ELECTRIC FIELD SENSING THROUGH
SECOND HARMONIC GENERATION
OF ULTRAFAST LASER PULSES

by
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ABSTRACT

In dynamic processes such as spark discharges, it is impossible to insert field probes to measure rapidly varying and localized electric fields. In this thesis, we investigate the use of second harmonic generation of an ultrafast laser pulse as a probe for an electric field. In a centrosymmetric medium, second harmonic generation is not allowed in the electric dipole approximation. However, breaking the symmetry of air with an applied electric field allows for the second harmonic generation to occur. The second harmonic signal holds information about the applied electric field and can be used to provide information on the vector structure of the electric field. Here we show that second harmonic generation can be used as a noninvasive field probe for the vector electric field applied over an air gap.

We find that there is a significant background second harmonic signal without an applied field that results from a quadrupole nonlinear response of the air. When characterizing this background we find that the polarization purity is crucial to minimizing the quadrupole signal. Subtracting this background, taking two polarization directions, and scanning the beam spatially around a blade electrode, we show that we can make a field map of the electric field that is consistent with an electrostatic calculation.

Analyzing the results from this work, we propose two methods for using this probe. The first is to use a pulse sequence and observe the second harmonics with a photomultiplier tube to see the voltage response over time. The pulse sequence will capture the electric field at each pulse and show the variations in the field over time. The second method is to use a beam that is simultaneously focused in space and in time. This will give a shorter depth of focus and should minimize the background second harmonic signal. The imaging is a possible probe for fields changing on the order of seconds. The efficiencies that need to be reached to image single shots are currently not available.
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LIST OF SYMBOLS

Charge density ............................................ $\rho$

Current density ........................................... $J$

Displacement Field ......................................... $D$

Electric field .............................................. $E$

Magnetic field ............................................ $B$ or $H$

Magnetization ............................................... $M$

Phase Velocity ............................................ $\nu$

Polarization vector ....................................... $P$

Refractive Index .......................................... $n$

Speed of Light ............................................ $c$

Susceptibility ............................................. $\chi$

Vacuum Permeability ..................................... $\mu_0$

Vacuum Permittivity ..................................... $\epsilon_0$

Wave Number ............................................. $k$
LIST OF ABBREVIATIONS

Background Second-Harmonic .............................................. BSH
Charge Coupled Device ...................................................... CCD
Chirped-Pulse Amplification ................................................. CPA
Colorado School of Mines ..................................................... CSM
Direct Current ...................................................................... DC
Los Alamos National Lab ....................................................... LANL
Photomultiplier Tube ............................................................ PMT
Second Harmonic .................................................................. SH
Second-Harmonic Generation ................................................. SHG
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CHAPTER 1
INTRODUCTION

The goal of this thesis is to explore the feasibility of a non-invasive field probe of the electric field vector through second harmonic generation (SHG) of femtosecond pulses.

Accidental electrostatic discharge can pose a serious safety risk for many industrial operations related to the Los Alamos National Labs (LANL) mission. Our group at the Colorado School of Mines (CSM) has been investigating electrostatic discharge to understand how much stored energy on a charged object can be delivered to a ‘victim’ load through the spark. To better understand the electrostatic discharge events, we have developed models of the plasma dynamics of the sparks. We are looking for laser diagnostic probes that can be used to characterize the dynamic conditions in the spark plasmas. Among the parameters of interest is the dependence of the electric field on position and time within the spark. Since it is possible to use a laser pulse to trigger the spark, we are looking for a method that will allow us to obtain as much information about the electric field as possible in a single shot.

Previous methods of measuring electric fields for plasmas have measured the electric field dynamics in plasmas using the fluorescence dip technique, with fluorescence from excited states of hydrogen [1] and Rydberg-state helium [2]. The fluorescence dip technique finds the electric field through the use of Stark splitting. With Stark splitting the excited-state transitions are observed. This method of finding the electric field had been shown to be precise, up to a few volts per centimeter, but has to be done at low pressures where the Stark effect is not masked by collisional (pressure) broadening. Additionally, for this technique, the laser must have a narrow-linewidth that is tuned to the resonant transitions of the species observed.

Gavrilenko et al. [3] and Goldberg et al. [4] showed measurement of electric fields through resonant four-wave mixing. The mixing process is similar to Coherent Anti-Stokes
Raman Scattering (CARS) and is non-intrusive. There are two driving beams with operating frequencies that differ by the vibrational Raman shift of a gas molecule. This approach has been used to measure the electric fields in hydrogen [5] and nitrogen [6]. The output beam for the hydrogen was measured at 2.4 µm and for the nitrogen at 4.29 µm, which were used to measure the electric fields. Although CARS is non-intrusive it does require different driving frequencies for two lasers, and the detection of different wavelength light, which is dependent on the molecular structure of the gas. As CARS is dependent on resonance, it does not work for all gases and has only been shown to be effective for H$_2$ and N$_2$.

Dominic and Feinberg have presented a method of imaging the electric field in bulk glass through the use of SHG [7]. The direct current (DC) field map is induced by the rearrangement of charges in the glass as the glass gets irradiated by two beams. The two beams are at the fundamental, $\omega$, and second harmonic, $2\omega$, frequencies of the pump laser. To map out the electric field, the SH signal is observed through two different orientations of the polarizer and moving the beam through a $x$-$y$ grid. This method requires two input signals: the fundamental and second harmonic.

Dogariu et al. shows a non-intrusive measurement technique of the electric field strength using SHG [8]. The experiments performed take the electric field strength up to 2000 V/cm. The paper shows that there is a quadratic response in the signal as the applied voltage is increased and the electric field strength varies from 0 V/cm to 2000 V/cm. The pulse energy was kept around 0.5 mJ per pulse, with a pulse width of 50 fs and a central wavelength of 800 nm. The SH signal was observed using a photomultiplier tube (PMT) and an oscilloscope. Dogariu et al. were able to map out time dependence of electric fields. The method of measuring electric field vector dependence is also non-invasive. There is only one input beam required and has no limitations on the atomic structure of the gas.

Our approach to a non-invasive probe is an extension of the Dogariu method. Similarly to Dogariu et al., for our experiments, a femtosecond laser is used with a central wavelength at 800 nm. The electric field strength is increased, close to the breakdown point of air, 30
kV/cm [9]. Instead of only using a PMT to observe and characterize the SH behavior, a camera will be used to image the SH and from that retrieve the local electric field structure.
This chapter provides the background required for the project. The process of second harmonic generation in second and third order susceptibility will be described. The theoretical form of the static electric fields will also be touched on.

2.1 Second Harmonic Generation

The nonlinear optical response of materials can be used to extend the available frequency range of a laser. One of these nonlinear phenomena is second-harmonic generation (SHG) which will be discussed in this section. As will be shown below, in the dipole approximation, SHG requires that the medium have an inversion asymmetry. SHG has been measured in anisotropic crystals, solid surfaces, and liquids or gases that have been oriented, for example with an external field [10]. The strength of the second harmonic (SH) signal depends on the symmetry of the medium, the phase matching between the fundamental and doubled frequencies, and the intensity of the laser beam within the nonlinear medium.

SHG can occur when a strong DC or quasi-DC field is applied to an isotropic gas, as the applied field breaks the symmetry of the gas. Therefore, we anticipate that the SH signal can be used to image an applied electric field at the focus of the fundamental laser. The SH can, therefore, be used to image the electric field and find the underlying vector structure.

2.1.1 Electromagnetic Theory

The mathematical framework for understanding second harmonic generation (SHG) is found in electromagnetism. In the following derivations, we largely follow the analysis and notation found in Boyd [10]. Recall Maxwell’s equations in SI units:

\[ \nabla \cdot \mathbf{D} = \rho \]  

(2.1)
\[ \nabla \cdot \mathbf{B} = 0 \quad (2.2) \]

\[ \nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \quad (2.3) \]

\[ \nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} + \mathbf{J} \quad (2.4) \]

In Maxwell’s equations \( \mathbf{E} \) is the electric field, \( \mathbf{D} \) is the displacement field, \( \mathbf{B} \) is the magnetic field, \( \mathbf{H} = \frac{1}{\mu_0} \mathbf{B} - \mathbf{M} \), \( \mathbf{M} \) is the magnetization and \( \mu_0 \) the permeability of vacuum, \( \mathbf{J} \) is the free current and \( \rho \) is the free charge density. The region of space of interest is assumed to carry no free charge, no current and is nonmagnetic. These assumptions simplify Maxwell’s equations.

\[ \rho = 0, \quad \mathbf{J} = 0, \quad \mathbf{B} = \mu_0 \mathbf{H} \quad (2.5) \]

\( \mathbf{D} \) and \( \mathbf{E} \) are related through polarization.

\[ \mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P} \quad (2.6) \]

Above \( \mathbf{P} \) is the polarization vector, and \( \epsilon_0 \) is the permittivity of vacuum. The material that the light propagates through is allowed to be nonlinear so that the polarization vector has a nonlinear dependence on the local electric field strength within the material. Taking the perturbation expansion of \( \mathbf{P} \) with \( \mathbf{E} \),

\[ \mathbf{P} = \epsilon_0 \left( \chi^{(1)} \mathbf{E} + \chi^{(2)} \mathbf{E}^2 + \chi^{(3)} \mathbf{E}^3 + \cdots \right) \quad (2.7) \]

we see the nonlinear dependence of \( \mathbf{P} \) in the terms with higher order dependence on \( \mathbf{E} \).

