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Exploring Novel Crystals and Designs for Acousto-Optic Devices

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Exploring Novel Crystals and Designs for Acousto-Optic Devices

by

Jonathan B. Pfeiffer

B.S., University of Colorado, 2009
M.S., University of Colorado, 2012

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has been approved for the Department of Electrical and Computer Engineering

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Kelvin H. Wagner

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Date _________________

The final copy of this thesis has been examined by the signatories, and we find that both the content and the form meet acceptable presentation standards of scholarly work in the above mentioned discipline.
Abstract

Acousto-optic devices are a versatile technology that are driven electronically to precisely and rapidly control the intensity, frequency, and propagation direction of a laser beam. Applications include acousto-optic scanners, filters, mode lockers, and modulators. Despite the popularity of acousto-optic devices, there currently is no UV transparent device that can satisfy the requirements of the atomic clock and quantum computing communities. In this thesis, I describe my experimental efforts for discovering a new UV transparent, acousto-optic crystal that can meet the experimental requirements. I also present my graphical representations for locating practical and efficient acousto-optic designs in a given medium.

The first part of this thesis describes how to measure the elastic-stiffness and photoelastic coefficients of a given crystal. The elastic-stiffness coefficients are essential for designing acousto-optic devices because they determine the velocity, diffraction, and polarization of acoustic waves in a given medium. I used both resonant ultrasound spectroscopy and a modified version of Schaefer–Bergman diffraction to measure elastic coefficients. I discuss in detail the strengths, differences, and similarities of the two experiments. The photoelastic coefficients are necessary for determining the diffraction efficiency of a given acousto-optic geometry. Similar to the elastic coefficients, I employ a modified version of the Schaefer–Bergmann experiment to measure the photoelastic coefficients. I corroborate the measured results with the well established Dixon experiment.

The second part of this thesis describes four different graphical representations that help locate practical and efficient acousto-optic designs. I describe in detail each algorithm and how to interpret the calculated results. Several examples are provided for commonly used acousto-optic materials.
The thesis concludes by describing the design and performance of an acousto-optic frequency shifter that was designed based on the culmination of my research effort.
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The quality of the research presented in this thesis would be a shadow of itself if not for my advisor Kelvin Wagner. The energy, knowledge, and creativity Kelvin provided facilitated the progress of my research. I also thank Kelvin for spending countless hours teaching me science and how to succeed as an experimentalist. Many thanks to Dana Anderson for providing me with my first researching position that over the course of two and half years taught me the fundamentals of research. Lastly, I thank Juliet Gopinath for the year and half I spent in her lab learning and growing as a researcher.
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Chapter 1

Introduction and Motivation

A quantum computer (QC) takes advantage of quantum-mechanical phenomena, such as superposition and entanglement, to simultaneously process a superposition of computation paths and produce a final state that is an interference of these paths [1]. This parallel processing power allows a QC to solve certain problems more efficiently than a classic computer. One popular example is Shor’s algorithm, which shows that a QC can factor large numbers, an important problem in cryptography, exponentially faster than a Turing machine [2]. Grover’s algorithm, which searches for an entry in a database, scales more efficiently as the size of the search space increases [3]. Of particular interest to physicists is modeling quantum mechanical systems, an intractable problem for modern computers but feasible with a QC because the computer itself is a quantum system [4]. In the last twenty years, significant research effort has been dedicated to the realization of a QC.

In an attempt to guide the research efforts for QCs, David DiVincenzo enumerated the requirements for a successful QC. These requirements are known as the DiVincenzo criteria [5]:

1. Well-defined, two-state quantum subsystems known as qubits (quantum bits).
2. The ability to initialize all qubits into a known quantum state.
3. Gate operation times that are much faster than the qubit decoherence times.
4. A universal set of quantum gates between the qubits that can be implemented with controllable interactions of the quantum system.
5. A method for reading out the state of any qubit with high fidelity.
(6) Scalable to a large number of qubits.

Any QC that satisfies all of the DiVincenzo criteria will be able to initialize, process, and store qubits.

Ignacio Cirac and Peter Zoller in 1995 proposed using a string of cold, trapped ions for quantum computation [6]. In their QC, the internal energy levels of the ions would act as the $|0\rangle$ and $|1\rangle$ states of the qubit. Focused laser light would individually address and manipulate each qubit, and the Coulomb force between the ions would induce coupled oscillations of the ions. Cirac and Zoller showed that this coupled oscillation of the ions could be used to create a controlled-NOT (CNOT) gate, which can create an universal set of quantum gates [7].

David Wineland’s group in Boulder at the National Institute of Standards and Technology (NIST) has been pursuing a QC, based on Cirac and Zoller’s design, that satisfies the DiVincenzo criteria [8]. They employ long-lived hyperfine states of $^9\text{Be}^+$ for their qubits (criteria 1 and 3), apply optical pumping to achieve state initialization (criteria 2), and use state-dependent fluorescence to readout the qubit state (criteria 5). In 1995 the group demonstrated a two-bit CNOT gate with stimulated Raman transitions in $^9\text{Be}^+$ ions (criteria 4) [9]. The group is currently working on scaling the QC to satisfy criteria 6 [10].

The experimental efforts of Wineland’s group requires precise frequency and power control of several ultraviolet (UV) laser wavelengths near 313 nm [10]. The state preparation, optical cooling, stimulated Raman transitions, and optical readout all require $\sim$ 313 nm laser light finely tuned in frequency and power. For their current and future experiments with $^9\text{Be}^+$, they would like to manipulate the 313 nm with:

- Efficient and precise frequency and phase shift of the laser beam over 1.24 GHz
- Fast sub-microsecond access time
- Sufficient resolution to address array of ions (32-100 well resolved spots)
- Multiple simultaneous spot addressing as well as single beam deflection
- Possibility of two or even three-dimensional array addressing
• On-off contrast ratio in excess of 10,000:1
• High diffraction efficiency in the UV and visible at specific ion frequencies
• Pulse timing jitter less than 1 nsec

Additional ions proposed for quantum computing paradigms include $^{24}\text{Mg}^+$ and $^{40}\text{Ca}^+$ that require precise optical manipulation at 280 and 395 nm, respectively [11, 12]. Also both $^{199}\text{Hg}^+$ and $^{27}\text{Al}^+$, which require 194 and 267 nm optical manipulation, have been proposed for making atomic clocks that would be an order of magnitude more precise than $^{133}\text{Cs}$ clocks [13]. Therefore, there is a great demand for an optical device that can modulate, frequency shift, deflect, and manipulate laser beams to satisfy all of the requirements enumerated above at several different UV wavelengths.

The required sub-microsecond speed eliminates solutions based on liquid crystal spatial light modulators (SLM), fast steering mirrors, or even two-dimensional micro-electro-mechanical (MEMS) SLMs, which all at best operate at KHz frame rates. Acousto-optic (AO) devices, on the other hand, have the potential of meeting all of the requirements for the quantum computing community. Figure 1.1 illustrates an AO device. In an AO device, a piezo-electric transducer driven by an RF frequency creates a traveling acoustic wave. The strain from the acoustic wave creates a moving phase grating that diffracts the incident beam and frequency shifts the diffracted beam by the frequency of the acoustic wave. The polarization of the diffracted beam may be, but not necessarily, changed by the acoustic wave to an orthogonal polarization with respect to the incident beam polarization. The intensity, propagation direction, phase, and frequency of the diffracted beam can be finely and rapidly tuned by electronically adjusting the drive signal of the device.

Unfortunately current AO crystals with high diffraction efficiency ($\text{TeO}_2$, $\text{PbMoO}_4$, $\text{GaP}$) are not transparent below 355 nm. Wineland’s group is currently using fused silica AO devices, but the intrinsic low diffraction efficiency of the devices necessitates RF powers in excess of 5 W to achieve the required diffraction efficiencies. These large RF powers along with acoustic absorption create thermal drifts in the device, causing both the diffracted and undiffracted optical beam to
Figure 1.1: Acousto-optic device driven by an 80 MHz signal.

Drift because the optical properties of the crystal are temperature dependent.

Recently new measurements have been made on the acoustic and AO properties of barium borate (BBO), a crystal transparent down to 189 nm [14, 15, 16]. There are two different phases of BBO. The low temperature $\beta$ phase ($3m$) lacks a center of inversion and thus possesses a non-vanishing second-order nonlinear optic tensor. The moderately large nonlinear optical coefficients of $\beta$-BBO combined with the deep UV transparency makes $\beta$-BBO a highly attractive material for frequency doubling, tripling, and quadrupling. Hence, hundreds of papers have been published about the nonlinear optical applications of $\beta$-BBO [17, 18]. The high temperature phase $\alpha$-BBO ($\bar{3}m$) is centrosymmetric, so it can not be used as a nonlinear crystal. As a result, few papers have been published about $\alpha$-BBO. However, the initial AO measurements of BBO have shown that the $\alpha$ phase has all the properties to emerge as the next AO material to satisfy all of the requirements of an UV scanner, frequency shifter, and modulator for the quantum computing community.

This thesis presents my research efforts for developing UV-transparent AO devices. Chapter 2 reviews the necessary physics for understanding the AO interaction. Chapter 3 discusses measuring the elastic-stiffness coefficients of a material with resonant ultrasound spectroscopy and Schaefer–Bergmann diffraction. Elastic-stiffness measurements of both $\alpha$ and $\beta$-BBO are presented
in Chapter 3. Chapter 4 explains how to measure the photoelastic coefficients of a material using AO diffraction with measurements of both fused silica and \(\alpha\)-BBO presented. Chapters 5 and 6 present different graphical representations for finding efficient AO geometries. The thesis concludes with Chapter 7 by discussing the performance of an \(\alpha\)-BBO AO device that I designed based on the culmination of my research efforts.
Chapter 2

Background Theory

This chapter provides a theoretical overview of the propagation of optical and acoustic waves and their interaction with each other in different media. For both the optical and acoustic waves, we assume plane wave solutions to solve for the velocity, wavelength, and polarization of the waves as a function of the propagation direction. A pictorial illustration in momentum space is provided that aides in understanding the solutions. The theory covered in this chapter will facilitate in understanding the measuring and designing techniques discussed in subsequent chapters.

2.1 Optical Waves in Homogeneous Media

In this section the allowed optical modes of propagation will be derived for different classes of crystals. The media in which the light is propagating is assumed to be linear, homogeneous, and free of both current and charge, resulting in the simplified Maxwell’s equations written in differential form:

\[
\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \quad \text{(Faraday’s law)} \tag{2.1}
\]

\[
\nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t} \quad \text{(Ampere’s law)} \tag{2.2}
\]

\[
\nabla \cdot \vec{B} = 0 \quad \text{(Gauss’s law for a magnetic field)} \tag{2.3}
\]

\[
\nabla \cdot \vec{D} = 0. \quad \text{(Gauss’ law for electric field)} \tag{2.4}
\]

Maxwell’s equations describe and relate with respect to each other the electric field intensity \( \vec{E} \), the magnetic field intensity \( \vec{H} \), the electric flux density \( \vec{D} \), and the magnetic flux density \( \vec{B} \). The
four fields presented in Maxwell’s equations have a total of 12 unknowns, with 3 unknowns for each component of a given field vector. Maxwell’s equations also provides eight different equations. The vector equations, Eq. 2.1 and 2.2, provide 3 equations each, while the scalar equations, Eq. 2.3 and 2.4, provide 1 equation each. Equation 2.3 however, is not independent of the other three Maxwell’s equations, which is proven by taking the divergence of Eq. 2.1 and noting that the divergence of the curl is equal to 0:

\[ \frac{\partial}{\partial t} (\nabla \cdot \vec{B}) = 0. \] (2.5)

The above equation shows that the divergence of the magnetic flux density \( \vec{B} \) must be constant for all space and time. This constant is readily set to 0 because no magnetic charge has ever been experimentally observed. Therefore, Maxwell’s equations contain 12 unknowns with only 7 independent equations.

To uniquely define an electromagnetic field, Maxwell’s equations are augmented with six more equations known as the constitutive relations:

\[ \vec{D}(t) = \varepsilon_0 \int_{-\infty}^{t} \bar{\varepsilon}(t - t') \cdot \vec{E}(t') dt', \] (2.6)

\[ \vec{B}(t) = \mu_0 \int_{-\infty}^{t} \bar{\mu}(t - t') \cdot \vec{H}(t') dt'. \] (2.7)

Here \( \varepsilon_0 \) and \( \mu_0 \) are the permittivity and permeability of free space, and \( \bar{\varepsilon} \) and \( \bar{\mu} \) are the relative dielectric and permeability tensors of the medium. For rapidly varying fields, such as the fields associated with optical waves, the medium does not respond instantaneously, so the response depends on the previous fields in addition to the field at time \( t \). This can be seen by the causal convolution in the constitutive relations, which assumes that the medium is linear and time-invariant. Equations 2.6 and 2.7 can be simplified by converting from the time to frequency domain with a Fourier transform:

\[ \vec{D} = \varepsilon_0 \bar{\varepsilon}(\omega) \cdot \vec{E}, \] (2.8)

\[ \vec{B} = \mu_0 \bar{\mu}(\omega) \cdot \vec{H}, \] (2.9)
where

\[ \tilde{\epsilon}(\omega) = \int_0^\infty \tilde{\epsilon}(t)e^{i\omega t} dt, \]  
\[ \tilde{\mu}(\omega) = \int_0^\infty \tilde{\mu}(t)e^{i\omega t} dt. \]  
(2.10)

(2.11)

The advantage of converting to the frequency domain is immediately obvious as the integral relations of Eqs. 2.6 and 2.7 reduce to algebraic relations in Eqs. 2.8 and 2.9. This conversion reveals that a medium is dispersive when it cannot instantaneously respond to a time varying electromagnetic field. If the medium could respond instantaneously, then the convolution operator in the time domain would be unnecessary and the Fourier transform to the frequency domain would result in a constant \( \tilde{\epsilon} \) and \( \tilde{\mu} \) for all \( \omega \).

