state HHG.
CHAPTER 8: CONCLUSION

In conclusion, solid-state HHG is a relatively new field that has the potential to be used in compact attosecond light sources and as a technique to study materials. However, understanding the generation mechanisms and the factors influencing the harmonic signal is important for many of these applications. We have looked at the influence of polarization, symmetry, and setup geometry on HHG in bulk crystal.

As bulk crystals in transmission are favorable in many situations, the Jones calculus technique presented has potential to help control the propagation effects from these thick crystals. We found that the harmonic signal seemed to be very polarization dependent, where the signals were seemingly entirely suppressed by a high ellipticity at 45 degrees. In support of this, a reflection geometry setup also produced an increase in signal at 45 degrees, although the direct comparison was not available with the limitations of the data. Future measurements of the reflection geometry will study the signals around 45 degrees and also work to understand the lack of angle dependence in the higher order harmonics. Overall, the comparison between these three setups suggests that a theoretical understanding is needed. One such method would be DFT calculations to help better understand the generation mechanism and influence of polarization.

In a different perspective on controlling HHG signal and the influences of polarization, we used a two-color interferometer to manipulate the electric field symmetry used to generate harmonics. We saw strong fringes at the overlap position, but also were able to observe a dependence on both the intensity of the second harmonic arm and the
angle between the polarizations of the two arms. This relationship has the potential in studying the long trajectories of the electrons, something that is currently unobserved about the generation in solid-state HHG. Future work involves investigation of these trajectories using the setup described in this thesis.

Overall, characterizing the influence of polarization, symmetry, and setup geometry on high harmonic generation in solids contributes to our understanding of the underlying mechanisms of HHG and improves our ability to control harmonic signal. This can lead to improved understanding of atomic processes and high powered light sources.
CHAPTER 9: REFERENCES