We transform Maxwell’s equations into a differential wave equation by taking the curl of both sides of (2.3). We use vector calculus and the assumptions stated in (2.5) to simplify the equation to the relation between \( \mathbf{E} \) and \( \mathbf{P} \) as

\[ \nabla \times \nabla \times \mathbf{E} + \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2} \quad (2.8) \]

Eq. (2.8) is the most general wave equation form in nonlinear optics, and can be simplified with further assumptions. The first term on the left hand side can be transformed using
vector calculus as

$$\nabla \times \nabla \times \mathbf{E} = \nabla (\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} \quad (2.9)$$

If there is no spatial dependence of the dielectric response, then $\nabla \cdot \mathbf{E} = 0$, i.e. $\nabla \epsilon = 0$. In nonlinear optics, the more generalized relation between $\mathbf{D}$ and $\mathbf{E}$ leads to a non-vanishing term, even in isotropic materials. In most nonlinear cases the first term on the right hand side of (2.9) can be shown to be small, especially with the slowly varying amplitude approximation. $\nabla \cdot \mathbf{E}$ is assumed to be small enough to neglect and the wave equation becomes

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2} \quad (2.10)$$

To see the nonlinearity come into (2.10), we take the polarization and displacement fields and separate them into their linear and nonlinear components

$$\mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{NL} \quad (2.11)$$

$$\mathbf{D} = \mathbf{D}^{(1)} + \mathbf{D}^{NL} \quad (2.12)$$

where the superscript denotes whether the vectors are linear, (1), or nonlinear, (NL). The linear component of the displacement field, $\mathbf{D}^{(1)}$ can be represented by

$$\mathbf{D}^{(1)} = \epsilon_0 \mathbf{E} + \mathbf{P}^{(1)} \quad (2.13)$$

After solving (2.13) for $\mathbf{E}$, we substitute that in for the second $\mathbf{E}$ in (2.10) along with (2.11) for $\mathbf{P}$,

$$\nabla^2 \mathbf{E} - \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{D}^{(1)}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}^{NL}}{\partial t^2} \quad (2.14)$$

The medium in which the nonlinear processes take place is now assumed to be isotropic, dispersion-less, and lossless. The assumptions on the medium allow $\mathbf{D}^{(1)}$ to be represented in terms of the electric field and a frequency-independent dielectric scalar, $\epsilon^{(1)}$

$$\mathbf{D} = \epsilon_0 \epsilon^{(1)} \mathbf{E} \quad (2.15)$$
With the assumption that the driven wave equation is satisfied by every frequency component of $\varepsilon^{(1)}$, (2.14) can be written as

$$-\nabla^2 E_j + \frac{\varepsilon^{(1)}(\omega_j)}{c^2} \frac{\partial^2 E_j}{\partial t^2} = -\frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P_{NL}^j}{\partial t^2}$$  \hspace{1cm} (2.16)

With the relation in (2.16) we can analyze the process of SHG. The most general process of SHG is shown in Figure 2.1. Two photons of the same frequency ($\omega$) add and a third photon at twice the frequency ($2\omega$) is released.

Figure 2.1: Diagram depicting second harmonic generation. Two photons at $\omega$ come in and one photon at $2\omega$ comes out. The dashed lines are virtual states.

The total electric field in the nonlinear medium is the sum of the fundamental and doubled frequency components.

$$E(z,t) = E_1(z,t) + E_2(z,t)$$  \hspace{1cm} (2.17)

where the 1 and 2 subscript represent the fundamental and doubled frequency components respectively, and for simplicity, we assume each field is a plane wave propagating in the $z$ direction. Each of the electric fields can be expressed as a complex amplitude $\tilde{E}$ and slowly varying amplitude $A_j(z)$,

$$E_j(z,t) = \tilde{E}_j(z) e^{-i\omega_j t} + \text{c.c.}$$  \hspace{1cm} (2.18)

and

$$\tilde{E}_j(z) = A_j(z) e^{ik_j z}$$  \hspace{1cm} (2.19)
In (2.18) the c.c. denotes that the complex conjugate of the first term is added. The index $j$ can be 1 or 2, representing the fundamental and doubled frequency components respectively. The wave number, $k_j$, is given in terms of the refractive index, $n_j$ as

$$k_j = \frac{n_j \omega_j}{c}, \quad n_j = \sqrt{\epsilon^{(1)}(\omega_j)}$$

(2.20)

We can also represent the nonlinear polarization in its fundamental and doubled frequency components just as the electric field.

$$P_{NL}(z, t) = P_j(z) e^{-i \omega_j t} + \text{c.c.}$$

(2.21)

where $j = 1, 2$. The lowest order nonlinear case of SHG has $P_{NL}^{NL}(\omega)$ as

$$P^{(2)}_i(2\omega) = \sum_{jk} \chi^{(2)}_{ijk} (2\omega, \omega) E_j(\omega) E_k(\omega)$$

(2.22)

where $i, j, k$ represent the Cartesian coordinates $x, y, z$ respectively. $\chi^{(2)}_{ijk}$ is the susceptibility tensor. The susceptibility represents the induced response in a material due to the presence of an electric field. Through (2.22), the intensity of the electric field in a medium, $E$, and the materials response, $\chi^{(2)}_{ijk}$, can be used to determine the nonlinear polarization. Given a fixed propagation and polarization direction, the susceptibility tensor can be represented by the scalar $d_{eff}$. The two polarization components can be represented as

$$P_1(z) = 4d_{eff}E_2E_1^* = 4d_{eff}A_2 A_1^* e^{i(k_2-k_1)z}$$

(2.23)

$$P_2(z) = 2d_{eff}E_1^2 = 2d_{eff}A_1^2 e^{2ik_1z}$$

(2.24)

Taking these forms of the polarization, substitute (2.17) and (2.21) into (2.16).

$$-\nabla^2 (A_1(z)e^{ik_1z} e^{-i\omega t}) + \frac{\epsilon^{(1)}(\omega_1)}{c^2} \frac{\partial^2}{\partial t^2} (A_1(z)e^{ik_1z} e^{-i\omega_1 t}) = -\frac{1}{\epsilon_o c^2} \frac{\partial^2 4d_{eff}A_2 A_1^* e^{i(k_2-k_1)z} e^{-i\omega_1 t}}{\partial t^2}$$

(2.25)

Applying the spatial and temporal derivatives to the terms on the left hand side, this becomes
\[- \left( \frac{\partial^2}{\partial z^2} A_1(z) + 2 i k_1 \frac{\partial}{\partial z} A_1(z) - k_1^2 A_1(z) + \frac{\omega_1^2 \epsilon^{(1)}(\omega_1)}{c^2} A_1(z) \right) e^{i k_1 z e^{-i \omega_1 t}} \quad (2.26)\]

the first three terms come from the two spatial derivatives and the fourth term comes from
the time derivatives. The last two term cancel each other out. The new form of the applied
wave equation becomes,

\[- \left( \frac{\partial^2}{\partial z^2} A_1(z) + 2 i k_1 \frac{\partial}{\partial z} A_1(z) \right) e^{i k_1 z e^{-i \omega_1 t}} = \frac{4 \omega_1^2 d_{eff}}{c^2} A_2 A_1^* e^{i(k_2-k_1)z} e^{-i \omega_1 t} \quad (2.27)\]

\[- \left( \frac{\partial^2}{\partial z^2} A_1(z) + 2 i k_1 \frac{\partial}{\partial z} A_1(z) \right) = \frac{4 \omega_1^2 d_{eff}}{c^2} A_2 A_1^* e^{i(k_2-2k_1)z} \quad (2.28)\]

In 2.28 the first term of the left hand side is assumed to be much smaller than the
second. This approximation is called the slowly-varying wave approximation, stating that
the amplitude of \( A_1 \) varies much slower that the spatial oscillations in \( e^{ikz} \). The double
spatial derivative is negligible compared to the other terms. With this approximation we
simplify the differential equation to

\[\frac{d A_1}{dz} = \frac{2 i \omega_1^2 d_{eff}}{k_1 c^2} A_2 A_1^* e^{-i \Delta k z} \quad (2.29)\]

We take the same approach to solve for the second differential equation, also using the
slowly-varying wave approximation to simplify the equation.

\[\frac{d A_2}{dz} = \frac{i \omega_2^2 d_{eff}}{k_2 c^2} A_1^2 e^{i \Delta k z} \quad (2.30)\]

where the phase mismatch, \( \Delta k = 2k_1 - k_2 \), represents how closely the fundamental
and doubled frequency wave vectors overlap.