For the remainder of this thesis, I will assume that the material is nonmagnetic \( \tilde{\mu} = \bar{I} \) (where \( \bar{I} \) represents the identity matrix), so Eq. 2.7 simplifies to \( \bar{B} = \mu_0 \bar{H} \). The dielectric tensor \( \tilde{\epsilon} \) is reduced by conservation of energy and by requiring that the electric and magnetic energy densities to always be positive, which reduces \( \tilde{\epsilon} \) to a positive definite, symmetric tensor \([19, 20]\). Every symmetric tensor can be transformed to an orthogonal coordinate system in which the tensor reduces to a diagonal tensor:

\[ \tilde{\epsilon} = \begin{bmatrix} \epsilon_1 & 0 & 0 \\ 0 & \epsilon_2 & 0 \\ 0 & 0 & \epsilon_3 \end{bmatrix}. \]

Here the real constants \( \epsilon_1, \epsilon_2, \) and \( \epsilon_3 \) are the eigenvalues. Note that this transformation can depend on the frequency \( \omega \), so \( \tilde{\epsilon} \) can have off-diagonal elements in the crystallographic coordinate system.

The symmetry of the crystal can further reduce \( \tilde{\epsilon} \). Every crystal belongs to 1 of the 32 distinct point groups, which describe the symmetry operations that leave the crystal unchanged. Any crystal that has a symmetry with a three-dimensional irreducible representation requires the three eigenvalues of \( \tilde{\epsilon} \) to be equivalent \( (\epsilon_1 = \epsilon_2 = \epsilon_3) \) \([21]\). These crystals, known as optically isotropic crystals, possess cubic symmetry. Similarly crystals with a single axis of greater than
two-fold symmetry, known as uniaxial crystals, will have a 2-dimensional irreducible representation that requires two of the $\tilde{\epsilon}$ eigenvalues to be equivalent. Biaxial crystals do not possess an axis with a symmetry greater than two; hence, place no restrictions on the eigenvalues of $\tilde{\epsilon}$.

2.1.1 Monochromatic Plane Waves

The simplest solution to Maxwell’s equations is a monochromatic plane wave with sinusoidal dependence in both time and space. Plane waves provide accurate approximations to many real wave problems, and more complicated fields in linear media can be represented by a superposition of plane waves. The results presented in this chapter assume plane waves for both the optical and acoustic waves. Below we provide an overview of plane waves so that the solutions presented in this chapter can be better understood.

Let us consider a monochromatic electromagnetic wave with

$$\vec{E}[\vec{r}, t] = \text{Re}[\vec{E}_0 e^{i(k \cdot \vec{r} - \omega t)}], \quad \vec{H}[\vec{r}, t] = \text{Re}[\vec{H}_0 e^{i(k \cdot \vec{r} - \omega t)}].$$

(2.12)

Here $\vec{E}_0$ and $\vec{H}_0$ are complex amplitude vectors of the physical field, $\vec{r}$ is a position vector, $\vec{k}$ is a constant vector called the wave vector, and $\omega$ is the angular frequency. Equation 2.12 is constant for a constant phase $\phi_1$:

$$\phi_1 = \vec{k} \cdot \vec{r} - \omega t.$$  

(2.13)

For a fixed time $t$, Eq. 2.13 reduces to a plane of constant phase described by

$$\hat{k} \cdot \vec{r} = \zeta = \frac{\phi_1 + \omega t}{k},$$  

(2.14)

where $\zeta$ is the distance of the plane from the origin, and the wave number $k$ and the wave normal $\hat{k}$ are the magnitude and direction of the wave vector ($\vec{k} = k \hat{k}$). Figure 2.1a illustrates a plane of constant phase as described by Eq. 2.14. The plane is normal to $\hat{k}$ and is infinitely large. As time increases, the distance of the plane from the origin $\zeta$ must increase to keep the phase of the plane constant. The velocity at which the plane propagates along $\hat{k}$ can be determined by taking the derivative of Eq. 2.14

$$v_p = \frac{\partial \zeta}{\partial t} = \frac{\omega}{k}.$$  

(2.15)
Figure 2.1: (a) Plane of constant phase $\phi_1$. (b) Planes of constant phase with a phase difference of $2\pi$ between planes, traveling along $\hat{k}$.

Here $v_p$ is the phase velocity and travels along $\hat{k}$ ($\vec{v}_p = v_p \hat{k}$). The phase velocity is the speed of planes of constant phase, but this does not necessarily equal the speed of energy propagation $v_e$.

The separation of two planes of equal phase, $\phi_2 = \phi_1 + 2\pi$ can be measured in both time and space. In time, the separation between two planes with a phase difference of $2\pi$ is simply equal to

$$T = \frac{2\pi}{\omega}, \quad (2.16)$$

where $T$ is called the period of the wave. In space, the two planes separated in phase by $2\pi$ can be determined by

$$\phi_2 - \phi_1 = 2\pi = \hat{k} \cdot (\vec{r}_2 - \vec{r}_1) = k\lambda \quad (2.17)$$

The wavelength of the plane wave $\lambda = \frac{2\pi}{k}$ determines the spatial separation of the two planes, as illustrated in Fig. 2.1b. With the properties of plane waves well defined, the solutions of optical and acoustic plane waves will now be presented.

### 2.1.2 Dispersion Equation

Faraday’s and Ampere’s laws (Eqs. 2.1-2.2) are two coupled equations that describe how the spatio-temporally varying electromagnetic fields relate to each other. The two equations can be
decoupled by first inserting the constitutive relations Eqs. 2.8-2.9 into Eq. 2.2, and then applying the time derivative to both sides:

\[
\frac{1}{\mu_0} (\nabla \times \frac{\partial \vec{B}}{\partial t}) = \epsilon_0 \vec{\epsilon} \cdot \frac{\partial^2 \vec{E}}{\partial t^2}.
\] (2.18)

Here \( \vec{\mu} = \vec{I} \) as I am assuming that the material is nonmagnetic. Inserting Eq. 2.1 into the above eliminates \( \vec{H} \), and produces an equation that depends on \( \vec{E} \) alone:

\[
\nabla \times \nabla \times \vec{E} = -\mu_0 \epsilon_0 \vec{\epsilon} \cdot \frac{\partial^2 \vec{E}}{\partial t^2}.
\] (2.19)

To solve for Eq. 2.19, plane waves solutions (Eq. 2.12) are assumed. This assumption simplifies the spatial and temporal derivatives to \( \frac{\partial}{\partial t} \rightarrow -i\omega \) and \( \frac{\partial}{\partial x} \rightarrow ik \), reducing Eq. 2.19 to

\[
-(\vec{k} \times \vec{k} \times \vec{E}) = \omega^2 \mu_0 \epsilon_0 \vec{\epsilon} \cdot \vec{E}.
\] (2.20)

With the aid of the antisymmetric matrix \( \vec{k} \times \vec{I} \) defined as

\[
\vec{k} \times \vec{I} = \begin{bmatrix} 0 & -k_3 & k_2 \\ k_3 & 0 & -k_1 \\ -k_2 & k_1 & 0 \end{bmatrix},
\]

Eq. 2.20 may be rewritten as

\[
[k_0^2 \vec{\epsilon} + (\vec{k} + \vec{I})^2] \cdot \vec{E} = 0.
\] (2.21)

Here \( k_0^2 = \omega^2 \mu_0 \epsilon_0 \). This equation can be rewritten as a generalized eigenvalue problem

\[
[\vec{\eta} \cdot (\vec{k} \times \vec{I})^2] \cdot \vec{E} = \lambda \vec{E}.
\] (2.22)

Here \( \lambda = -k_0^2/\gamma^2 \) and \( \vec{E} \) are the eigenvalue and eigenvector of the matrix \( [\vec{\eta} \cdot (\vec{k} \times \vec{I})^2] \) and \( \vec{\eta} \) is the inverse of \( \vec{\epsilon} \) (\( \vec{\eta} = \vec{\epsilon}^{-1} \)). For a given plane wave direction \( \vec{k} \), the eigenvalues determine the magnitude of \( \vec{k} \), which determines the phase velocity of the plane wave \( v_p = \omega/k \), and the eigenvectors determine the polarization of \( \vec{E} \).

Faraday’s and Ampere’s laws (Eqs. 2.1-2.2) reveal that a spatio-temporally varying electric field \( \vec{E} \) creates a spatio-temporally varying magnetic field \( \vec{H} \); hence, a magnetic field plane wave
accompanies every solution of the electric field plane wave. Equation 2.22 determines the exact speed and polarization required of the electric field $\vec{E}$ such that the spatio-temporally varying $\vec{E}$ and $\vec{H}$ fields will continually create each other without losing energy.

For Eq. 2.22 to have a nontrivial solution, the determinant of the characteristic equation must vanish:

$$|k_0^2\vec{I} + \vec{e}^{-1} \cdot (\hat{k} \times \vec{I})^2| = 0,$$

(2.23)

where $|\vec{a}|$ represents the determinant of the matrix $\vec{a}$. Equation 2.23 is known as the dispersion equation, which can be solved for in explicit form:

$$(\hat{k} \cdot \vec{e} \cdot \hat{k})n^4 - (\hat{k} \cdot [(\text{adj} \vec{e})_t \vec{I} - \text{adj} \vec{e}] \cdot \hat{k})n^2 + |\vec{e}| = 0.$$

(2.24)

Here the index of refraction $n = k/k_0$ and $\text{adj} \vec{a}$ and $\vec{a}_t$ represent the adjoint and trace of the tensor $\vec{a}$, respectively [19]. Equation 2.24 is quadratic in $n^2$ and has four solutions for a given $\hat{k}$ (two solutions each for forward and backward propagating plane waves).

Gauss’s law combined with the constitutive relationship Eq. 2.8, $(\hat{k} \cdot \vec{\varepsilon}_0 \vec{\varepsilon}) \cdot \vec{E} = 0$, requires $\vec{E}$ to be orthogonal to $\hat{k} \cdot \vec{\varepsilon}_0 \vec{\varepsilon}$ for all propagating modes. Therefore, all propagating modes exist in a plane, which only takes two vectors to span. The two eigenvectors with non-zero eigenvalues from Eq. 2.22 span the plane orthogonal to $\hat{k} \cdot \vec{\varepsilon}_0 \vec{\varepsilon}$. The third mode does not propagate and therefore its eigenvalue is equal to zero, causing Eq. 2.24 to be quadratic in $n^2$ and not $n^3$.

Below I present the explicit solutions to Eq. 2.22 for isotropic and uniaxial media, and briefly discuss the solutions for biaxial media. I also present in each subsection a graphical tool called momentum space ($k$-space for short) to help visualize the solutions and physics as a function of propagation direction $\hat{k}$.

### 2.1.3 Isotropic Media

The symmetry of optically isotropic crystals and liquids requires $\varepsilon = \varepsilon_1 = \varepsilon_2 = \varepsilon_3$, which reduces the dielectric tensor to $\vec{\varepsilon}$. This reduction simplifies Eq. 2.24 to

$$\varepsilon_1 n^4 - 2\varepsilon n^2 + \varepsilon^3 = \varepsilon[n^2 - \varepsilon] = 0.$$

(2.25)
Here the $\hat{k} \cdot \hat{k} = 1$ terms have been removed. With $\hat{k}$ absent from Eq. 2.25, the solutions for $n$ are independent of the plane wave propagation direction $\hat{k}$. Therefore, these crystals are called optically isotropic because there is no way to distinguish different directions in the crystal with optical waves. Additionally Eq. 2.25 only depends on $n^2$, so there are only two solutions: one forward and one backward propagating wave.

Solving for the index of refraction for a forward propagating wave in Eq. 2.25 results in

$$n = \sqrt{\varepsilon},$$

$$k = nk_0 = \omega \sqrt{\mu_0 \varepsilon_0} = \frac{\omega}{c} n,$$

$$v_p = \frac{\omega}{k} = \frac{1}{\sqrt{\mu_0 \varepsilon_0 \varepsilon}} = \frac{c}{n},$$

(2.26)

for all propagation directions $\hat{k}$. In free space $\varepsilon = 1$, so $v_p = \frac{1}{\sqrt{\mu_0 \varepsilon_0}} = c$, where $c$ is known as the speed of light in free space. The index of refraction is almost always greater than 1, making light slower in a material $v_p = \frac{c}{n} < c$.