The solutions to these coupled differential equations describe the wave build up of the
SH signal, \( A_2 \), and the fundamental, \( A_1 \). In our case, where the conversion efficiency is
extremely low, we can safely assume that the power in the fundamental beam is not depleted
significantly. Therefore, we can work with just (2.30) and hold \( A_1 \) constant.
2.1.2 Phase-Matching Condition

From (2.20) the wave number for the fundamental and doubled frequency are

\[ k_\omega = \frac{\omega n_\omega}{c}, \quad k_{2\omega} = \frac{2\omega n_{2\omega}}{c} \]  

(2.31)

The electric field at the fundamental frequency locally produces the second harmonic field in phase with the fundamental. This SH field coherently adds to the SH signal produced at other \( z \) positions in the medium. For maximum efficiency, the phase velocity of the SH signal must be equal to that of the fundamental electric field.

\[ v_p = \frac{2\omega}{2k_\omega}, \quad v_E = \frac{2\omega}{k_{2\omega}} \]  

(2.32)

\[ k_{2\omega} = 2k_\omega \]  

(2.33)

where the \( v_p \) is the phase velocity of the fundamental (pump) beam, and \( v_E \) is the phase velocity of the SH signal.

Perfect phase matching condition is shown in (2.33), which sets \( \Delta k = 0 \). With a perfect phase matching the SHG process is most efficient, Figure 2.2a. The refractive index, \( n_\omega \), of most materials in the visible part of the spectrum increases with frequency, leading to negative phase matching conditions. Using a birefringent nonlinear crystal the phase matching can be angle-tuned to be positive or negative by placing the harmonic beam along the lower refractive index axis. In Figure 2.2b the case of positive phase mismatch is depicted. With a positive \( \Delta k \), SHG is still allowed because a Gaussian beam has an inherent angular spread of wave numbers.

![Figure 2.2: Phase matching shown under (a) perfect phase matching condition, and (b) positive phase matching condition.](image)
The conversion efficiency from the fundamental to the doubled frequency is strongly dependent on the wave number mismatch. To see this dependence, start by looking at the intensity of each wave. The intensity is given by the time average pointing vector,

\[ I_j = \frac{n_j c}{2\pi} |A_j|^2 \]  (2.34)

Integrating (2.30) over \( z \) from \( z_o \) to \( z' \),

\[ A_2 = \frac{4\pi i \omega_2^2 d_{eff}}{k_2 c^2} A_1^2 \left( \frac{e^{i\Delta k z'} - e^{i\Delta k z_o}}{i\Delta k} \right) \]  (2.35)

The amplitude, \( A_2 \), is now in terms of the axial position. The same can be done for \( A_1 \), taking the form for \( A_1 \) and \( A_2 \) put them in (2.34), and use (2.20) to simplify, the doubled frequency intensity as a function of axial position becomes

\[ I_2 = \frac{32\pi^3 \omega_2^2 d_{eff}^2}{n_1^2 n_2 c} I_1^2 L^2 \text{sinc}^2 \left( \frac{\Delta k L}{2} \right), \quad L = z' - z_o \]  (2.36)

In (2.36) \( L \) is the length of the nonlinear medium. The intensity of the second harmonic depends on the square of the fundamental intensity, showing the efficiency of SHG dependence on strong input intensity. The phase matching parameter is an argument of the sinc function. Poor phase matching can easily reduce the conversion efficiency of the fundamental to SH signal.

One approach to solving the more general case of finite beam size is to expand the field and the nonlinear polarization in terms of modes that solve the homogeneous wave equation, i.e. the left hand side of (2.16). The simple case of harmonic generation of Gaussian beams is worked out in Boyd [10]. For SHG, the maximum conversion efficiency in the tight focusing limit is at \( \Delta k = 0 \). The tight focusing limit denotes propagation for many Rayleigh ranges on both sides of the focus. The conversion efficiency is nonzero for \( \Delta k > 0 \), but peaks at 0.

2.1.3 Negative Phase Matching

As stated above the phase matching is usually controlled with a birefringent crystal. For the purpose of this thesis, there was no control over the phase mismatch. The index of refraction for air goes as, [11]

---

11
\[ n(\lambda) = 1 + \frac{0.05792105}{238.0185 - \lambda^{-2}} + \frac{0.00167917}{57.362 - \lambda^{-2}} \]  

(2.37)

Using the refractive index for air the \( \Delta k \) can be calculated for the lab. In the equation above \( \lambda \) is the wavelength in microns. Taking the fundamental and doubled frequency wavelengths as 800 and 400 nm,

\[ \Delta k = -121.2 \text{ m}^{-1} \]  

(2.38)

The coherence length of the phase mismatch is

\[ L = \frac{2\pi}{\Delta k} = 51.9 \text{ mm} \]  

(2.39)

The coherence length denotes the distance traveled for one period of the oscillation for the wave vector of the fundamental and doubled frequency to aligned again. As we will be discussing later, in our experiment, the coherence length is much smaller than the lens focal length, but comparable to or larger than the electric field structures we are measuring.

### 2.1.4 Inversion Symmetry

As stated before, for SHG to be possible the medium must have a particular symmetry, there must be high intensity, and adequate phase matching. The last two points have been shown mathematically above. The first requirement states that there must be a lack of inversion symmetry. The symmetry of a medium can be seen in the susceptibility tensor. Taking the simple form of the polarization, assuming that the material’s polarization instantaneously responds to an applied electric field,

\[ \mathbf{P}(t) = \chi^{(2)} \mathbf{E}^2(t) \]  

(2.40)

With a lack of inversion symmetry, when the sign on the electric field is changed, the sign on the polarization must also change

\[ -\mathbf{P}(t) = \chi^{(2)}(-\mathbf{E})^2(t) \]  

(2.41)

If the nonlinear medium holds inversion symmetry, the susceptibility tensor does not change sign. From (2.40) and (2.41), \( \mathbf{P}(t) = -\mathbf{P}(t) \), which can only be true if \( \chi^{(2)} \) is zero. This
condition results in the conclusion that no SHG is expected to take place in a centrosymmetric nonlinear materials. However, we will see in the next section that anything that breaks this symmetry will allow SHG to take place.

2.2 Quadrupole Expansion in the Electric Field

The case of SHG discussed above is the easiest and simplest SHG process. The medium used in the experiment is air. Air is considered a centrosymmetric medium which means it holds inversion symmetry. Above the conclusion was drawn that there can be no SHG in a centrosymmetric medium, but with the laser system used there is a nonzero second harmonic signal without externally applying any symmetry breaking.

The analysis above, and most derivations in literature, expand the fundamental electric field out to the dipole term, and ignores higher order terms. Taking the electric field expansion out to the quadrupole term suddenly allows for SHG in a centrosymmetric medium. These higher order SH terms tend to be weak enough that they can be ignored, but the laser system used has high enough intensities to create a measurable amount of SH signal through the quadrupole term.

If we allow for a pair of fields \( E_1 \) and \( E_2 \) that can interact in a medium, the polarization given by the quadrupole interaction between the fields and their field gradients is given by

\[
P_Q(\omega_1 + \omega_2) = \chi_Q(\omega_1, \omega_2) \left[ -\frac{2}{3} \left( \nabla \cdot E_1 \right) E_2 + \left( \nabla E_1 \right) \cdot E_2 + E_2 \cdot \left( \nabla E_1 \right) \right]
+ \chi_Q(\omega_2, \omega_1) \left[ -\frac{2}{3} \left( \nabla \cdot E_2 \right) E_1 + \left( \nabla E_2 \right) \cdot E_1 + E_1 \cdot \left( \nabla E_2 \right) \right]
\]

(2.42)

Compared to the polarization in (2.40) this form of the polarization, \( P_Q \), does hold inversion asymmetry [12]. Bethune shows similar structure for the polarization due to the magnetic dipole interaction with the magnetic field, and the nonuniform field-induced quadrupole moment per volume. All three of these polarization’s have similar electric field dependence and can be combined for a more complete polarization.
As we are only looking at the electric field terms we stick with the form in (2.42). The spatial part of the electric field in (2.42) are all Gaussian beams. As every terms holds a spatial derivative of the field, each term of $P_Q$ goes as $x e^{-x^2}$. When applying coordinate inversion, the extra $x$ will flip the sign of $P_Q$. Now we can see that second harmonic generation can take place even in a medium with inversion symmetry. Effectively, it is the gradient in the intensity that breaks the symmetry.

Taking the general form of the polarization due to the quadrupole term, (2.42), we apply the conditions for SHG. For SHG, $\omega_1 = \omega_2 = \omega$ and $E_1 = E_2 = E$,

$$P(2\omega) = \chi_Q(\omega, \omega) \left[ -\frac{2}{3} (\nabla \cdot E)E + \frac{1}{2} \nabla E^2 + (E \cdot \nabla)E \right]$$

The polarization in (2.43) is due to a dipole-forbidden second-order susceptibility [12]. The background SH signal that is observed is due to the polarization above. From before, $\nabla \cdot E = 0$ if the medium is uniform and has no free charge, eliminating the first term. $\nabla E$ will not lead to a propagating electromagnetic wave since it has zero curl, so the third term in (2.43) will dominate [12].

2.3 Third-Order Susceptibility Process

The analysis above considered SHG occurring only for second-order susceptibility processes. These required either a medium that does not have inversion symmetry, or a quadrupole expansion of the electric field. The SH signal that will be used to map out the electric field strengths and vectors is a third order susceptibility process.

A third-order process yields a signal that depends on the third power of the input field(s). So for example we can get third-harmonic generation from the fundamental,

$$(e^{-i\omega t})^3 = e^{-i3\omega t}$$

If a DC field is present, there is another mechanism for generating the SH signal. With three wave mixing between the laser and the DC electric field:

$$\omega + \omega + 0 = 2\omega$$
This is the mechanism that allows us to use SHG as a probe of the DC electric field.

For the second-order susceptibility process the polarization obeyed the form in (2.22). An equivalent equation is found in the case of third-order susceptibility processes. A third-order process involves four photons, where the fourth photon is the output, $\omega_1 = \omega_1 + \omega_2 + \omega_3$. The most general formula of the polarization at $\omega_4$ is given by

$$P_i(\omega_4) = \frac{1}{4} \varepsilon_o \sum_p \sum_{ijkl} \chi^{(3)}_{ijkl}(\omega_4; \omega_1, \omega_2, \omega_3) E_j(\omega_1) E_k(\omega_2) E_l(\omega_3) \tag{2.46}$$

Here $ijkl$ can all be 1, 2, or 3, associated with $x$, $y$, and $z$ respectively. $\sum_p$ refers to the sum over all distinct permutations of $\omega_1$, $\omega_2$, and $\omega_3$ [13]. In the case of SHG through a third-order process, $\omega_1 = \omega_2 = \omega$ and $\omega_3 = 0$ such that $\omega_4 = \omega_1 + \omega_2 + \omega_3 = 2\omega$. Taking the sum over $p$ in (2.46) the polarization becomes

$$P_i(2\omega) = \frac{1}{4} \varepsilon_o \sum_{ijkl} \left( \chi^{(3)}_{ijkl}(2\omega; \omega, \omega, 0) \hat{E}_j(\omega) \hat{E}_k(\omega) \hat{E}_l(0) + \chi^{(3)}_{ijkl}(2\omega; \omega, 0, \omega) \hat{E}_j(\omega) \hat{E}_k(0) \hat{E}_l(\omega) + \chi^{(3)}_{ijkl}(2\omega; 0, \omega, \omega) \hat{E}_j(0) \hat{E}_k(\omega) \hat{E}_l(\omega) \right) \tag{2.47}$$

There are three terms left in the polarization sum, as there are three distinct permutations of $\omega$, $\omega$, 0.

In (2.46) and (2.47), $\chi^{(3)}_{ijkl}$ is the third-order susceptibility. $\chi^{(3)}_{ijkl}$ has 81 elements that describe a medium’s reaction to the presence of an electric field. In materials that lack symmetry, all 81 elements are independent of each other. Materials which obey symmetry rules can reduce the number of independent elements. The focus will be brought to isotropic materials such as gases, liquids, and vapors.

An isotropic material has no intrinsic axis and holds inversion symmetry, so each coordinate axes must be equivalent. In an isotropic material an applied electric field in the $x_j$ direction should not induce a field in the $\pm x_k$ direction. For every nonzero element of $\chi^{(3)}_{ijkl}$, the index numbers should appear an even number of times. The inversion symmetry leaves 21 out of the 81 susceptibility elements non zero [10, 13]. The 21 nonzero elements are not
all independent of one another, and the susceptibility elements can be related as

\[ \chi_{1111} = \chi_{2222} = \chi_{3333} \]
\[ \chi_{1122} = \chi_{1133} = \chi_{2211} = \chi_{2233} = \chi_{3311} = \chi_{3322} \]
\[ \chi_{1212} = \chi_{1313} = \chi_{2323} = \chi_{2121} = \chi_{3131} = \chi_{3232} \]
\[ \chi_{1221} = \chi_{1331} = \chi_{2112} = \chi_{2332} = \chi_{3113} = \chi_{3223} \] (2.48)

All nonzero elements of \( \chi^{(3)}_{ijkl} \) are shown in (2.48). The relations between the elements shows that there are only four distinct elements that must satisfy

\[ \chi_{1111} = \chi_{1122} + \chi_{1212} + \chi_{1221} \] (2.49)

leaving \( \chi^{(3)}_{ijkl} \) with 3 independent elements.

Expanding the sum over \( jkl \) in (2.47) for \( P_1(2\omega) \) gives 21 terms. The 21 terms can be combined into 7 terms by using intrinsic permutation symmetry. Intrinsic permutation symmetry states

\[ \chi^{(3)}_{ijk}(\omega_3; \omega_1, \omega_2) = \chi^{(3)}_{ikj}(\omega_3; \omega_2, \omega_1) \] (2.50)

The relation between the permutations in the susceptibility indices relate the 21 different \( \chi^{(3)}_{ijkl} \) to three independent values,

\[ \chi_{1111}(2\omega; \omega, \omega, 0) = \chi_{1111}(2\omega; \omega, 0, \omega) = \chi_{1111}(2\omega; 0, \omega, \omega) \]
\[ \chi_{1122}(2\omega; \omega, 0, \omega) = \chi_{1122}(2\omega; \omega, 0, \omega) = \chi_{1212}(2\omega; \omega, \omega, 0) = \chi_{1212}(2\omega; 0, \omega, \omega) \]
\[ = \chi_{1221}(2\omega; \omega, 0, \omega) = \chi_{1221}(2\omega; 0, \omega, \omega) = \chi_{1133}(2\omega; \omega, \omega, 0) \]
\[ = \chi_{1133}(2\omega; \omega, 0, \omega) = \chi_{1313}(2\omega; \omega, \omega, 0) = \chi_{1313}(2\omega; 0, \omega, \omega) \]
\[ = \chi_{1331}(2\omega; \omega, 0, \omega) = \chi_{1331}(2\omega; 0, \omega, \omega) \]
\[ \chi_{1112}(2\omega; 0, \omega, \omega) = \chi_{1122}(2\omega; \omega, 0, \omega) = \chi_{1212}(2\omega; \omega, \omega, 0) = \chi_{1133}(2\omega; 0, \omega, \omega) \]
\[ = \chi_{1313}(2\omega; \omega, 0, \omega) = \chi_{1331}(2\omega; \omega, 0, \omega) \] (2.51)

The 21 terms that were initially produced in the \( \sum_{jkl} \) in (2.47) are now condensed down to
\[ P_1(2\omega) = \frac{1}{4} \varepsilon_o \left[ 3\chi_{1111}(2\omega; \omega, \omega, 0)E_1(\omega)E_1(\omega) + 6\chi_{1122}(2\omega; \omega, \omega, 0)(E_1(\omega)E_2(\omega)E_2 + E_1(\omega)E_3(\omega)E_3) \right. \\
\hspace*{3cm} \left. + E_1(\omega)E_3(\omega)E_3 \right] + 3\chi_{1221}(2\omega; \omega, \omega, 0) \left( E_1E_1(\omega)E_1(\omega) + E_1E_2(\omega)E_2(\omega) + E_1E_3(\omega)E_3(\omega) \right) \]  

(2.52)

Further simplifications can be made by using (2.49). Applying the intrinsic permutation symmetry to (2.49), \( \chi^{(3)}_{1111} \) can be turned into two terms.

\[ \chi_{1111}(2\omega; \omega, \omega, 0) = \chi_{1122}(2\omega; \omega, \omega, 0) + \chi_{1212}(2\omega; \omega, \omega, 0) + \chi_{1221}(2\omega; \omega, \omega, 0) \]

\[ = 2\chi_{1122}(2\omega; \omega, \omega, 0) + \chi_{1221}(2\omega; \omega, \omega, 0) \]  

(2.53)

The polarization for the second harmonic along one axis becomes,

\[ P_1(2\omega) = \frac{1}{4} \varepsilon_o \left[ 6\chi_{1122}(2\omega; \omega, \omega, 0)(E_1(\omega)E_1(\omega) + E_1(\omega)E_2(\omega)E_2 + E_1(\omega)E_3(\omega)E_3) \right. \\
\hspace*{3cm} \left. + 3\chi_{1221}(2\omega; \omega, \omega, 0) \left( E_1E_1(\omega)E_1(\omega) + E_1E_2(\omega)E_2(\omega) + E_1E_3(\omega)E_3(\omega) \right) \right] \]

Taking a closer look at the two grouped terms separately shows that there is a dot product within each of the terms.

\[ P_1(2\omega) = \frac{1}{4} \varepsilon_o \left[ 6\chi_{1122}(2\omega; \omega, \omega, 0)(E_1(\omega)E_1(\omega) + E_1(\omega)E_2(\omega)E_2 + E_1(\omega)E_3(\omega)E_3) \right. \\
\hspace*{3cm} \left. + 3\chi_{1221}(2\omega; \omega, \omega, 0) \left( E_1E_1(\omega)E_1(\omega) + E_1E_2(\omega)E_2(\omega) + E_1E_3(\omega)E_3(\omega) \right) \right] \]

(2.54)

A similar process can be taken to find the form for \( P_2(2\omega) \) and \( P_3(2\omega) \). Combining the three terms to form a collective \( \mathbf{P}(2\omega) \) gives

\[ \mathbf{P}(2\omega) = \frac{1}{4} \varepsilon_o \left[ 6\chi_{1122}(2\omega; \omega, \omega, 0)(\mathbf{E}(\omega) \cdot \mathbf{E}_{DC})\mathbf{E}(\omega) + 3\chi_{1221}(2\omega; \omega, \omega, 0) \left( \mathbf{E}(\omega) \cdot \mathbf{E}(\omega) \right) \mathbf{E}_{DC} \right] \]

(2.55)

The SH signal that will lead to finding the vector field of the electric field will be of the form above. There is a term that will be mainly dependent on the vector structure of the fundamental, and another that is dependent on the vector structure of the applied electric field.
By controlling the orientation of the input laser polarization relative to the DC electric field, we can map out the vector direction of the electric field.