The eigenvalues of Eq. 2.22 for isotropic media can be solved for by setting $\lambda = -\frac{1}{\varepsilon}$:

$$\left[ \frac{1}{\varepsilon} \cdot (\hat{k} \times \hat{I}) \right] \cdot \vec{E} = \frac{1}{\varepsilon} \cdot \vec{E}.$$ \hspace{1cm} (2.27)

After eliminating the $\frac{1}{\varepsilon}$ term, the above equation can be rewritten as

$$\hat{k} \times \hat{k} \times \vec{E} + \vec{E} = 0.$$ \hspace{1cm} (2.28)

Using the vector identity $\vec{A} \times \vec{B} \times \vec{C} = \vec{B}(\vec{A} \cdot \vec{C}) - \vec{C}(\vec{A} \cdot \vec{B})$ and once again noting that $\hat{k}^2 = 1$, the above reduces to

$$\hat{k} \cdot \vec{E} = 0.$$ \hspace{1cm} (2.29)

Equation 2.29 states that any vector $\vec{E}$ orthogonal to $\hat{k}$ is an eigenvector.

With $\hat{k}$ and $\vec{E}$ defined, the directions of the three remaining electromagnetic fields can be
determined using Maxwell’s and the constitutive equations:

\[ \mathcal{H} = \frac{1}{\eta} (\hat{k} \times \mathcal{E}), \]
\[ \mathcal{D} = \epsilon_0 \mathcal{E} \cdot \mathcal{E}, \]
\[ \mathcal{B} = \mu_0 \mathcal{H}. \]  

(2.30)

Here \( \eta = \frac{\mu_0}{\epsilon_0 \epsilon} \) is the intrinsic impedance of the medium. The bottom equation reveals that \( \mathcal{B} \) and \( \mathcal{H} \) are parallel. For an isotropic material \( \mathcal{E} = \mathcal{I} \), so \( \mathcal{D} \) is parallel with \( \mathcal{E} \). From Eqs. 2.29 and 2.30, we see that the vectors \( \hat{k}, \mathcal{E}, \) and \( \mathcal{H} \) form a right-handed orthogonal triad of vectors, as illustrated in Fig. 2.2. This triad represents a transverse electromagnetic plane wave with the polarization field vectors \( \mathcal{E} \) and \( \mathcal{H} \) lying in a plane of constant phase. For isotropic media, the vectors \( \mathcal{E} \) and \( \mathcal{H} \) may rotate around \( \hat{k} \) and still be eigen-polarizations as long as they remain orthogonal with respect to each other.

The Poynting vector \( \mathbf{S} \), which gives the propagation direction of the energy of the optical beam, is calculated by \( \mathbf{S} = \frac{1}{2} (\mathcal{E} \times \mathcal{H}) \). For isotropic media, the Poynting vector \( \mathbf{S} \) is parallel with the wave vector \( \hat{k} \), as illustrated in Fig. 2.2.

For a constant optical frequency \( \omega \), the magnitude of the wave vector \( \hat{k} \) can be plotted as function of propagation direction \( \hat{k} \). The resulting surface is known as the wave-vector surface, the dispersion surface, or as will be referred to as in the remainder of this thesis, the momentum surface. The momentum surface is a powerful tool and displays all the information of plane wave
propagation. The magnitude, slope, and curvature of the surface all reveal useful information [22].
The radius of the sphere determines the wave number $k$, and therefore the index of refraction $n$, phase velocity $v_p$, and wavelength $\lambda$. The Poynting vector can be shown to be orthogonal the surface [22, 20], so the slope or first derivative of the momentum surface determines the direction of the power flow. In addition, the electric and magnetic fields are orthogonal to the momentum surface as shown by Eqs. 2.29 and 2.30. The curvature of the momentum surface can determine the diffraction of a finite beam composed of a superposition of planes waves. This will be discussed more in the case acoustic waves because the curvature does not vary significantly for optical momentum surfaces.

For isotropic medium, the momentum surface is a sphere with a radius of $k = nk_0$, where $n$ is the index of refraction of the medium. The single sheeted surface reflects the fact that the two non-zero eigenvalues of Eq. 2.22 are degenerate, which only limits the eigenvector $\hat{\mathcal{E}}$ to a tangent plane of the momentum surface. Figure 2.3 shows two eigenvectors that span a plane for various propagation directions $\hat{k}$. The Poynting vector is orthogonal to the surface, which is equivalent to $\hat{k}$ for a sphere.
Figure 2.4 illustrates a cross-section of the momentum surface and compares momentum space to real space. Every point on the momentum surface represents a plane wave. As already discussed, the geometry of the surface determines the wavelength, polarization, and the Poynting vector of the plane wave. Figure 2.4 illustrates a point in momentum space and the corresponding plane wave in real space. The polarization is not provided because for isotropic materials it is not uniquely defined for a given propagation direction.

2.1.4 Uniaxial Crystals

For uniaxial crystals, $\epsilon_1 = \epsilon_2 \neq \epsilon_3$. Traditionally the z-axis is associated with the axis of highest symmetry; hence, $\epsilon_3$ is associated with the z-axis. Solving for the dispersion equation (Eq. 2.24) for uniaxial crystals results in

$$\left( k^2 - n_o^2 k_0^2 \right) \left( \frac{k_x^2 + k_y^2}{n_e^2} + \frac{k_z^2}{n_o^2} - k_0^2 \right) = 0. \quad (2.31)$$

Here $n_o^2 = \epsilon_1$ and $n_e^2 = \epsilon_3$ are the ordinary and extraordinary indices of refraction and $k_0 = \frac{2\pi}{\lambda_0}$ is the wave vector in free space. There are two solutions to Eq. 2.31. The first solution, corresponding
to leftmost factor equaling zero, is a sphere of radius

\[ k = n_o k_0. \]  \hspace{1cm} (2.32)

The second solution, corresponding to the rightmost factor equaling zero, is an ellipsoid with rotational symmetry about the z-axis:

\[ \frac{k_x^2 + k_y^2}{n_e^2} + \frac{k_z^2}{n_o^2} = k_0^2. \]  \hspace{1cm} (2.33)

Switching to spherical coordinates sets \( k_z^2 = k_0^2 n_e^2(\theta) \cos^2 \theta \) and \( k_x^2 + k_y^2 = k_0^2 n_e^2(\theta) \sin^2 \theta \), which allows us to solve for \( n_e(\theta) \):

\[ \frac{1}{n_e^2(\theta)} = \frac{\cos^2 \theta}{n_o^2} + \frac{\sin^2 \theta}{n_e^2}. \]  \hspace{1cm} (2.34)

Figure 2.5 illustrates the momentum surface for a negative uniaxial crystal \((n_e < n_o)\). The red surface, known as the extraordinary surface, is the ellipsoid and the blue surface, known as the ordinary surface, is the sphere. Both surfaces touch along the z-axis, which is commonly known as the optic axis. This single axis where the eigenvalues are degenerate is why the media is known as uniaxial.

The ordinary and extraordinary electric field eigen-polarizations \( \hat{E} \) lie along the latitudes on the sphere and the longitudes of the ellipsoid, as illustrated in Fig. 2.5. Both eigen-polarizations are tangent to their respective momentum surfaces. In spherical coordinates where, \( \hat{k} = (\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta) \), the ordinary and extraordinary eigen-polarizations for \( \hat{E} \) and \( \hat{D} \) are

\[ \hat{E}_o = (-\sin \phi, \cos \phi, 0) \quad \hat{D}_o = (-\sin \phi, \cos \phi, 0) \] \hspace{1cm} (2.35)

\[ \hat{E}_e = \left( \frac{\cos \phi \cos \theta}{n_o^2}, \frac{\sin \phi \cos \theta}{n_o^2}, \frac{-\sin \theta}{n_e^2} \right) \sqrt{\frac{\cos^2 \theta}{n_o} + \frac{\sin^2 \theta}{n_e^2}} \quad \hat{D}_e = (\cos \theta \cos \phi, \sin \phi \cos \theta, -\sin \theta) \]

On the optic axis, the degeneracy of the eigenvalues results in a plane of eigen-polarizations, instead of just two different eigen-polarizations. This is the same effect as seen in isotropic crystals where the degeneracy of the momentum surface results in a plane of eigen-polarizations. In isotropic crystals, however, this degeneracy occurs in every direction instead of just along the optic axis.

Figure 2.6 provides a comparison of momentum and real space for a negative uniaxial crystal. For the ordinary surface, \( \hat{S}_o \) and \( \hat{k}_o \) are always parallel as in the isotropic case. For the extraordinary
Figure 2.5: Momentum surface for a negative uniaxial media with the eigen-polarizations imposed on the surface \( (n_o = 1.9, n_e = 1.5) \). The ordinary surface (in blue) is sphere and the extraordinary surface (red) is an ellipsoid. The two surfaces touch and are rotational symmetric around the z-axis.

surface, on the other hand, \( \hat{S}_e \) and \( \hat{k}_e \) are not always parallel. In this case the optical power propagates away from the wave normal \( \hat{k} \) at angle \( \alpha \), which is know as the walk-off angle. The walk-off angle is calculated by

\[
\tan \alpha = \frac{n_e(\theta)}{2} \sin 2\theta \left( \frac{1}{n_e^2} + \frac{1}{n_o^2} \right).
\]

Figure 2.7a plots the walk-off angle \( \alpha \) as a function of the polar angle \( \theta \).

The field vectors for the ordinary wave are the same as for isotropic crystals, which is illustrated in Fig. 2.2. For the extraordinary wave, the electric field \( \hat{E} \) and \( \hat{D} \) are not parallel, and therefore neither are \( \hat{k} \) or \( \hat{S} \). Figure 2.7b shows the relative field orientations as determined by \( \hat{S} = \frac{1}{2}(\hat{E} \times \hat{H}) \) and Eqs. 2.29 and 2.30.

2.1.5 Biaxial Crystals

In biaxial crystals, \( \epsilon_1 \neq \epsilon_2 \neq \epsilon_3 \). The resulting momentum surfaces are no longer simple quadric surfaces, but are more complicated and cannot be defined by a simple equation as done
Figure 2.6: Momentum space versus real space in uniaxial media.

Optical Walk-Off

Figure 2.7: (a) Walk-off in a uniaxial crystal with $n_o = 1.9$ and $n_e = 1.5$. (b) Relative orientations of the wave normal $\hat{k}$, the Poynting vector $\hat{S}$, and the electromagnetic vectors $\hat{E}$, $\hat{D}$, $\hat{B}$, and $\hat{H}$ for the extraordinary wave.

with isotropic and uniaxial crystals. For biaxial crystals, there are two optical axes, hence the name ‘biaxial’. Research on biaxial crystals will not be presented in this thesis, so discussion on them will be limited.

Figure 2.8 illustrates a biaxial momentum surface for $n_x = 1.2$, $n_y = 1.5$, $n_z = 1.8$. The
eigenvalues and eigen-polarizations were calculated using Eq. 2.22. Also illustrated in the figure are the optic axes, which occur in the plane of the largest and smallest indices of refraction.

2.2 Acoustic Waves in Homogeneous Media

In the previous section, we saw that the coupling between the electromagnetic fields $\vec{E}$ and $\vec{H}$ leads to plane wave propagation given the correct orientation and propagation speed of the electromagnetic fields. Similarly for acoustic waves, the correct coupling of the strain $\vec{S}$ and stress $\vec{T}$ fields in an elastic medium will result in plane wave propagation. This section begins by first discussing the stress and strain fields along with their relation to each other. The solutions for acoustic plane wave propagation are then derived and the corresponding acoustic momentum surfaces are presented and discussed.

2.2.1 Stress and Strain Fields

The particle displacement field $u(\vec{r}, t)$ is a continuous variable that describes the particle displacement from equilibrium as a function of space and time. However, the particle displacement field $u(\vec{r}, t)$ fails to represent deformation because it is non-zero for uniform translations that leave
the material undeformed. Similarly, the displacement gradient tensor $Q_{ij}(\bar{r}, t) = \frac{\partial u_i(\bar{r}, t)}{\partial r_j}$ fails to represent material deformation because it is non-zero for rigid rotations [23]. The linearized strain tensor $S_{ij}$, on the other hand, is only non-zero when a material is deformed, and is defined as the symmetric part of the gradient tensor $Q_{ij}$:

$$S_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial r_j} + \frac{\partial u_j}{\partial r_i} \right). \quad (2.37)$$

The antisymmetric contribution to the displacement gradient matrix $Q_{ij}$ can be shown to be associated with local rotations that do not experience a restoring force. The $S_{11}$ component represents a plane wave propagating and polarized along $\hat{x}$. $S_{12}$ and $S_{21}$ represent a shear wave propagating in the $xy$-plane and polarized in a direction orthogonal to the propagation direction but in the $xy$-plane as well.

In continuous materials, stress is the traction force that neighboring particles exert on each other. Stresses in a medium are defined by the stress tensor $T_{ij}$, which assumes that the material is composed of volume elements of an orthogonal coordinate system (e.g. cubes or rectangular parallelepipeds). Each component of the stress tensor $T_{ij}$ represents the $r_i$ component of force per unit area between two parts of the medium separated by an imaginary plane perpendicular to the $r_j$ axis. The $T_{xx}$ component, for instance, represents a compressional force on an $yz$-plane in the $\hat{x}$ direction. The $T_{xy}$ component, on the other hand, represents a shear force on an $xz$-plane in the $\hat{x}$ direction.

In order to determine the total stress over the surface of an object of volume $V$ and surface $S$, all of the traction forces exerted on the object must be summed

$$T_{tot} = \int_S \bar{T} \cdot dS. \quad (2.38)$$

By applying the divergence theorem, Eq. 2.38 becomes

$$\int_S \bar{T} \cdot dS = \int_V \left( \nabla \cdot \bar{T} \right) dV. \quad (2.39)$$

Assuming no external forces, then Newton’s second law ($F = ma$) states that

$$\int_V \left( \nabla \cdot \bar{T} \right) dV = \int_V \rho \frac{\partial^2 \bar{u}}{\partial t^2} dV. \quad (2.40)$$
Setting the integrands equal to each other, we get

$$
\nabla \cdot \bar{T} = \rho \frac{\partial^2 \bar{u}}{\partial t^2},
$$

which is the translational equation of motion for a vibrating medium with no applied external forces.