It is also worth noting that, as shown in Bethune [12], quasi-DC fields can be set up in the medium through the creation of a plasma with charge separation. A quasi-DC field can also produced in a neutral gas, a bound-charge polarization is set up in the gas by the gradient in the laser intensity. These are potential electric field sources that could be imaged.
CHAPTER 3
EXPERIMENTAL METHOD

This chapter discusses the laser system, experimental setup, and data collection methods used. The laser system consists of an oscillator, a stretcher, an amplifier and a compressor. In the following section, we will describe the laser system, the delivery optics and the experimental apparatus to measure electric field strengths in air using the second harmonic signal.

3.1 Optical System

The laser system used to produce the pulses for experiment is an in-house laser system. A block diagram of the laser system is shown in Figure 3.1. The mode-locked Ti:sapphire oscillator produces pulses that carry energy on the order of nanojoules per pulse. The pulses from the oscillator cannot be focused to intensities sufficiently high to produce measurable SHG in air. An amplification stage takes the pulses from the oscillator and amplifies the peak power of the pulses.

Figure 3.1: Block diagram of Ti:Sapphire laser. With four main stages: (1) oscillator, (2) stretcher, (3) amplifier, and (4) compressor.

The amplification stage is based on chirped-pulse amplification (CPA). CPA allows for short pulses to build up their peak intensities by stretching, amplifying and re-compressing...
the pulse [14–17]. Further discussion below gives more detail on each individual block of the in-house laser system. The system uses CPA to produce femtosecond pulses that have an average maximum power of 600 mW, at a 1 kHz repetition rate, and each pulse carries about 0.9 mJ.

The oscillator produces the initial pulses used to seed the amplification system. At the output of the oscillator is a 93 MHz beam, with a central wavelength at approximately 800 nm, a pulse width of 20 fs and an average power of 110 mW.

The pulses from the oscillator travel to the stretcher. In the stretcher the pulses are taken from femtosecond pulses to picosecond pulses. The output beam from the stretcher is several picoseconds in pulse width at a 93 MHz repetition rate.

The output pulses from the stretcher go into the regenerative amplifier. The repetition rate of the pulses gets reduced to 1 kHz for the pulses that get amplified. Out of the regenerative amplifier come pulses at a 1 kHz repetition rate with a maximum pulse power of 15 GW, still at a pulse width of approximately 100 ps.

In the compressor the pulses from the regenerative amplifier get re-compressed to a pulse width on the order of femtoseconds. The pulses from the compressor get used in the experiment. The in-house laser system provides pulses that are approximately 40 femtoseconds in duration, with a central wavelength of 800 nm, and a beam average power of 600 mW at 1 kHz repetition rate.

The pulse intensities are high enough to ionize the air when the beam is focused tightly. In the laser system, between the regenerative amplifier and the compressor is a half-wave plate polarizer combination that can be used to reduce the power of the beam.

3.2 DC Electric Field

The goal of the research is to find structure of electric fields by observing the second harmonic generated in the field. The electric field imaged is therefore a controlled variable and two setups will be considered. The two electrode configuration provided different structures that can be checked against modeling software. The electrode configurations are a parallel
plate capacitor and a thin blade with a perpendicular plate. The modeling software used is COMSOL.

![Circuit Diagram](image)

Figure 3.2: Basic circuit diagram of an resistor-capacitor circuit. The circuit layout is used to produce the probed electric fields, where the capacitor represents the electrode setup.

Figure 3.2 shows the basic resistor-capacitor circuit that is used to create the electric fields. The capacitor (C) is the two different electrode configurations.

### 3.2.1 Parallel Plate Capacitors Model

The first of the two electrode configurations to examine is a parallel plate capacitor (PPC).

![Parallel Plate Capacitor](image)

Figure 3.3: A parallel plate capacitor from top view. Top plate is at a nonzero voltage, $V_o$, and the bottom plate is grounded. The gap is filled with air.

Figure 3.3 shows the top down view of two parallel conducting plates separated by a gap of air ($d$). The bottom plate is held at 0 V and the top plate is held at a constant nonzero voltage $V_o$. Both of the plates have a large area $A$, such that the length of the...
plates is much larger in proportion to the plate separation. With these dimensions there is translational symmetry with an invariance in the direction parallel to the plates so that the electric potential and field between the plates only holds \( z \) dependence \[18\].

\[ V(r) = V(z) \quad \mathbf{E}(r) = E_z(z)\mathbf{k} \] (3.1)

Within the gap there is no charge, and the electric potential must satisfy Laplace’s equation

\[ \nabla^2 V(z) = \frac{d^2 V(z)}{dz^2} = 0 \] (3.2)

\[ \frac{dV(z)}{dz} = A \] (3.3)

\[ V(z) = Az + B \] (3.4)

The boundary condition \( V(0) = 0 \) sets \( B = 0 \) and \( V(d) = Ad = V_o \) sets \( A = \frac{V_o}{d} \). The electric field is \( \mathbf{E} = -\nabla V \)

\[ \mathbf{E} = -\nabla V = -\frac{d}{dz} \left( \frac{V_o}{d}z \right) = \frac{-V_o}{d} \mathbf{k} \] (3.5)

In (3.5) the electric field between the electrodes of the parallel plate capacitor is shown to be constant throughout the whole region. The PPC can easily be modeled in COMSOL, and the electrostatic potential between the plates of the PPC and electric field vectors for the PPC can be seen in Figure 3.4.

In the model the bottom surface is held at a positive nonzero voltage, and the top surface is grounded. As expected the vectors, in the right image of Figure 3.4, point from the positive plate straight to the grounded plate. Near the edge of the plates there is a fringing effect. The focus of the laser is placed in the center of the plates, so that it falls within the constant electric field, and does not get affected by the fringing.

The parallel plates used in the experiment have circular surfaces with a diameter of 30 mm. Near the breakdown voltage of air, 3 kV/mm, the capacitance of the parallel plates will be
Figure 3.4: Left: Voltage dependence in the gap between a positively charged (bottom) and grounded (top) surface of a finite length. The color bar is measured in kV. Right: normalized electric field vectors in between the positively charged and grounded sides. The vectors point straight up from the positive surface to the grounded surface in between the plates. Near the edge of the plates there are the fringing effects.

\[ C = \frac{\epsilon_0 A}{d} = 6.27 \text{ pF} \]  \hfill (3.6)

and the energy stored on the capacitor is

\[ U = \frac{1}{2} CV^2 = 28 \mu J \]  \hfill (3.7)

Structure wise this electrode setup is simple, and the model shows the expected electric field form we found in (3.5).

3.2.2 Parallel Plate Capacitor Experiment Configuration

The first electrode setup use is a parallel plate capacitor (PPC). The PPC is chosen for the simplicity. The data acquired with the PPC is a good tool to check if the electric field calculations are performed properly.

The parallel plates of the capacitor are two elevator bolts, with 30 mm diameters. The bolts are held in place by insulating plastic. One of the bolts is attached to the positive lead of a voltage supply, while the other is connected to the negative lead (or ground). Figure 3.5 shows a top, and a side view image of the setup.
Figure 3.5: Left: Top view of the parallel plate capacitors, shows the centering on the focus, with the beam in the center. Right: tilted-side view of the capacitor, showing the height centering of the plates to the focus.

A resistor is placed between the voltage supply and grounded plate. The resistor ensures that there is a load to drop the voltage over if there is an electrostatic discharge event between the plates.

The voltage supply is a BERTAN 377P high voltage supply, able to supply up to 7 kV. Breakdown of air occurs at an electric field strength of $3 \times 10^6$ V/m, or 3 kV/mm [9]. The plate separation determines the maximum voltage that the plates can hold before an electrostatic discharge even occurs. The plates straddle the focus and are brought in close to each other. The gap spacing is brought down to 1-2 mm. With a 1 mm gap the plates can be brought up to almost 3 kV before breakdown occurs.

3.2.3 Pin and Plate Model

To observe the vector structure in the electric field another electrode setup is used. Figure 3.6 shows a conducting plate held at a nonzero voltage ($V_o$) and a grounded pin. The pin has a rounded tip.

The electrode configuration as shown in Figure 3.6 is modeled using COMSOL. The COMSOL model has a positively charged bottom plate and a grounded, rounded tip, needle. The model is symmetric about the 0 position on the x-axis. Figure 3.7 shows the electric potential and the normalized vector dependence of the electric field.
Figure 3.6: Top view of a grounded pin close to a plate held at nonzero $V_o$. The space around them is air.

Figure 3.7: Left: Electric potential in the space between a positively charge surface (bottom) and a grounded pin. Right: normalized electric field vector structure. The electric potential, on the left in Figure 3.7, shows the positively charged bottom plate and the grounded surface of the needle. The vector plot on the right shows the expected field structure of a small pin and a large plate electrode configuration. Figure 3.8 shows the electric field vector dependence with scaled vectors. The larger vectors represent the stronger electric field.

Structure that hold the same proportions as the pin and plate configuration are expected to have similar vector dependence in the electric field as seen in Figure 3.8. As you move further way from the pin the electric field gets weaker. The Electric field is strongest along the center line of the pin and the plate. Electric field lines become perpendicular to any surface because of the continuity equations in the electric field. As well, when you look
further away from the pin, the curvature of the electric field lines becomes weaker.

### 3.2.4 Blade and Plate Experimental Configuration

The rounded needle in the COMSOL modeling above is replaced by a long thin blade for the experiment. From the oncoming view of the laser this configuration still looks like a needle and a plate, but instead of being at one location, it is along a longer portion of the focus.

The plate is an elevator bolt (diameter = 3 cm) and the blade is the blade from a utility knife. The plate is connected to the positive terminal of the voltage supply and the blade is attached to a resistor that goes to ground. Figure 3.9 shows a top and side view of the electrode configuration.

The blade and plate are collectively on a translation stage that moves perpendicular to the beam propagation direction. On that translation stage, the plate is on another translation stage to set the spacing between the blade and the plate. The blade is on a vertical translation stage to move the blade vertically.
The translation stages can be used to move the electrodes with respect to the laser. Figure 3.10 shows the grid used for data collection. The grid spacing is 0.2 mm in both directions. As the electric field is symmetric across the blade, only the top or bottom half has to be considered in the grid.

### 3.3 Data Collection Methods

Retrieving the electric field vector dependence from the SH signals can be done by using the vector dependence of the SH signals. To collect information on the SH signal two data collection methods were used to observe the SH generated. The first is with the use of a photomultiplier tube (PMT), and the second uses a CCD (charge coupled device) camera.
3.3.1 Retrieving the Vector Field

The goal is to use the SH signal produced from (2.55) and find the vector structure of $E_{DC}$. There are two approaches to finding the spatial dependence. Given a polarization direction for the input fundamental, $E(\omega)$, the electric field will be at some angle relative to the input polarization, as shown in Figure 3.11. To retrieve the $E_{DC}$ spatial dependence a polarizer is placed in front of the camera.

$$E(\omega)$$

$$E_{DC}$$

Figure 3.11: The applied electric field is at some angle $\theta$ to the fundamental electric field polarization, $E(\omega)$.

When the polarizer is aligned along the polarization of the input beam, the signal reaching the camera will have the form

$$P(2\omega) \cdot \hat{e}_\parallel = \frac{1}{4} \varepsilon_o (6\chi_{1122}(2\omega; \omega, \omega, 0) + 3\chi_{1221}(2\omega; \omega, \omega, 0)) E_{DC} E^2 \cos \theta$$

(3.8)

The polarizer can also be set perpendicular to the fundamental polarization. The polarization then follows the form

$$P(2\omega) \cdot \hat{e}_\perp = \frac{1}{4} \varepsilon_o 3\chi_{1221}(2\omega; \omega, \omega, 0) E_{DC} E^2 \sin \theta$$

(3.9)

With either of the polarizer orientations the intensity on the camera with have the form

$$I_{signal} = P(2\omega)^2 = \Gamma E_{DC}^2 (E^2)^2 = \Gamma E_{DC}^2 I_\omega^2$$

(3.10)

here $\Gamma$ represents the square of the constants in (3.8) and (3.9).
For both scenarios, the polarizer being parallel or perpendicular to the fundamental input polarization, two data points are taken. The two data points are taken with two different input polarization’s that are 90° to each other, horizontally and vertically polarized light.

By getting the signal in both the horizontal $(x)$ and vertical $(y)$ directions, vector points can be made by retrieving the electric field information form each image and combining them into $(x, y)$ vectors.

### 3.3.2 Expected Background Second-Harmonic Structure

The signal due to the quadrupole expansion in the electric field is referred to as the background second-harmonic (BSH) signal. For the BSH it is easy to get the intensity profile by squaring the dominant term of the polarization equation in (2.43).

$$P(2\omega) = \chi_Q(\omega, \omega)(E \cdot \nabla)E$$

The square of the polarization equation will give the expected spatial dependence of the BSH signal.

![Figure 3.12: Theoretical structure of the BSH signal. The structure is both found for horizontally (left) and vertically (right) polarized input light.](image-url)
The structure of the BSH signal is shown in Figure 3.12. The left and right plots are for a horizontally and vertically polarized input beam respectively. The expected BSH structure is a doubly lobed structure where the lobes lie along the direction of the input polarization.

As the BSH signal is polarized in the direction of the input polarization, tuning the polarizer perpendicular to the input beam polarization will remove most of the BSH. The cross-polarization would cut out the BSH signal and only image the SH due to the applied electric field.

### 3.3.3 PMT Set Up

A photomultiplier tube (PMT) is used to observe the generated SH signal. A top view of the lab table layout is shown in Figure 3.13.

![Figure 3.13: Top view of PMT collection layout. After the beam is focused between the electrodes, the beam is collimated and sent onto a prism that takes the second-harmonic away from the fundamental so that it can be shown onto the PMT.](image)

The incoming beam is focused between the electrodes using lens L1. After the focus the beam is collimated, with L2, and redirected onto a prism. The prism separates the second-harmonic from the fundamental. The second-harmonic is redirected onto the PMT after passing through infrared filters. The mirrors M1 and M2 are coated for 400nm at 0°. Through the use of the coated mirrors, the dichroic mirror (DM), the prism and the infrared
filters the fundamental signal is cut out and the resulting signal on the PMT is due to the second-harmonics.

The PMT is connected to an oscilloscope where the voltage response to signal can be observed. The PMT used is a Burle negative voltage PMT that can be brought up to 2000 V and has a high quantum efficiency in the 400 nm region. The oscilloscope records the voltage profile from the PMT triggering on the 1 kHz signal from the laser system.

With the PMT the SH signal voltage can be recorded for analysis, and for confirmation of the presence of a signal.

3.3.4 CCD Camera Setup

The second method of data collection is imaging the second-harmonic response on a CCD camera. As with the PMT, the beam is focused between the electrodes and collimated after using L2. Using mirrors coated for 400 nm light the beam is redirected onto the camera, see Figure 3.14. The beam is focused onto the camera using another lens (L3). The dichroic mirror (DM), turning mirrors (M1 and M2), and the infrared filters are used to filter out the fundamental from the second-harmonic signal.

![Figure 3.14: Top view of CCD camera data collection. The laser gets focused between the electrodes and collimated as the beam is redirected towards the camera. The beam gets focused thought IR filters onto the camera. The mirrors (M1 and M2), dichroic mirror (DM), and IR filters are used to decrease the amount of fundamental light that reaches the camera.](image)

Before the initial focusing lens is a half-wave plate (1/2 WP) that is used to set the input polarization to horizontally or vertically polarized light. A polarizer (PL) can also be placed...
in the beam path to the camera to observe the signal polarization dependence.

The CCD camera used is a cooled PIXIS 1024F camera. The quantum efficiency (QE) of the camera is shown in Figure 3.15. The black curve in the figure is the QE for the specific camera used. The SH signal has a central wavelength around the 400 nm and as shown on the QE plot the camera is only a couple percent efficient at seeing the signal.

![Figure 3.15: Quantum efficiency of PIXIS 1024 Cameras. The F series is the camera used in the experiments for this thesis. The QE of the camera is only 2-3% efficient for the SH signal. [19]](image)

The CCD camera is controlled with the program LightField. Within LightField the cooling temperature (set to -70°C) and exposure time can be set. Through LightField, .tiff files are created to be analyzed further after the experiment is completed. The .tiff files hold the information of counts per pixel.
The goal is to find the vector dependence of an electric field. Here we aim to get an image of the electric field strength and vector direction. The signal from the background second-harmonics (BSH) is sufficiently high that we need to minimize it and subtract it off our images with an applied electric field. We need to understand the BSH well to be able to see if there are ways to optically cancel it out. We characterize the electric field dependent signal when the electric field is uniform, and set up electrodes with a spatially varying electric field to make an image of the vector field.

4.1 Background Second-Harmonics

The intensities in the beam are strong enough to create a second-harmonic signal when the beam is focused down in air. As discussed in Chapter 2, the SH signal is classically forbidden as air is a centrosymmetric medium, but owing to quadrupole and other higher order effects, there is a SH signal that is comparable in strength to the electric field induced signal. The SH signal is classified as the background SH (BSH) signal, which is present with or without externally removing the centrosymmetry of air. The BSH is characterized by looking at the SH structure on the camera, and the dependence on input polarization. The BSH response to input laser power and focal length of focusing lens is also observed. The BSH can be minimized or potentially eliminated by understanding and characterizing the signal.