The stress $S_{ij}$ and strain tensors $T_{ij}$ are related through Hook’e’s law, which states that there is a linear relationship between stress and strain:

$$
T_{ij} = C_{ijkl} S_{kl}
$$

Here $C_{ijkl}$ is the fourth-rank elastic-stiffness tensor and linearly relates the two second rank tensors $\bar{S}$ and $\bar{T}$. The stiffness coefficients, which are in units of $N/m^2$, measure a materials resistance to deformation.

Equation 2.42 contains nine equations with nine strain variables each, so the stiffness tensor has 81 coefficients. However, the symmetry of $S_{ij}$ and $T_{ij}$ allow for $C_{ijkl}$ to be reduced. The strain tensor $S_{ij}$ is symmetric ($S_{ij} = S_{ji}$) by definition (Eq. 2.37). The stress tensor $T_{ij}$ is symmetric as long as the solid is not subject to an applied torque [24], which will be assumed here. With $S_{ij}$ and $T_{ij}$ both being symmetric tensors, the first two or last two indices of the stiffness tensor can be interchanged without changing the value of the stiffness coefficient:

$$
C_{ijkl} = C_{jikl} = C_{ijlk}.
$$

This reduces the number of independent elastic stiffness coefficients from 81 to 36. With 36 coefficients, the elastic stiffness tensor may be written as a $6 \times 6$ matrix $C_{IJ}$, where the capital subscripts imply use of the reduce subscript notation. In addition, $S_{ij}$ and $T_{ij}$ can be written as six-element vectors because they are symmetric $3 \times 3$ matrices, which only have six independent coefficients. This abbreviated notation is known as Voight notation and can be convenient when writing out the stiffness tensor $C_{IJ}$ and trying to visualize the symmetry of the crystal [23, 25]. The stiffness tensor $C_{IJ}$ is required to be symmetric by conservation of energy [23, 24], which reduces the number
of independent elastic coefficients to 21. Symmetry of the medium further reduces the number of independent elastic coefficients. Only triclinic crystals require 21 independent coefficients.

2.2.2 Acoustic Plane Wave Solutions

With $S_{ij}$, $T_{ij}$, and their respective relationship defined, we can derive the solutions for acoustic waves. From the symmetry of $S_{ij}$, $T_{ij}$, and $C_{ijkl}$, Hooke’s law (Eq. 2.42) can be rewritten as

$$T_{ij} = C_{ijkl} \frac{\partial u_k}{\partial x_l}.$$  \hspace{1cm} (2.43)

Combining Eq. 2.41 with Eq. 2.43 produces the wave equation:

$$\rho \frac{\partial^2 u_j}{\partial t^2} = C_{ijkl} \frac{\partial^2 u_k}{\partial x_i \partial x_l}.$$  \hspace{1cm} (2.44)

As with optical waves, assume plane wave solutions will be assumed

$$\tilde{u}(\tilde{r}, t) = W \hat{U} e^{i(\hat{K} \cdot \tilde{r} - \Omega t)}$$  \hspace{1cm} (2.45)

Here $W$ is the amplitude of the acoustic wave, $\hat{U}$ is the polarization, $\Omega$ is the radian frequency, $K = 2\pi/\Lambda$ is the magnitude of the acoustic wave vector $\hat{K} = K \hat{m}$ with an acoustic wavelength $\Lambda$, and $m_j$ are the Cartesian components of the unit vector along the acoustic wave vector $\hat{K}$. This plane wave assumption reduces the time and spatial derivatives to multiplication factors of $-i\Omega$ and $i\hat{K}$; thus, the wave equation simplifies to the Christoffel equation

$$\rho V_a^2 U_i = C_{ijkl} m_j m_k U_l.$$  \hspace{1cm} (2.46)

Here $V_a = \Omega/K(\hat{m})$ is the acoustic velocity. Using the second-rank Christoffel tensor $\Gamma_{il} = C_{ijkl} m_j m_k$, the Christoffel equation reduces to

$$\Gamma_{il} U_l = \rho V_a^2 U_i.$$  \hspace{1cm} (2.47)

Equation 3.10 is a generalized eigenvalue problem of the matrix $\Gamma_{il}$. The eigenvalues $\rho V_a^2$ determine the velocity and the eigenvectors $U_i$ determine the polarization of an acoustic wave propagating in the $\hat{m}$ direction. The symmetry of the stiffness tensor $C_{ijkl}$ forces the Christoffel tensor $\Gamma_{il}$ to
be symmetric; therefore, its eigenvalues are real and its eigenvectors are orthogonal [24]. Figure 2.9 illustrates how the acoustic polarizations form an orthogonal triad of vectors. The polarization that is most parallel to the propagation direction $\hat{K}$ is referred to as the quasilongitudinal mode while the other two modes are referred to as quasishear modes.

In order for Eq. 3.10 to have a nontrivial solution, the determinant of the characteristic equation must vanish:

$$|\Gamma_{il} - \rho V_a^2 \delta_{il}| = 0. \quad (2.48)$$

Solving for the determinant of Eq. 2.48 results in a sixth-order equation, which reveals that there are three different solutions for a given propagation direction $\hat{m}$ and symmetric forward and backwards solutions [22]. There are three solutions because, unlike for optical waves, longitudinal waves are allowed.

The left column of Fig. 2.10 illustrates the coordinate plane cross-sections of the slowness surface ($1/V_a(\theta, \phi)$) for $\alpha$-BBO with the projection of the polarization vectors imposed on the surfaces. The magnitude and the polarization of the surfaces were solved for using Eq. 3.10. The quasilongitudinal mode is usually the fastest mode in the crystals, and is thus represented by the smaller of the three surfaces. The polarization for the longitudinal mode is nearly parallel with the propagation direction $\hat{m}$. The elliptical-shaped-shear mode in the YZ cross-section is a pure mode
with its polarization out of the plane in the \( \hat{x} \)-direction. The polarization vectors are not visible because the three-dimensional vectors are projected into the two-dimensional plane, so vectors that are perpendicular to the cross-section plane will not appear on the plot. The slow-shear mode for the XY-cross-section is not a pure mode, but its polarization is dominantly in the \( \hat{z} \)-direction.

The acoustic surfaces intersect along acoustic axes, which are very similar to optic axes. In the YZ-cross-section, there are four different acoustic axes. The acoustic axis along the \( \hat{z} \)-axis is required by the \( C_3 \) rotation symmetry about the \( z \)-axis, which requires that the crystal and all of its physical qualities (e.g. acoustic polarization) are left unchanged by \( 2\pi/3 \) rotations about the \( z \)-axis. As a consequence, the longitudinal mode must be polarized purely along the \( z \)-axis in order to remain unchanged from \( 2\pi/3 \) rotations. No single shear polarization can satisfy the \( C_3 \) symmetry, so the quasishear eigenvalues become degenerate to make the eigen-polarizations define a plane instead of a single vector. This example reflects the fact that anytime an axis possess \( C_n \) symmetry where \( n > 2 \), then the axis must be an acoustic axes and the longitudinal mode must be purely polarized along \( \hat{m} \). The remaining three acoustic axes in the YZ cross-section are not required by symmetry, so the longitudinal mode is not necessarily polarized along \( \hat{m} \) and the plane defined by the quasi-shear modes is not necessarily perpendicular to \( \hat{m} \).

We can easily convert the acoustic slowness surfaces calculated from the Christoffel equation to momentum space with a simple multiplication factor: \( K_a(\phi, \theta) = \frac{\Omega}{V_a(\phi, \theta)} \). As with optical momentum surfaces, the acoustic momentum surfaces contain an abundance of information. Figure 2.10 demonstrates the information contained in the acoustic momentum surfaces by comparing the fast and slow-shear modes in both real and momentum space. To begin with, the magnitude of the momentum surface determines the magnitude of the momentum vector \( K_a \) and the phase velocity \( V_a \) of the acoustic wave. Figure 2.11 illustrates how the slow-shear mode (blue) when compared to the fast shear mode (red), has a larger momentum vector \( \bar{K}_a \) that results in a shorter acoustic wavelength in real space.

The slope, or the first derivative, of the momentum surface determines the direction of the energy velocity vector \( \hat{V}_e \) and the acoustic Poynting vector. The right column of Fig. 2.10 illustrates
Figure 2.10: Acoustic slowness cross-sections with polarization (left column) and walk-off (right column) of $\alpha$-BBO. The black lines are orthogonal to the momentum surface and illustrate walk-off. The dashed gray lines are in the direction of $\bar{K}_a(\phi, \theta)$ and provide a reference for the black lines.
the direction of the energy velocity vector, which is orthogonal to the slowness surface. The energy velocity vector $\vec{V}_e$ determines the direction of the energy transport of the acoustic wave, and is calculated by [24]

$$V_i^e = \frac{C_{ijkl}U_j U_l \hat{K}_k}{\rho V_a}.$$ (2.49)

In Fig. 2.11, the momentum surface is rotated by $11^\circ$ from $K_z$ to a symmetry axis of the ellipse so that the first derivative of the slow-shear mode is zero, resulting in no acoustic walk-off in real space. The fast-shear mode, on the other hand, has a non-zero slope that results in a large acoustic walk-off in real space.

The energy flow of an acoustic beam is normal to the momentum surface. A beam of finite width $D$ in real space requires a bandwidth of transverse spatial frequencies in momentum space of about $\Delta k_t \approx \frac{2\pi}{D}$. The plane waves associated with the different transverse spatial frequencies will have different normal vectors to the momentum surface, or in other words different Poynting vectors, so the beam will diverge because the energy flow of the plane waves that compose the beam flow in different directions. The divergence of the beam can be quantified with the Rayleigh Range $Z_r$, which is the distance over which the cross-sectional area of the beam doubles [26]:

$$Z_r = \frac{D^2}{b\lambda}.$$ (2.50)

Here $b$ is the relative curvature to the isotropic curvature [22], and is calculated by

$$b = \frac{1 + 2(K'/K)^2 - (K''/K)}{[1 + (K'/K)^2]^{3/2}},$$ (2.51)

where the primes represent differentiation of $K(\theta)$ with respect to the angle. For isotropic medium, $b = 1$. For a perfectly collimated beam, the Rayleigh-range goes to infinity, requiring $b = 0$. This will only occur for a perfectly flat momentum surface.

Figure 2.11 illustrates the diffraction of the two shear modes in $\alpha$-BBO. The fast-shear mode has very little curvature in the direction of the acoustic propagation, which results in small acoustic diffraction in real space. In contrast, the slow-shear mode has large curvature, resulting in large acoustic diffraction in real space. The divergence of the beam can be mitigated by increasing the
Figure 2.11: Comparison of the fast (red) and slow-shear (red) modes in momentum and real space. The magnitude, slope, and curvature of the momentum surface determines in real space the acoustic wavelength, walk-off, and diffraction respectively.

size of the transducer $D$, which decreases the width of the transverse Fourier uncertainty $\Delta K_t = \frac{2\pi}{D}$.

2.3 Acousto-Optic Interaction

The previous two sections discussed the propagation of optical and acoustic plane waves in a given media. This section discusses how the optical and acoustic waves can interact and create acousto-optic diffraction. I begin by describing the elasto-optic effect in which the optical properties of a medium are changed from a propagating acoustic wave. This change in the material can cause optical diffraction. The section concludes by deriving the diffraction efficiency of an incident optical beam from a given acousto-optic interaction.

2.3.1 Elasto-Optic Effect

The elasto-optic effect describes the change in the optical properties of material through an applied mechanical stress. The elasto-optic effect was first observed by Sir David Brewster in 1815.
using jellies. In 1816 Brewster placed a strip of glass between two crossed polarizers. With the glass being optically isotropic, no light passed through the second polarizer. Brewster then stretched the glass, and light was then seen passing through the second polarizer. What Brewster observed was that the applied stress to the glass created an artificial birefringence that rotated the polarization of the incident light; thus, allowing for some of the light to pass through the second polarizer. Below I present the theory of the elasto-effect first presented by Pockels in 1889 and then completed by Nelson and Lax in 1971 [27, 28].

With the elasto-optic effect, we are interested in the change of the index of refraction \( n \) of a material. However, the index of refraction can not be represented as a tensor because it does not transform like a tensor, so the elasto-optic analysis must be conducted with either the relative permittivity tensor \( \epsilon_{ij} \) or the impermeability tensor \( \eta_{ij} \). The impermeability tensor \( \eta_{ij} \) is the inverse of \( \epsilon_{ij} \) \( (\epsilon_{ij}\eta_{jk} = I_{jk}) \). By determining the change to \( \epsilon_{ij} \) or \( \eta_{ij} \), the change to the index of refraction can be determined with Eq. 2.22.

In deriving the elasto-optic effect, Pockels assumed a linear relation to the change of \( \eta_{ij} \) with respect to an applied strain \( S_{kl} \):

\[
\Delta \eta_{ij} = p_{ijkl} S_{kl}.
\]  

(2.52)

Following Pockels notation, extensional strains are considered positive. The fourth-rank photoelastic tensor \( p_{ijkl} \) relates the nine components of \( S_{ij} \) to the nine components of \( \eta_{ij} \). Similar to the stiffness tensor \( C_{ijkl} \), \( p_{ijkl} \) can be simplified by noting that both \( S_{ij} \) and \( \eta_{ij} \) are symmetric matrices; therefore, \( p_{ijkl} = p_{jikl} = p_{ijlk} = p_{jilk} \). This reduces the number of independent photoelastic coefficients from 81 to 36, allowing Eq. 2.52 to be written in reduced subscript notation

\[
\Delta \eta_l = p_{l,j} S_j.
\]  

(2.53)

Unlike \( C_{IJ} \), \( p_{l,j} \) is not required to be symmetric \( (p_{l,j} \neq p_{j,l}) \), but the symmetry of the crystal will reduced the number of independent coefficients.

In Pockels’ derivation, he only considered the symmetric gradient of the displacement vector \( u_i \) as represented by the strain tensor \( S_{kl} \), and erroneously assumed that the contribution from the
antisymmetric gradient of the displacement $\omega_{ij}$ to be negligible. However, $\omega_{ij}$ is associated with local rotations that can cause a rotation of the principal axes of the medium. Such rotations in highly optically-anisotropic materials can lead to additional $\Delta \eta$ that contribute to AO diffraction. Nelson and Lax were the first to notice the contribution from $\omega_{ij}$ and measure the asymmetry of $p_{ijkl}$.

The general definition for the photoelastic effect is given by

$$\Delta \eta_{ij} = P_{ijkl} \frac{\partial u_k}{\partial x_l}. \quad (2.54)$$

Here $P_{ijkl}$ is the general photoelastic tensor and is only symmetric in $i$ and $j$ because $\eta_{ij}$ must remain symmetric. Both $\frac{\partial u_k}{\partial x_l}$ and $P_{ijkl}$ can be decomposed into their symmetric and antisymmetric tensors:

$$\frac{\partial u_k}{\partial x_l} = \frac{1}{2} \left( \frac{\partial u_k}{\partial x_l} + \frac{\partial u_l}{\partial x_k} \right) + \frac{1}{2} \left( \frac{\partial u_k}{\partial x_l} - \frac{\partial u_l}{\partial x_k} \right) = S_{kl} + \omega_{kl},$$

$$P_{ijkl} = \text{Sym}(P_{ijkl}) + \text{Antisym}(P_{ijkl}) = p_{ijkl} + P_{ij(kl)}. \quad (2.55)$$

Noting that symmetric and antisymmetric matrices are orthogonal with respect to each other ($\text{Sym}(A) \cdot \text{Antisym}(B) = 0$), Eq. 2.54 reduces to

$$\Delta \eta_{ij} = p_{ijkl} S_{kl} + P_{ij(kl)} \omega_{kl}. \quad (2.56)$$

Explicit definitions for $P_{ij(kl)}$ can be determined when the orientation of the principle dielectric axis are fixed, which excludes triclinic and monoclinic crystals. Nelson and Lax showed this explicit expression to be

$$P_{ij(kl)} = -\frac{1}{2} \left( \frac{1}{n_i^2} - \frac{1}{n_j^2} \right) (\delta_{ik} \delta_{jl} - \delta_{jk}). \quad (2.57)$$

The above equation reveals that the antisymmetric part of the photoelastic tensor $P_{ijkl}$ is zero for optically isotropic materials. For most crystals, the Nelson and Lax correction is so small that it can be safely ignored, but for crystals with a large birefringence and small index of refraction, like in BBO, the Nelson and Lax correction can not be neglected.
After determining the change to the impermeability tensor $\eta_{ij}$ with Eq. 2.56, the change to the index of refraction can be determined either explicitly with Eq. 2.22 or approximately by

$$\Delta \eta = \Delta \left( \frac{1}{n^2} \right) = p_{ijkl}S_{kl} + P_{ij(kl)}\omega_{kl},$$

$$-\Delta n - \frac{2}{n^3} = p_{ijkl}S_{kl} + P_{ij(kl)}\omega_{kl},$$

$$\Delta n = -\frac{1}{2}n^3(p_{ijkl}S_{kl} + P_{ij(kl)}\omega_{kl}).$$

(2.58)

The change to the permittivity tensor $\Delta \bar{\epsilon}$ can be determined by first noting that

$$\bar{\epsilon} \bar{\eta} = \bar{I}$$

(2.59)

Differentiating the above relation generates

$$\Delta \bar{\epsilon} \bar{\eta} + \bar{\epsilon} \Delta \bar{\eta} = 0$$

(2.60)

Multiplying by $\bar{\eta}$ and inserting both Eqs. 2.56 and 2.59, we have

$$\Delta \bar{\epsilon} = -\bar{\epsilon}(\bar{p}\bar{S} + \bar{P}\bar{\omega})\bar{\epsilon}.$$  

(2.61)

Regardless if we think of $\Delta \bar{\eta}$, $\Delta n$, or $\Delta \bar{\epsilon}$, the important point to note is that strain changes the optical properties of the crystal. Hence, a traveling acoustic plane wave will create sinusoidally varying strain, which in turn creates a moving anisotropic dielectric phase grating in the medium that can diffract incident optical light.

### 2.4 Acousto-Optic Interaction

There are numerous derivations that describe the diffraction of light through a dielectric perturbation. Most derivations are either a coupled-wave or modal analysis [29, 30]. Here we present a Fourier analysis using a derivation first presented by [31]. This Fourier analysis assumes that the optical diffraction is so weak that the incident optical field remains undepleted (Born approximation); therefore, it is not accurate in the large diffraction efficiency regime. However, the Fourier analysis provides a simple pictorial description of the acousto-optic interaction that facilitates in designing and evaluating acousto-optic devices.
We begin the derivation with two coupled waves with a dielectric perturbation \( \bar{\varepsilon}(\vec{r}, t) \):

\[
\nabla^2 [E_i(\vec{r}, t) + E_d(\vec{r}, t)] + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \left[ \bar{\varepsilon} + \bar{\varepsilon}(\vec{r}, t) \right] [E_i(\vec{r}, t) + E_d(\vec{r}, t)] = 0. \tag{2.62}
\]

Here \( E_i \) and \( E_d \) are the incident and diffracted electric fields and \( c \) is the speed of light. We separate the above equation into coupled equations for the incident field of frequency \( \omega \) and each monochromatic angular frequency component \( \omega_m \) of the diffracted field:

\[
\nabla^2 E_i(\vec{r}, \omega) + \frac{\omega^2 \varepsilon}{c^2} E_i(\vec{r}, \omega) = -\frac{\omega^2}{c^2} = \sum_m \bar{\varepsilon}(\vec{r}, \Omega_m) E_d(\vec{r}, \omega_m),
\]

\[
\nabla^2 E_d(\vec{r}, \omega_m) + \frac{\omega^2_m \varepsilon}{c^2} E_d(\vec{r}, \omega_m) = \frac{-\omega^2_m}{c^2} \bar{\varepsilon}(\vec{r}, \Omega_m) E_i(\vec{r}, \omega). \tag{2.63}
\]

Here \( \bar{\varepsilon}(\vec{r}, \Omega_m) \) represents the dielectric perturbation from an acoustic wave with an angular frequency \( \Omega_m \). For Eq. 2.63 to be true fall all time, then \( \omega_m = \omega + \Omega_m \), which is required by conservation of energy. The conservation of energy requirement can be simplified by noting that \( \Omega_m \ll \omega \), so \( \omega_m \approx \omega \).

We now Fourier expand the diffracted field \( E_d \) into plane-wave eigenmodes of the unperturbed medium that evolve along the \( z \)-direction:

\[
E_d(\vec{r}, \omega_m) = \int \mathcal{E}_d^{\omega_m}(\vec{k}_t, z) e^{i k_{zd} \cdot \vec{k}_t} e^{i k_{zd}(\vec{k}_t)z} d\vec{k}_t. \tag{2.64}
\]

Here \( k_{zd}(\vec{k}_t) = \sqrt{k_0^2 - k_t \cdot \vec{k}_t} \) is the longitudinal component of the wavevector, \( \vec{k}_t = k_{tx} \hat{x} + k_{ty} \hat{y} \) is the transverse component of the wavevector, and \( k_d = 2\pi n_d(\vec{k}_t, \omega_m) / c \) is the magnitude of the diffracted wavevector. Substituting Eq. 2.64 into Eq. 2.63 results in the following equation:

\[
\left[ 2ik_{zd}(\vec{k}_t) \frac{\partial}{\partial z} \mathcal{E}_d^{\omega_m}(\vec{k}_t, z) + \frac{\partial^2}{\partial z^2} \mathcal{E}_d^{\omega_m}(\vec{k}_t, z) \right] e^{i k_{zd}(\vec{k}_t)z} d\vec{k}_t = \frac{-\omega^2_m}{c^2} \bar{\varepsilon}(\vec{r}, \Omega_m) E_i(\vec{r}, \omega). \tag{2.65}
\]

We can simplify the above equation by applying the slowly varying envelope approximation (SVEA) \( k_{zd} \frac{\partial}{\partial z} \mathcal{E}_d^{\omega_m}(\vec{k}_t, z) \gg \frac{\partial^2}{\partial z^2} \mathcal{E}_d^{\omega_m}(\vec{k}_t, z) \), giving

\[
\int \int 2ik_{zd}(\vec{k}_t) e^{ik_{zd}(\vec{k}_t)z} \frac{\partial}{\partial z} \mathcal{E}_d^{\omega_m}(\vec{k}_t, z) e^{i(xk_x + yk_y)} dk_x dk_y = \frac{-\omega^2_m}{c^2} \bar{\varepsilon}(\vec{r}, \Omega_m) E_i(\vec{r}, \omega). \tag{2.66}
\]

Applying the SVEA reduces Eq. 2.66 from a second order to a first order equation, which reduces the number of solutions and the required boundary conditions from two to one. Next we take the
transverse Fourier transform in $x$ and $y$ to get

$$e^{ikzd(k_x,k_y)z} \frac{\partial}{\partial z} \mathcal{E}_d^{\omega m}(k_x, k_y, z) = \frac{i\omega_m^2}{2c^2 kzd(k_x,k_y)} \int \int \tilde{\delta}(\bar{r}, \Omega_m) E_i(\bar{r}, \omega) e^{-i(xk_x+yk_y)} dx dy.$$  \hspace{1cm} (2.67)

If we apply the Born approximation and assume that the diffraction is so weak that both the incident electric field $E_i$ and the dielectric perturbation $\delta \varepsilon$ do not lose power during the interaction, then we can directly integrate the above equation, which yields the field of the diffracted wave $E_d$ at the exit face

$$\mathcal{E}_d^{\omega m}(k_x, k_y, L) = \frac{i\omega_m^2}{2c^2 kzd(k_x,k_y)} \int_0^L FT_{xy} \left\{ \delta\tilde{\varepsilon}(\bar{r}, \Omega_m) E_i(\bar{r}, \omega) \right\} e^{-ikzd(k_x,k_y)z} dz.$$  \hspace{1cm} (2.68)

Here $L$ is the length of the interaction. This last integral can be reformulated as a 3D Fourier transform by noting that $\delta \varepsilon$ term vanishes outside of the interaction length of the acoustic column ($z \in \{0, L\}$). This allows us to extend the limits of the $z$ integration to infinity, and therefore substitute a three-dimensional Fourier transform into Eq.2.68:

$$\mathcal{E}_d^{\omega m}(k_x, k_y, L) = \frac{i\omega_m^2}{2c^2 kzd(k_x,k_y)} \int \delta(k_z - kzd(k_x,k_y)) FT_{xyz} \left\{ \delta\tilde{\varepsilon}(\bar{r}, \Omega_m) E_i(\bar{r}, \omega) \right\} dk_z,$$

$$= \frac{i\omega_m^2}{2c^2 kzd(k_x,k_y)} \int \delta(k_z - kzd(k_x,k_y)) \left( \tilde{\delta}(\bar{K}, \Omega_m) \ast \mathcal{E}_i(\bar{k}, \omega) \right) dk_z \hspace{1cm} (2.69)$$

This equation states that the angular spectrum of the diffracted field equals the convolution of the Fourier transform of the incident optical field with the Fourier transform of the dielectric perturbation; however, the $\delta$ function requires that the convolution is an allowed eigenmode of the light in the unperturbed medium.

Figure 2.12 provides an illustrative example of Eq. 2.69 for optical isotropic diffraction from an acoustic wave. In this two-dimensional example, an incident beam with an uniform intensity and optical aperture of width $A$ is diffracted by an acoustic beam with an uniform intensity and width $L$. In this example, the convolution of the Fourier transform of the incident optical beam with the Fourier transform of the acoustic beam perfectly resides on the optical momentum surface; therefore, the diffraction efficiency is maximized. If the angle of the incident beam or the frequency of the acoustic beam were changed, then the convolution would be displaced causing a decrease in the diffraction efficiency.
Figure 2.12: Comparison of AO diffraction in momentum and real space.
Chapter 3

Measuring Elastic Stiffness Coefficients

The elastic-stiffness coefficients are a fundamental property of a material. As the second derivatives of the free energy with respect to strain, they are closely related to the thermal properties through the Debye theory [32]. Of more importance to the contents of this thesis, the stiffness coefficients are critical in designing acousto-optic devices because they are vital in determining the diffraction efficiency and the velocity, walk-off, and diffraction of the acoustic wave. Hence, the complete and accurate determination of the elastic-stiffness coefficients of a material is required for designing acousto-optic devices.