4.1.1 BSH - Spatial Structure

In Chapter 3 the structure of the BSH is discussed. The expected BSH structure is two lobes oriented along the input polarization direction as shown in Figure 3.12. Figure 4.1 and Figure 4.2 show BSH signals on the camera. For both the sets of images the laser power was
set to 150 mW and the exposure time on the camera was 100 s. For the images in Figure 4.1 a half wave plate was used to set the input polarization to horizontally and vertically polarized light before the focusing lens, L1 in Figure 3.14.

![Figure 4.1: SH response on camera without an applied voltage to the electrodes. The input polarization is set to horizontal (left) and vertical (right).](image)

The signals in Figure 4.1 show the expected double lobe structure with an additional round spot in the center. The lobe orientation behaves as expected and is oriented along the direction of the input polarization. The strength of the round spot in the center is dependent on the input polarization. The round spot in the SH response comes from the addition of the half-wave plate in the beam path. The input polarization from the laser, before the half-wave plate, is close to vertically polarized light. The round spot is stronger for horizontally polarized light as the half wave plate has to turn the polarization from the laser system more than for vertically polarized light. The BSH was observed without the use of the half-wave plate. Figure 4.2 shows these responses.

In Figure 4.2 the input polarization is set by the laser system, and is mostly vertical polarization. A polarizer was placed after the collimation lens, L2 in Figure 3.14. In the left image of Figure 4.2 the polarizer was oriented to let through horizontally polarized light, and in the right image the polarizer was set to let through vertically polarized light.
Figure 4.2: SH response on the camera with no voltage applied to the electrodes, and no half-wave plate used. Left: polarizer after the collimating lens was set to let through horizontally polarized light. Right: the polarizer was set to let through vertically polarized light.

The BSH spatial structure without the half-wave plate has the expected two lobe structure. With the half-wave plate the BSH signal is stronger in the round spot and can be minimized. The voltage response of the BSH on the PMT shows that the SH signal has a strong dependence on the tip-tilt orientation of the half-wave plate. When the half-wave plate is set slightly off-axis in the beam path there are large voltage spikes on the PMT. The half-wave plate is required to orient the input polarization to horizontally and vertically polarized light. With the BSH going to the PMT, the half-wave plate can be oriented in a way to minimize the voltage response on the PMT, and decrease the BSH before starting an experiment.

4.1.2 BSH - Polarization Dependence

The polarization of the input beam was set to be horizontal, and the SH signal was directed onto the PMT. A polarizer set to let horizontally polarized light through was placed behind the collimation lens. The voltage response on the PMT was recorded as a function of the polarizer angle relative to the input polarization. Figure 4.3 shows the normalized voltage (to maximum voltage recorded) response to the angle between the input polarization and the polarizer.
The voltage response follows the expected sinusoidal response as the angle between the input polarization and polarizer varies from 0 to 360°. As seen in Figure 4.3 there is an constant offset, as the normalized voltage never reaches the 0 %. The offset is present without the half-wave plate to set input polarization. This tells us that there is some SH signal in the background that is not in the direction of the input polarization. From these findings we can tell that there is more to the BSH signal than represented by the second-order susceptibility process with the quadrupole expansion in the electric field. We suspect that the $\nabla E^2$ term in (2.43) might be making a contribution, since that would produce a radially polarized signal.

4.1.3 BSH Response to Laser Power

Using the PMT, the background second-harmonic response to increasing laser power was measured. The focal length of the focusing lens used was 400 mm. The polarizer located between the regenerative amplifier and compressor was used to vary the laser power from 30 to 500 mW. While varying the laser power the voltage response on the PMT was recorded.

The BSH on the PMT starts with a quadratic response, from 30 to 100 mW, as seen on Figure 4.4. The voltage response becomes linear from 150 to 270 mW and then levels off to a
constant response. Between the 100 and 150 mW there is a bump, which is still unexplained. The bump might be due to movement of the half-wave plate when it was bumped during the experiment. Future experiments will look into imaging the SH signal while varying the laser power to observe changes in the SH signal.

The quadratic and linear response are expected in the response, as well as the flattening out at higher power. Near the rounding off of the signal the air becomes visibly ionized, around the 270 mW.

All data collected for the SH response with a voltage applied to the electrodes was in the lower power range so that there was no ionization of the air. Most data was taken with the laser power between the 100 and 200 mW.

4.1.4 BSH Response to Focal Length

The BSH signal was imaged with two different focal length lenses for L1 in Figure 3.14. For all images in Figure 4.5 and Figure 4.6 the laser power was 150 mW and the camera exposure time was set to 100 s. The lenses have focal lengths of 200 and 400 mm. The BSH was observed without a half-wave plate in the beam path, these SH signals are shown in Figure 4.5.
Figure 4.5: BSH signal through polarizer set to let through horizontally (top) and vertically (bottom) polarized light for a 200 (left) and 400 mm (right) focal length lens. (a) shows double lobe structure in line with the horizontal polarization. (b) the signal is too weak to be picked up on the camera in 100 s. (c) double lobe structure is present as expected. (d) there is a faint double lobe structure.
The BSH signal appears to be stronger for the shorter focal length lens. In Figure 4.5 the images in the left column are for the 200 mm lens, and in the right column the 400 mm lens. The top row shows the SH signal through a polarizer set to let through horizontally polarized light, and the bottom had the polarizer set to let through vertically polarized light.

The same was done for the beam passing through the half-wave plate to rotate the input beam polarization to horizontally and vertically polarized light.

Figure 4.6 shows the BSH response when the half-wave plate is used. The round spot response in the center of the image is back. The top and bottom row are for horizontally and vertically polarized light in, and the left and right columns are for the 200 and 400 mm lenses respectively.

Both Figure 4.5 and Figure 4.6 show that the BSH signal is stronger for the shorter focal length lens. With the half-wave plate in place the double lobe structure is weaker than the round spot due to the half-wave plate. For the 400 mm signal the double lobe structure of the BSH is completely drowned out by the central dot.

Ideally the longer focal length lens is used to probe the electric field. The fact that the desired SH signal is drowned out by the half-wave plate makes the distinction between the desired SH signal and the BSH signal difficult to see. To see the SH signal for the cases with an applied voltage on the plates the 200 mm lens was used so that there was a significant change in the intensities in the images compared to the BSH.

4.2 Parallel Plate Capacitor

Using the half-wave plate the polarization of the input beam was set to horizontal and vertical polarization and sent between the plates of the parallel plate capacitor (PPC). For each polarization, images of the SH signal were acquired with a 20 s exposure time on the camera for voltages on the parallel plates from 0 to 2.4 kV in steps of 200 V. The horizontally and vertically polarized light are set to be perpendicular and parallel to the PPC plate surfaces respectively.
Figure 4.6: BSH signal for horizontally (top) and vertically (bottom) polarized input beam with a 200 (left) and 400 mm (right) focal length lens. (a) see the expected double lobe structure that is being overshadowed by the central circular signal. (b) no sign of a double lobe structure, just the circular signal. (c) double lobe structure is there with similar strength to the central circular signal. (d) no sign of a double lobe structure.
The SH signal on the camera with horizontal and vertical polarization are shown in Figure 4.7. The left and right sets of 4 × 3 images are for horizontal and vertical input polarizations respectively. For the last image in both sets there were electrostatic discharge events occurring between the two plates while the camera was imaging the SH signal.

![Figure 4.7: SH response on the camera with 20 s exposure time. In steps of 200 V applied to the PPC the SH signal was measured. These SH images have the 0 V signal already subtracted. (a) horizontally polarized beam. (b) vertically polarized beam.]

From the SH images with horizontal input, Figure 4.7a, a clear increase in the SH response is seen with an increase in voltage. For the vertical polarization there is a small increase, but most of the signal looks like background noise. The image of the fundamental signal is used to retrieve the electric field from the SH images. The fundamental signals are shown in Figure 4.8

The electric field at the focus from the horizontally polarized beam are shown in Figure 4.9. The images show a clear increase in the strength of the electric field with an increase in potential difference between the plates. At the higher voltages there is a small fringing effect along the boarders of the electric field image. The fringing is due to the slight miss overlap in the fundamental and SH centers. Within the calculation for the electric field the center of the fundamental and SH signal are aligned, with an error of ±5 in pixels on a 80 × 80 image. The same is done for the SH due to the vertically polarized beam, as seen in
Figure 4.8: Images of the fundamental signal for horizontally (left) and vertically (right) polarized light. Filters were used to block the SH signal from reaching the camera.

Figure 4.10.

The fringing effect is largest when the applied electric field is 3 kV/mm, the last image. During the exposure time of the image there were several electrostatic discharge events, so the plates were not held at a constant 2.4 kV during the exposure time. The inconsistent electric field in the area between the plates results in a non-uniform electric field, which is seen in the image.

The electric field in the vertical direction for the different voltages is shown in Figure 4.10. The overall signal looks noisy and there is no clear increase in the strength with an increase in the voltage.