I employed both Schaefer–Bergmann diffraction (SBD) and resonant ultrasound spectroscopy (RUS) to measure the elastic-stiffness coefficients of both $\alpha$ and $\beta$-BBO. The elastic-stiffness coefficients for both $\alpha$ and $\beta$ were previously measured using pulse-echo experiments [14, 17], but the pulse-echo experiment does not accurately measure off-diagonal elastic coefficients and is susceptible to large errors due to acoustic walk-off. Thus, I improved the accuracy of the measured stiffness coefficients by using both SBD and RUS measurements.

3.1 Measurements

In the SBD measurement, a transparent specimen is filled with acoustic plane waves of various polarizations and propagation directions at a monochromatic RF frequency. A collimated laser beam propagates through the crystal and interacts with the spectrally rich acoustic field. Only the acoustic plane-wave components that satisfy the Bragg-matching condition produce a
diffracted optical beam. A Fourier transform lens maps the resulting SBD onto a camera, giving a cross-section of the acoustic slowness surface [33]. Previous SBD experiments calculated the elastic-stiffness coefficients by solving a set of linear equations using measurement points along the SBD pattern that determine the acoustic velocities for a certain propagation direction [34, 35, 36, 37]. I present an alternative method in which I use analytic solutions for the acoustic slowness-surface cross-sections to fit to an image of the entire measured SBD pattern, including the strong speckled diffraction peaks and the weak envelope outlining the entire contour. The elastic coefficients are calculated by iteratively adjusting the elastic coefficients until the overlap between the measured and calculated cross-sections is maximized.

RUS has become the standard technique for measuring elastic stiffness coefficients with accuracies approaching 0.1% [38]. RUS determines the elastic coefficients of a cut and polished specimen by exciting and measuring 30-100 of the lowest order resonant modes in the kHz-MHz range. The sample can be of any shape and orientation with dimensions from a few hundred microns to several centimeters [39]. To determine the elastic coefficients, the resonance spectrum is calculated from the current estimate of the elastic coefficients along with the dimensions, density, and geometry of the sample. The elastic coefficients are then refined by varying the coefficients until the error between the calculated and measured resonance spectra is minimized.

RUS and my new approach to SBD have intriguing similarities. In both measurements, the elastic coefficients are determined by measuring the resonant elastic modes of the sample, and then fitting to the eigenvalues of a Lagrangian. For the SBD measurement, the specimen is assumed to be so large that the solutions to the elasto-dynamic Lagrangian are acoustic plane waves and the eigenvalues and eigenvectors, solved for using the acoustic Christoffel equation, determine the velocities and polarizations of the plane waves. The specific crystal cut and the propagation direction of the incident optical beam determine the acoustic slowness-surface cross-section that is measured by acousto-optic diffraction. When AO diffraction angles are measured at just a few angles, the linear system giving the elastic coefficients can be solved with a matrix inverse. I, however, measure the entire multi-sheeted slowness surface cross-section (the SBD pattern) and
iteratively refine the values of the elastic coefficients with an optimization algorithm until the measured and calculated eigenvalues agree. For the RUS measurement, the Lagrangian accounts for the finite size of the specimen so that the eigenvalues and eigenvectors of the Lagrangian represent the resonant frequencies, or modes, and resonant deformations of the sample. These resonances are determined by the elastic constants, and also by the shape, orientation, and size of the crystal. As with the SBD measurement, the elastic coefficients can not be calculated directly from the measured eigenvalues (resonances), but are instead determined by an optimization algorithm. In the next few sections, I describe, compare, and contrast the two measurements in detail to make the experimental and computational similarities more apparent.

3.2 Schaefer-Bergmann Diffraction

Clemens Schaefer and Ludwig Bergman first observed SBD in 1934, an impressive 26 years before the invention of the laser [40]. In 1935 E. Fues and H. Ludloff determined that the SBD patterns are the result of light diffraction from many different elastic waves propagating orthogonally to the incident light. They discovered that the three different surfaces are from the three different acoustic modes in the crystal, and that the shape of the SBD patterns are determined by the velocity of the acoustic wave as a function of propagation direction [41]. Fues and Ludloff’s

Figure 3.1: Measured SBD of (a) fused silica, (b) $\alpha$-BBO, and (c) $\beta$-BBO.
analysis allowed for experimentalists to measure the elastic-stiffness coefficients of a sample using SBD patterns [36]. With the invention of the laser, the SBD experiment attracted more attention from researchers [42, 43], but never achieved the popularity of pulse-echo in the 1960s-1980s or RUS in the last thirty years.

3.2.1 SBD Experiment

In my SBD experiment, as illustrated in Fig. 3.2, I use the crystallite phenyl benzoate, which melts at 67° C, to temporarily bond the α-BBO specimen to a fused silica reference cell that has an attached piezo-electric transducer that operates in the 10-50 MHz range. I bond the α-BBO and fused silica crystals by heating up the crystals above 67° C with a thin layer of melted phenyl benzoate between the two crystals. I allow the crystals, while pressed together, to cool to room temperature so that the phenyl benzoate will recrystallize and temporarily bond the two crystals. This bond is easily removed by reheating the specimen. With this bonding scheme and using both a shear and longitudinal transducer, I can launch all three polarizations of the acoustic waves along any of the X, Y, or Z axis.

With the α-BBO and fused silica crystals bonded, I launch acoustic waves into the fused silica using a shear transducer. The acoustic impedance mismatch between the two media and the phenyl bond leads to partial transmission of the acoustic wave into the α-BBO with partial reflection back into the fused silica. Surface scratches on the polished surfaces and inhomogeneities of the recrystallized bond layer scatters some of the transmitted acoustic waves into a variety of directions in the α-BBO and can even mode convert the acoustic wave into other acoustic polarizations. After several additional reflections and scatterings off of edges, faces, and corners within the α-BBO, the crystal is filled with plane-wave components with a variety of polarizations and propagation directions. The resonating field at these RF frequencies could be described in real space throughout the crystal, as is done in RUS, but expanding the resonant field in acoustic plane-wave components that satisfy the Christoffel equation is much better for describing the Bragg-matched acousto-optic diffraction. Although these acoustic waves attenuate with propagation, I launch the acoustic waves
at such low frequencies (10-50 MHz) that the acoustic waves have a chance to bounce around many times before attenuating; thus, creating a diffuse, spectrally rich resonating acoustic field containing nearly all plane-wave modes. Some modes, however, are much stronger because of resonant conditions off the crystal faces, edges, and corners, and because the transducer launches the acoustic power primarily in the direction perpendicular to the transducer. The uniformity of the distribution of the acoustic power could be improved by placing the transducer at the corner of the crystal or by using a smaller or non-rectangular transducer to encourage acoustic diffraction in the crystal [44].

I pass a collimated 532 nm optical beam with either ordinary or extraordinary eigenpolarization through an orthogonal face of the $\alpha$-BBO crystal to interact with the diffuse, spectrally rich resonating acoustic field excited in the crystal. The individual acoustic plane-wave components acousto-optically diffract the incident optical wave, but only the Bragg-matched acoustic plane-wave components that conserve momentum produce substantial diffraction. The power of a particular acoustic plane-wave component and the effective photo-elastic coefficient determine the magnitude of the diffracted optical wave. A Fourier transform lens maps the resulting angles of propagation of the diffracted light onto a camera to record the SBD pattern [34, 44], as illustrated in Fig. 3.2.

I determine the elastic coefficients by comparing the measured SBD patterns to the analytic cross-sections, which I calculate using Eq. 3.10 to solve for $V_a^{-1}$ and then multiply by $2\pi f_a$ to convert to momentum space. I convert the measured SBD patterns to momentum space by applying an appropriate scaling factor $S$. For a given acoustic wavevector $K_a$ and assuming small diffraction angle $\theta_m$, the light will be diffracted at an angle

$$\theta_m \approx \frac{K_a}{k_0 n_m}. \quad (3.1)$$

Here $n_m$ is the index of refraction of the medium. Upon exiting the crystal, the light will diffract according to Snell’s law, where once again the small angle approximation will be used:

$$n_a \theta_a = n_m \theta_m. \quad (3.2)$$
Figure 3.2: (a) Illustration of the SBD experiment. A piezoelectric transducer launches a monochromatic acoustic wave into the fused silica crystal. The acoustic wave refracts, reflects, and scatters between the fused silica and BBO crystal creating a diffuse spectrally-rich resonating acoustic field. An incident, collimated optical beam interacts with the acoustic field and is diffracted by various acoustic plane-waves. A Fourier transform lens maps the diffraction onto a CCD camera; thus, recording the SBD. A beam block, not illustrated, blocks the undiffracted light to prevent the CCD camera from saturating. (b) Flowchart of SBD optimization algorithm.

Here $\theta_a$ and $n_a$ are the diffraction angle and the index of refraction of free space, respectively. Combing Eqs. 3.1 and 3.2 and setting $n_a = 1$, we find that $\theta_a = \frac{K_a}{k_o}$. This diffraction angle $\theta_a$ will lead to a displacement $R$ in the Fourier plane of the lens from the DC spot of

$$R = \frac{K_a}{k_o} F. \tag{3.3}$$

Here $F$ is the focal length of the Fourier transform lens. I measure the SBD with a CCD camera that has pixel separation $\Delta p$, so distances in the recorded SBD are measured in number of pixels $N_p$ where

$$N_p = \frac{R}{\Delta p}. \tag{3.4}$$

Combing Eq. 3.3 with Eq. 3.4, the scaling factor $S$ to convert from pixel space to momentum space becomes

$$S = \frac{K_a}{N_p} = \frac{k_o \Delta p}{F} = \frac{2\pi \Delta p}{\lambda F}. \tag{3.5}$$

This shows that the size of the measured SBD on the CCD camera is proportional to both $\lambda$ and
$F$, but not the index of refraction of the medium. Therefore, the index of refraction of the medium is not required for SBD measurements as long as the small angle approximation for the diffracted light is accurate.

I refine the scaling factor $S$ by replacing the $\alpha$-BBO sample with a 300 line/in Ronchi grating and record the resulting diffraction pattern with the CCD camera. I measure the location of each diffraction order by fitting each spot to a Gaussian distribution to get sub-pixel accuracy. The diffraction grating has a transverse momentum wavevector of $K_g = \frac{2\pi}{\Lambda_g}$, where $\Lambda_g$ is the separation of the grating lines. By measuring the separation of the various diffraction orders and knowing $K_g$, I can experimentally determine the scaling factor $S$. For my SBD experiments, I use a lens with a focal length of $F = 750$ mm and a MQ042RG-CM Ximea camera with a pixel separation of $\Delta p = 5.5$ $\mu$m. I also use a beam expander with a magnification of 0.4 to scale the SBD so that it would fit onto the camera. With this experimental set-up, the scaling factor should be $S = 217 \frac{m^{-1}}{\text{pixels}}$. With the Ronchi grating, I measured a 1.2% difference with $S = 214 \frac{m^{-1}}{\text{pixels}}$.

Similar to RUS, the elastic coefficients cannot be determined directly from a measured SBD, but the SBD patterns can be calculated from the elastic coefficients. I determine the elastic coefficients of trigonal crystals by iteratively refining the elastic coefficients until the overlap of the measured and calculated SBD patterns reach a maximum, as shown in Fig. 3.2b. The overlap is calculated by integrating the square root of the measured SBD pattern weighted by a calculated slowness surface. I use the square root of the SBD to prevent the optimization algorithm from solely focusing on a few bright spots, and instead focus on the entire SBD pattern. By taking the square root, the optimizer is less likely to converge to a local minimum. If I take the log of the measured SBD, the optimizer is less sensitive to the difference from the signal and the speckled noise. I give the calculated SBD pattern a Gaussian width of about two pixels to represent the finite size of the focused beam on the detector and to help the optimizer search for the maximum overlap. In addition to optimizing the stiffness coefficients, I optimize the location and rotation of the calculated SBD patterns to maximize the overlap with the measured SBD patterns.
3.2.2 SBD Computation

To understand the measured SBD patterns, I begin by solving for the eigenvalues and eigenvectors of an infinitely large sample. Centimeter sized samples can be approximated as being infinity large because the acoustic wavelengths are very small by comparison (between 10-100 µm).

The kinetic and potential energies for a uniform crystal with no applied external forces are

\[ KE = \frac{\rho}{2} \int V \left( \frac{\partial u_i}{\partial t} \cdot \frac{\partial u_i}{\partial t} \right) dV, \quad PE = \frac{1}{2} \int V C_{ijkl} \frac{\partial u_i}{\partial x_j} \frac{\partial u_k}{\partial x_l} dV. \] (3.6)

Here \( \rho \) is the material density, \( u_i(\vec{r}, t) \) is the displacement vector field, \( C_{ijkl} \) is the elastic stiffness tensor, and \( V \) is the volume of the specimen. Taking the difference of the kinetic and potential energies creates the following Lagrangian

\[ L = \frac{1}{2} \int dt \int V \left[ \left( \rho \frac{\partial u_i}{\partial t} \cdot \frac{\partial u_i}{\partial t} \right) - C_{ijkl} \frac{\partial u_i}{\partial x_j} \frac{\partial u_k}{\partial x_l} \right] dV. \] (3.7)

Applying the Euler-Lagrange equation [45] to Eq. 3.7 and assuming that \( \rho \) and \( C_{ijkl} \) are independent of time and space results in the spatio-temporal anisotropic acoustic wave equation:

\[ \rho \frac{\partial^2 u_i}{\partial t^2} - C_{ijkl} \frac{\partial^2 u_k}{\partial x_j \partial x_l} = 0. \] (3.8)

For an infinitely large crystal, the eigensolutions are displacement-vector plane-waves

\[ u_k = U_k e^{i(\omega t - K \cdot \vec{m})}, \] (3.9)

where \( U_k \) is the unit polarization vector, \( K \) is the length of acoustic wavevector \( \vec{K}_a = K \hat{m} \), and \( m_j \) are the Cartesian components of the unit vector along \( \vec{K}_a \). Substituting Eq. 3.9 into Eq. 3.8, yields the Christoffel equation from Chapter 2:

\[ [C_{ijkl} m_j m_l - \rho V_a^2 \delta_{ik}] U_k = 0. \] (3.10)

Here \( V_a = \omega / K^p(\hat{m}) \) is the phase velocity of the acoustic wave in the direction \( \hat{m} \) for one of the \( p = 3 \) polarizations. Solving the Christoffel equation for a given direction \( \hat{m} \) results in three eigenvalues and eigenvectors that represent the phase velocities \( V_a^p \) and corresponding eigenpolarizations \( \hat{U}^p \) of
three different acoustic waves. Figure 3.3a illustrates the triple-sheeted slowness surface, which is a plot of the inverse velocity of the acoustic waves $V_a^{-1}$, of $\alpha$-BBO by solving the Christoffel equation as a function of the acoustic wavevector direction $\vec{m}$ and using the elastic coefficients determined from SBD.

For an acoustic wave to efficiently diffract light, the effective photoelastic coefficient must be nonzero and the energy transfer from the input optical wave to the diffracted wave must be synchronous in phase, which is commonly known as Bragg-matching. This will occur when the
acoustic and optical momentum vectors sum to an allowed optical propagation mode

$$|\vec{k}_d| = \frac{2\pi n_o}{\lambda} = |\vec{k}_i \pm \vec{K}_a|,$$

(for ordinary diffracted wave)

$$\left|\left(\frac{1}{n_e}, \frac{1}{n_e}, \frac{1}{n_a}\right) \cdot \vec{k}_d\right| = \frac{2\pi}{\lambda} = |\vec{k}_i \pm \vec{K}_a|/n_e(\theta),$$

(for extraordinary diffracted wave) (3.11)

Here $\vec{k}_d$ and $\vec{k}_i$ denote the diffracted and incident optical wavevectors, respectively. The Bragg-matched intersections for a specific incident plane-wave in momentum space are solved for by intersecting the double-sheeted optical and triple-sheeted acoustic momentum surfaces. For a negative uniaxial crystal like $\alpha$-BBO, the optical momentum surface is an ellipsoid inside a sphere with the two surfaces touching along the z-axis and scaled by the frequency-dependent indices of refraction $n_e(\omega)$ and $n_o(\omega)$ and the vacuum optical wavelength $\lambda_o (k = \frac{2\pi n(\theta)}{\lambda_o})$. The corresponding ordinary and extraordinary eigenpolarizations lie along the latitudes on the sphere and the longitudes of ellipsoid, respectively. The acoustic wavevector $\vec{K}_a(\phi, \theta) = \frac{2\pi}{V_a(\phi, \theta)} f_a$ is proportional to the applied acoustic frequency $f_a$, which allows for the size of the acoustic momentum surface to be controlled. Also, the acoustic wavevector is proportional to the acoustic slowness $V_a^{-1}[\frac{\mu_s}{mm}]$, so the acoustic slowness surface can be converted to momentum space $[m^{-1}]$ by multiplying by the angular frequency $\Omega = 2\pi f_a$ [rad/s].

Figure 3.3b shows in momentum space the SBD geometry for the case when the incident optical light is propagating along the z-axis, and the acoustic frequency is set to $f_a = 1 \text{ GHz}$ to exaggerate the size of the acoustic-momentum surface for viewing purposes. The acoustic-momentum surface is centered on the incident optical wavevector $\vec{k}_i$ to illustrate the summation of $\vec{k}_i$ with every possible acoustic wavevector. The intersection of the optical and acoustic-momentum surfaces represent the diffracted wavevectors that satisfy Bragg-matching (Eq. 3.11). The acoustic momentum surface in Fig. 3.3b represents acoustic waves propagating in every possible direction in the crystal. For each acoustic propagation direction, the acoustic wave can have three different possible acoustic polarizations, which is represented by the three different acoustic surfaces in Fig. 3.3a. Each acoustic wave induces an elasto-optic grating in the direction of its propagation, and the wavelength $\Lambda$ of the grating is proportional to the magnitude of the diffraction angle and inversely
proportional to the acoustic velocity $V_a (A = V_a/f_a)$. Therefore, slower acoustic waves diffract the optical light at larger angles. The resulting diffraction pattern represents the speed, wavelength, and propagation direction of the various elasto-optic diffraction gratings (acoustic waves) in the crystal. Figure 3.3b illustrates that only the acoustic waves propagating essentially perpendicularly to the incident light will be Bragg matched and therefore diffract the light.

For my SBD calculations, I assume that the acoustic-momentum surface is so small compared to the optical-momentum surface that I ignore the curvature of the optical-momentum surface and approximate the intersection of the momentum surfaces as a planar cross-section, perpendicular to $\vec{k}_i$, of the acoustic momentum surface. For instance, incident light propagating along the z-axis would measure the XY cross-section of the acoustic momentum surface. Therefore, the SBD represents a cross-section of the acoustic-momentum surface, which is proportional to the acoustic-slowness surface.

Ignoring the curvature of the optical-momentum surface in the SBD calculations causes some errors. Figure 3.3d shows in solid lines the analytic solution to the XY cross-section, and the pink dots illustrate what the measured SBD pattern after appropriate scaling would be in momentum space for $f_a = 1$ GHz. The resulting errors from ignoring the curvature of the optical-momentum surfaces are shown in Fig. 3.3e. The errors are larger for the slower acoustic waves because approximating the optical momentum surfaces as planes becomes less accurate for larger acoustic momentum vectors $\vec{K}_a$. Figure 3.3f shows how the RMS error from ignoring the acoustic curvature of the slow-shear acoustic wave for the three different cross-sections depends on the applied acoustic frequency $f_a$. For my measurements I use acoustic frequencies of about $\sim 30$ MHz so that the RMS percent error would not exceed 0.25\%.

3.2.3 Fused Silica Measurements

With the experimental set-up (illustrated in Fig. 3.2), I am able to measure SBD from the reference crystal fused silica and any crystal bonded to the fused silica. When measuring the fused silica SBD, a crystal needs to be bonded to the reference cell to scatter the acoustic waves. Without
the second cell accompanied with the required bonding layer, the acoustic waves predominately stay in the acoustic mode launched by the transducer. The resulting SBD pattern is just two bright spots and nearly nothing else.

Fused silica is an isotropic crystal, which means that there is no way to distinguish two different directions in the crystal from the crystal properties. As a consequence, the acoustic slowness-surface is two concentric spheres. The inner sphere represents the pure longitudinal mode and the outer sphere represents the degenerate, pure shear modes. If the slowness surfaces did demonstrate any asymmetry or the shear modes were not degenerate, then different directions in the crystal would be distinguishable and the crystal would no longer be isotropic.

Figure 3.4 illustrates the SBD and corresponding fit for fused silica. Fused silica has only two independent elastic-stiffness coefficients: 1) $C_{11}$ that determines the radius of the longitudinal mode and 2) $C_{44}$ that determines the radius of the shear modes for the acoustic slowness surface. Imeasured $C_{11}$ and $C_{44}$ for fused silica to be 78.5 and 31.3 GPa, which is within a 0.4\% agreement of the accepted values. This simple measurement tests the calibration accuracy and validity of my experimental and computational analysis, and could be used to recalibrate subsequent measurements
3.2.4 SBD $\alpha$-Barium Borate Measurements

With my SBD measurement, I measured the SBD patterns of the XY, XZ, and YZ cross-sections. The top row of Fig. 3.6 shows images of SBD of $\alpha$-BBO from $\hat{z}$, $\hat{y}$, and $\hat{x}$-propagating
optical beams (532 nm). To prevent camera saturation, I blocked the optical DC spot in the middle of the diffraction pattern in an intermediate Fourier plane, making the diffraction pattern more visible. The optical power is not evenly distributed because the acoustic power and effective photoelastic coefficient vary for each direction. Also, the random nature of the resonating modes of the various acoustic waves causes a random, speckled intensity in the SBD pattern, but the outlines are clear especially for the YZ cross-section. Missing data do not prevent the optimizer from converging, but will increase the error bars.

The bottom row of Fig. 3.6 illustrates the experimental orientation of the crystals and the polarization of the transducer for each cross-section. For the XZ and YZ cross-sections, I launched the acoustic wave along the z-axis. I chose to propagate the acoustics along Z because the effective photoelastic coefficient is equal to zero for this geometry in trigonal crystals, preventing camera saturation from the diffracted light. In my SBD experiments, the acoustic mode launched by the transducer contains so much of the acoustic power that the light diffracted by this mode can be significantly brighter than the other acoustic modes, as seen in the XY cross-section. Camera saturation broadens the SBD pattern, making the optimization algorithm less sensitive to change in the shape of the calculated SBD pattern. By launching the acoustics along the z-axis for the XZ and the YZ cross-sections, I can minimize the deleterious effects of camera saturation. The z-axis is also an acoustic axis so the acoustic diffraction is large, which helps populate other acoustic modes. For the XZ cross-section, I set the transducer polarization along the z-axis to put more acoustic power into the quasi-y-polarized shear mode, which has a relatively small effective photoelastic coefficient. By doing so, I was able to see all three acoustic modes. For the YZ cross-section, I set the polarization along the y-axis because the x-polarized mode is invisible for this cross-section, so any z-polarized acoustic power would be wasted. For the XY cross-section, I launched a z-polarized acoustic mode to improve the visibility of the slow quasi-shear mode. Camera saturation is clearly visible for this cross-section because the effective photoelastic coefficient for the x-propagating shear modes are non-zero.

The elliptical acoustic momentum surface in the YZ cross-section is not visible because the YZ
Figure 3.6: (a) and (c) are measured SBD patterns for the XY and YZ cross-sections of β-BBO. (b) and (d) show the cross-sections of the slowness surface from the optimized elastic coefficients on top of the measured SBD pattern.

plane, for trigonal crystals, is a mirror plane. The YZ mirror plane requires one of the shear modes to be a pure mode with the particle motion purely in the \( \hat{x} \)-direction. If the mode were not pure but had some polarization in the YZ plane, then the symmetry of the mirror plane would be broken. The other two modes have their polarizations in the YZ plane, and thus also obey the symmetry of the mirror plane. For the \( \hat{x} \)-polarized shear mode with an elliptical momentum surface cross-section, the effective photoelastic coefficient is equal to zero for an incident \( \hat{x} \)-propagating optical beam in a trigonal crystal.

For crystals with trigonal symmetry, like \( \alpha \)-BBO, I have solved for the analytic solutions of the acoustic velocity for the XY, XZ, and YZ cross-sections; therefore, I can analytically calculate the slowness cross-section for my optimization algorithm for collimated light propagating perpendicular to the crystallographically cut faces. Other angles would require numerical solutions using the Christoffel equation of the appropriate cross-section. The cross-section analytic solutions are listed in Appendix A. Using the analytic solutions, I have optimized the elastic-stiffness coefficients by fitting to the measured SBD patterns. The middle row of Fig. 3.6 shows the resulting fits after optimization. Table 3.3 lists the optimized elastic-stiffness coefficients.
### 3.2.4.1 SBD \(\beta\)-Barium Borate Measurements

\(\beta\)-BBO is a piezoelectric crystal because, as it belongs to the 3m point group, it lacks inversion symmetry. Within a piezoelectric medium, the electromagnetic field and mechanical stress are coupled. Therefore, the electric constitutive relation (Eq. 2.8) and Hooke’s law (Eq. 2.42) need to be augmented to include the piezoelectric effect:

\[
D_i = \varepsilon_{ikl}S_{kl} + \epsilon^S_{ik}E_k
\]

(3.12)

\[
T_{ij} = C^E_{ijkl}S_{kl} - \epsilon_{ijk}E_k
\]

(3.13)

Here \(\epsilon_{ijk}\) is the third-rank piezoelectric stress coefficient. The superscripts \(E\) and \(S\) denote values measured under constant electric field and constant strain, respectively.

With Eqs. 3.12 and 3.13, the Christoffel equation can be rederived to include the piezoelectric effect [25, 24]:

\[
\left(C^E_{ijkl} + \frac{(\epsilon_{pij}K_{p})(\epsilon_{qkl}K_{q})}{\epsilon^S_{jk}K_{j}K_{k}}\right)K_{j}K_{k}U_{l} = \rho V_a^2 U_i.
\]

(3.14)

As the above equation shows, the piezoelectric effect essentially stiffens the stiffness coefficients to make the acoustic waves travel faster in the medium. When fitting to \(\beta\)-BBO SBD patterns, I use Eq. 3.14 to numerically solve for the SBD patterns.

For \(\beta\)-BBO, I was only able to get a crystal with four sides polished. I chose to have the YZ and XY faces polished, so I was only able to measure these two cross-sections with SBD. The specimen also had several cracks, but I was able to navigate the incident beam through the crystal without propagating through these defects. I do not believe that these defects impaired my SBD analysis.