The second-harmonic signal in the horizontal (Figure 4.9) and vertical (Figure 4.10) directions shows that the electric field is mainly perpendicular to the plates as expected. There is some electric field signal in the vertical direction because the plates are not perfectly flat, nor perfectly parallel to each other. With the majority of the signal in the horizontal direction, the electric field behaves like the expected form in (3.5).

The method of retrieving the electric field is shown to be correct from the PPC experiment as they are constant in the center. In addition, the PPC experiment clearly shows the increase in SH response to the increase in electric field, and that the expected constant
Figure 4.9: Plots of the electric field structure from the SH signals and the fundamental signal for the horizontally polarized light. The label above the blocks refers to the electric field strength on the PPC, measured in V/mm.

electric field exists in between the parallel plates. The slight fringing effect could be due to the simplification in the relationship between the signal on the camera and the calculated electric field.

4.3 Vector Field: Blade and Plate

The blade and plate electrode configuration has an expected electric field as seen in Figure 3.7. The plate and blade were moved with respect to the laser to create a grid as in Figure 3.10. The grid points are spaced 0.2 mm both in the $x$ and $y$ direction. The laser power was kept around the 180 mW at the focusing lens.

The spacing between the tip of the blade and the edge of the plate was 1.8 mm, and the potential difference between the blade and the plate was set to 3.2 kV. Figure 4.11 shows the images collected with the camera, with a 20 s exposure time. The left and right side are for horizontal and vertical polarization inputs respectively. The images have the BSH already
Figure 4.10: Electric field signals for vertically polarized light from the SH and fundamental signals. Above each image is a label that refers to the electric field strength, in V/mm, that is applied to the plates.

When setting the input polarization either to horizontal or vertical, the PMT was first used to minimize the BSH so that the wanted SH signal was not lost in the images. The individual images in the $19 \times 10$ sets in Figure 4.11 and Figure 4.12 are the data points taken at the respective grid points. For these sets of images the positively charged plate is on the right side, and the blade is in the lower left. For the first 7 elements of the second to last row, and first 10 elements of the last row the beam path was partially or completely disrupted by the blade.

The SH images are used to find the electric field signals at each point on the grid. The electric field images can be seen in Figure 4.12.

The left and right sides of Figure 4.12 are for the horizontal and vertical input polarization’s. With respect to the grids the blade is in the lower left. The blade was in the way of the beam for the first 9 elements of the bottom row. The plate is located to the right of the
Figure 4.11: SH images on camera with 20 s exposure time. Left: horizontal polarization in with the laser, and a horizontal polarizer after the focus. Right: vertical polarization in with the laser, and a vertical polarizer after the focus.

Figure 4.12: Electric field per image taken over a grid spacing of 0.2 mm near the blade edge. Left: The electric fields for the horizontally polarized laser in. Right: Vertical polarization electric fields images.

For one grid point the maximum electric field strength for the horizontal and vertical input polarization are combined to form a \((x, y)\) vector that indicates the direction of the electric field at that point. The same is done for all other grid points and the vectors are combined with the grid in a vector plot. Figure 4.13 shows the vector plot of the electric field.

The plot of the vector field for the blade and plate resembles the expected field structure shown in Figure 3.7. Near the edge of the plate the vector lines are perpendicular to the surface of the plate, and near the surface of the blade the vector lines become perpendicular to it. Directly along the line of the tip of the blade the vector lines all line up and point horizontally from the plate to the blade.
As the vector plot shows, the expected vector dependence of the electric field can be retrieved using an imaging method. The imaging method works well for the constant electric field.
CHAPTER 5
FUTURE DEVELOPMENT

The work in this thesis has shown that with the current measuring setup, we can find the vector dependence of constant-static electric fields. An improvement would be to image a time dependent electric field. Future work will consider applying similar methods as those described above to image the electric field of an electrostatic discharge (ESD).

ESD events are only a few nanoseconds long, and the current techniques will be insufficient to make measurements at such a short time scale.

5.1 Improved Imaging

The images taken during the experiments have exposure times of 20 to 100 s. To image a transient field in a spark discharge, we have two options. First, if we can trigger the spark repetitively in a way that produces the spark without temporal jitter that is larger than the transients we wish to observe, we can integrate the signal over many shots. Second, if the signal strength and the detection sensitivity can be made sufficiently high, we can collect data on a single shot.

Improvements can be made in the quantum efficiency of the camera, the transmission of the infra-red filters, and the amount of SH signal produced at the focus.

For a strong field, 2000 V/mm, over a 12 × 12 grid of pixels centered on the SH signal there are a total of 330,000 counts with an exposure time of 20 s. The quantum efficiency of the camera can be improved, and be brought from 3% to 60%. This is a factor 20 better, and could take the exposure time down to 1 s. During 1 s of our laser, there are still 1000 pulses that are being recorded.

For these measurements the shorter focal length lens, 200 mm, was used to focus down the beam. At this shorter focal length lens the power was brought close to the breakdown point of air, limiting the maximum pulse power. If the pulse width is stretched out, from 40
fs pulses to 80 fs pulses, then the input power could be doubled. This should give the same intensity and get the same conversion efficiency to the SH while leaving us with twice the output energy. This could take the exposure time down to 0.5 s to get a similar pixel count.

The IR filter set on the camera transmits 60% of the incoming light. These could be replaced by filters that let transmit 85%. By improving the transmission the exposure time could be brought down to 0.35 s.

For data taken with the 400 mm lens, for 60 second exposure time at 160 mW the counts on a 12 × 12 pixel grid over the center ranged from 400,000 to 700,000 for strong electric field strengths, between the 2400 and 2800 V/mm. The power can be increased up to 270 mW before ionizing the air. With that increase in power the exposure time can be brought down to 36 s. With a quantum efficiency increase of the camera for 3% to 60% the exposure time could be taken down to 1.8 s. By then also replacing the IR filters the exposure time could set to 1.2 s to get the same signal.

The improvement of the imaging system can be used to image time-varying electric field that vary of the scales of seconds to microseconds. The chances of using imaging to see the development of an ESD event are very slim.

Instead of focusing down to a spot, we can focus down to a line. With the 200 mm lens the spot size is about 35 µm. If we can instead focus down in one direction to 5 µm and expand the beam to 180 µm in the other direction, we can keep the same peak intensity with the same energy. Trying to go to a 180 µm spot would require 36 times more energy, which we do not have available to us. This can provide useful information when a line focus goes across or along a spark channel. This does not improve the exposure time of the camera much, but it will give more information about the electric field structure along the line focus in one single shot.

5.2 Pulse Sequence

A time varying electric field can be set up so that there is a well defined $t_0 = 0$ where the electric field starts to vary. For example there could be an electrostatic discharge event
that is started by a laser, and let that be set to $t_o$. To observe a time dependent field, measurements can be taken at different delay times with respect to $t_o$. This can be done at a shot-by shot basis, or with a sequence of pulses.

The current PMT set up can see SH generation from just focusing the laser in air starting at an power of 30 mW in our 1 kHz beam. The maximum power the beam can have in our system is around the 600 mW. The single beam can be split into 6 individual pulses each holding 80mW. A pulse train can come in to the time varying electric field and the waveform on the PMT can show the electric field strength variations.

A multipass ring could be built around the electrodes. The ends of the cavities would be two curved mirrors that are each placed 1 focal length away from the spark (or electric field of interest). The mirrors are tilted off axis in the same direction a bit so that the beam can go in a triangular path around the electrodes. The side mirror is adjusted to make the beam take multiple passes. For such a configuration the full pulse energy would be required. The path length of triangular ring will determine the pulse spacing. This type of configuration has been used in an amplification stage build be Backus et. al [20]. Their amplifying system was able to have 16 passes through the focus.

A pulse sequence will allow us to sample the electric field at multiple instances in time and provided the time dependence of the electric field. This is useful for electric fields that are fluctuation such as an electrostatic discharge.

### 5.3 Simultaneous Spatial and Temporal Focusing (SSTF)

Current experiments have operated with a beam from the normal compressor. Our in-house lasers system has another compressor that simultaneously focuses in space and in time (SSTF). This methods of compressing allows for a short depth of focus while maintaining a larger sport size than the regularly compressed beam would have for the same depth of focus. With a shorter depth of focus there is expected to be less background second harmonic generation.
The SSTF beam would help get around any self-focusing issues that are seen in the regularly compressed beam at high enough powers. The SSTF beam is fairly wide and would require larger aperture optics, which can put a limitation of its use.

5.4 Final Thoughts

The method presented for finding a non-invasive field probe of the vector electric field works well for static electric fields. With the current camera the images made of the electric fields had relatively high exposure times which would make it difficult to observe time dependent electric fields. A camera with a higher quantum efficiency for observing the SH could lead to imaging time dependent electric fields on the ms scale.

The goal is to apply the method of using SHG to gather information from the electric field to electrostatic discharge. The electrostatic discharges happen on nanosecond scales and imaging the time dependence would be impossible. To observe the time depended electric field we can also in a series of pulses, and have a PMT capture the wave form.

The current system will need some rearranging and upgrading before we are able to use second harmonic generation to probe the electric field of a plasma of an electrostatic discharge. But when a collection method is established, the SH signal will be able to provide us information about the electric field of the electrostatic discharge event.
REFERENCES CITED