### 3.2.5 SBD Error Analysis

To help determine the error bars for my SBD measurements, I have taken the derivative of the analytic solutions to the XY, XZ, and YZ cross-sections of \(\alpha\)-BBO with respect to each independent elastic-stiffness coefficient. Figure 3.7 plots the results for the three different acoustic eigenmodes.
Figure 3.7: Illustration of the derivatives (in units of pixels/GPa) of the XY, XZ, and YZ SBD pattern with respect to the six independent elastic stiffness coefficients (green-solid: slow-shear mode, blue-dashed: fast-shear mode, red-dashed-dotted: longitudinal mode).

Of α-BBO. A larger derivative results in a smaller error bar because a larger derivative implies that a smaller change to the independent variable is required to transform the calculated SBD pattern beyond the experimental errors. Figure 3.7 shows how the slow-shear mode, illustrated by a green solid line, produces the largest derivatives, while the longitudinal mode, red dashed-dotted line, is barely visible in most of the plots, meaning that the slow-shear mode produces more information about the elastic coefficients than the longitudinal mode. Also, the asymmetry of the plots reveals how the SBD patterns unevenly carry information about the stiffness coefficients.

Of the six independent elastic coefficients, the two coefficients most associated with shear modes, $C_{44}$ and $C_{14}$, produce the largest derivatives. Also, the largest derivatives of $C_{11}$ and $C_{12}$ can be attributed to their contribution to the shear elastic coefficient $C_{66} = \frac{1}{2}(C_{11} - C_{12})$. Figure 3.7 shows the similarities of the derivatives of $C_{11}$ and $C_{12}$, which reveals that the difference of $C_{11}$ and $C_{12}$, or in other words $C_{66}$, has a smaller error than the individual magnitudes of $C_{11}$ and $C_{12}$. The elastic coefficients $C_{13}$ and $C_{33}$ also demonstrate coupling similar to $C_{11}$ and $C_{12}$ that
increases their respective error bars.

In general the shear elastic coefficients will have smaller errors than the longitudinal coefficients, and the diagonal coefficients will have smaller errors than the off-diagonal coefficients. However, this is assuming that there are no missing data in the measured SBD patterns. Any variation in the intensity distribution in the SBD pattern will affect the error analysis.

### 3.3 Resonant Ultrasound Spectroscopy

Resonant ultrasound spectroscopy (RUS) is the study of resonance frequencies to obtain information about a material. A good example of the resonance spectrum containing information is a bell. The resonance frequencies of a bell depend on its shape, composition, and size. Any change to the properties of the bell or any added defect would change the harmonics. As a result, information about the bell can be gained by studying the resonance frequencies.

The first RUS experiment to measure the elastic-stiffness coefficients of a specimen was conducted by D. B. Fraser and R. C. LeCraw in 1964 [46]. They measured the elastic-stiffness coefficients of isotopic spheres with \( \text{mm} \) diameters by comparing measured resonance frequencies to analytic solutions. They also measured the decay of vibration for various resonant modes to determine the internal friction.

H. Ekstein and T. Schiffman were the first in 1956 to numerically calculate the resonance frequencies of a specimen, although not very accurately, by applying the Rayleigh-Ritz method to the boundary value problem of a specimen [47]. They also applied group theory to their basis functions to simplify their numerical calculations and to classify the eigenmodes according to their symmetry properties. R. Holland in 1968 numerically solved for the resonance frequencies of a specimen by applying the Rayleigh-Ritz method to the Lagrangian of a specimen with no applied external forces. The eigenvalues and eigenvectors of the Lagrangian represented the resonance frequencies and resonant deformations of the specimen, respectively [48]. H. Demerast, in 1971, improved on Holland's work by choosing Legendre polynomials as the basis set. Demarest was also able to apply the method and accurately calculate the resonance frequencies of a rectangular...
Figure 3.8: (a) Illustration of the RUS experiment and resulting data. Two spring-loaded, piezoelectric transducers lightly hold the specimen by its corners. A direct-digital-synthesis (DDS) function generator drives the bottom transducer through a range of frequencies from 1kHz-1MHz. The top transducer measures deformations in the specimen and a real-time spectrum analyzer (RSA) records the resonant frequencies. (b) Picture of RUS experiment.

parallelepiped specimen of isotropic fused silica [49]. I. Ohno in 1976 applied Demerast’s theory to rectangular parallelepiped crystals of general symmetry and exhaustively showed how to calculate, using simplifications from group theory, the resonance frequencies of an orthorhombic crystal [50]. The culmination of Ohno’s work is commonly referred to as the rectangular parallelepiped resonance (RPR) method.

The choice of a Legendre polynomial basis set restricts RUS to rectangular parallelepiped specimens. A. Migilori was able to lift this restriction by using a Cartesian power series $x^i y^m z^n$, which allows the specimen to be of any shape. Migilori demonstrated the Cartesian power series on a variety of shapes including cones, pyramids, and potatoes [39]. Migilori also contributed to RUS by exhaustively describing the specimen preparation, the electronics, the transducers, and error analysis required for RUS.
3.3.1 RUS experiment

A typical RUS measurement is illustrated in Fig. 3.8a in which a specimen is lightly held between two piezoelectric transducers. Any stress from the transducers will cause a positive shift in the measured resonance frequencies. However, the measured accuracy will be on the order of a tenth of a percent as long as the loading is only slightly larger than the weight of the specimen [51]. The transducers do not need to be bonded to the specimen and strong coupling is not required or wanted [52]. To measure the resonance spectrum, a function generator sweeps through a range of frequencies on the order of kHz-MHz. This signal is amplified and then drives one of the transducers. The transducer undergoes periodic oscillations pushing on the corner of the sample and will excite a resonant mode in the specimen if the transducer oscillates at one of the resonance frequencies. As the mode oscillates, the specimen will periodically undergo a characteristic sequence of displacements that will push on the receiving transducer, causing the transducer to compress and output an oscillating voltage. The amplitude of the transducer voltage is measured for a range of frequencies; thus, determining the resonances. The transducers will not measure a resonance mode if either transducer is placed at a node, and missing a resonance mode in the RUS data makes the optimization of stiffness coefficients significantly more difficult. To avoid placing the transducers at nodes, the transducers are placed at locations of low symmetry, such as edges or corners. A single RUS measurement can yield hundreds of the lowest order modes, although less than 50 or so of these resonance frequencies are needed to optimize the non-zero elastic-stiffness coefficients [51]. The amplitude of the resonances will vary depending on the motion of the coupling point of the transducer, but this does not affect the measurement since only the resonance frequencies are used for determining the elastic coefficients.

3.3.2 RUS Computation

With the measured resonance spectrum, the next step is to calculate the spectrum from a ‘best guess’ $C_{ijkl}$ set and compare the two spectra. Currently no analytic solutions exist for exactly
calculating the resonant modes of a specimen, except for a few special cases [51]. The solutions, however, can be approximated by applying the Rayleigh-Ritz method to the free vibration of an anisotropic elastic body [49].

The Rayleigh-Ritz method is applied by expanding the displacement vector field \( u_i(\bar{r}, t) \) from the kinetic and potential energies of Eq. 3.6 in an orthogonal basis

\[
u_i(\bar{r}, t) = a_{ip} \Phi_p(\bar{r}) e^{i\omega t}.
\]

(3.15)

Here \( \omega \) is the angular frequency of the vibrations, \( \Phi_p(\bar{r}) \) are the basis functions, and \( a_{ip} \) are the expansion coefficients to be determined indexed by the displacement direction \( (i = 1, 2, 3) \) and basis order \( p \). Inserting Eq. 3.15 into Eq. 3.6 and dropping the common \( e^{i\omega t} \) time dependence, the RUS Lagrangian becomes

\[
L = \frac{1}{2} a_{ip} a_{kq} \left( \rho \omega^2 \int_V I_{ik} \Phi_p \Phi_q dV - \int_V C_{ijkl} \frac{\partial \Phi_p}{\partial x_j} \frac{\partial \Phi_q}{\partial x_l} dV \right),
\]

(3.16)

where \( I_{ik} \) is the Kronecker delta matrix. Equation 3.16 can be written more compactly using vector notation where \( a_{ip} \rightarrow \bar{a} \) (with size \( 3N \) for \( N \) basis functions polarized along each axis \( i \)):

\[
L = \frac{1}{2} \left( (\rho \omega^2) \bar{a}^T \bar{E} \bar{a} - \bar{a}^T \bar{\Gamma} \bar{a} \right).
\]

(3.17)

Here the integrals appearing in Eq. 3.16 are now elements of the \( 3N \times 3N \) matrices \( \bar{\Gamma} \) and \( \bar{E} \). Applying the Euler-Lagrange equation [45] to Eq. 3.17 and assuming that \( \rho \) and \( C_{ijkl} \) are independent of time and space results in

\[
\bar{\Gamma} \bar{a} = \rho \omega^2 \bar{E} \bar{a}.
\]

(3.18)

Equation 3.18 is a generalized eigenvalue equation and the eigenvalues \( \lambda_n = \rho \omega_n^2 \) yield the resonance frequencies, while the eigenvectors \( \bar{a}_n \) determine the mode of vibration as a weighted sum of the orthogonal basis \( \bar{a}_n^T \Phi \).

Calculating the resonance frequencies of a specimen requires calculating both \( \bar{\Gamma} \) and \( \bar{E} \) and solving for the eigenvalues. Appropriate choice of the basis functions \( \Phi_p \) can greatly simplify this calculation. For instance Demarest and Ohno preferred normalized Legendre polynomials
\(P_\lambda(x)P_\mu(y)P_\nu(z)\), where \(\lambda\), \(\mu\), and \(\nu\) are integer values determining the order of Legendre polynomials \(P\) along \(x\), \(y\), and \(z\), because for rectangular-parallelepiped specimens \(\bar{E}\) reduces to the identity matrix and \(\bar{\Gamma}\) reduces to a block-diagonal matrix with the number of blocks depending on the symmetry of the specimen [49, 50]. Migolori, on the other hand, favored a series expansion \(x^\lambda y^\mu z^\nu\) because it works for various shapes; however, the \(\bar{E}\) matrix no longer reduces to the identity matrix [39]. I chose to work with Legendre polynomials as the basis function because our specimens are all rectangular parallelepipeds.

### 3.3.3 Calculating \(\Gamma\)

As discussed above, RUS solves for the resonance frequencies by solving a generalized eigenvalue problem, Eq. 3.16, where the potential and kinetic energies are represented by large matrices \(\bar{\Gamma}\) and \(\bar{E}\), respectively. The basis set for the RPR method are normalized Legendre polynomials:

\[
\Phi = \frac{1}{L_1L_2L_3} P_{\lambda}\left(\frac{x_1}{L_1}\right) P_{\mu}\left(\frac{x_2}{L_2}\right) P_{\nu}\left(\frac{x_3}{L_3}\right) u_i.
\]  

(3.19)

Here \(L_i\) is the half-length of the specimen along \(x_i\) and \(u_i\) is the unit vector in the \(x_i\) direction. The half-length \(L_i\) appears in the denominator and in front of the polynomials so that normalized Legendre polynomials remain orthonormal for the integral calculations for \(\bar{E}\). As a result, \(\bar{E}\) reduces to the identity matrix \(\bar{I}\) and the eigenvalue problem in Eq. 3.16 simplifies to

\[
\bar{\Gamma}\bar{a} = \rho\omega^2 \bar{\delta}\bar{a}.
\]  

(3.20)

With this reduction, only the matrix values of \(\bar{\Gamma}\) need to be determined.

The matrix values of \(\bar{\Gamma}\) are determined by the integral

\[
\Gamma_{pq} = \int_V C_{ijkl} \frac{\partial \Phi_p}{\partial x_j} \frac{\partial \Phi_q}{\partial x_l} dV.
\]  

(3.21)

Ohno provides a detailed solution for \(\bar{\Gamma}\) for a crystal of general symmetry by breaking \(\bar{\Gamma}\) into 9 different blocks:

\[
\bar{\Gamma} = \begin{pmatrix}
(1,1) & (1,2) & (1,3) \\
(2,1) & (2,2) & (2,3) \\
(3,1) & (3,2) & (3,3)
\end{pmatrix}.
\]  

(3.22)
Each block represents different polarization states of the basis functions $\Phi_p$ and $\Phi_q$ in Eq. 3.21. For instance $(2,3)$ is for $\Phi_p$ and $\Phi_q$ polarized along the $x_2$ and $x_3$ axes. $\bar{\Gamma}$ is a symmetric matrix, so Ohno [50] provides solutions to the upper triangle of $\bar{\Gamma}$, except he provides the solution of $(3,1)$ instead of $(1,3)$ for some reason. There is also a typographical error for the $(3,3)$ solution which should read as

$$(3,3) = C_{55}g_1 + C_{44}g_2 + C_{33}g_3 + C_{34}g_4 + C_{35}g_6 + C_{45}g_8 + C_{45}g_9. \tag{3.23}$$

Here $C_{ij}$ are elastic-stiffness coefficients and $g_i$ are given by Ohno and depend on the order of the Legendre polynomials $\lambda, \mu, \nu$ of the basis functions $\Phi_p$ and $\Phi_q$ [53].

The size of each block in Eq. 3.22 depends on the number of basis functions polarized along each direction $x_i$. To limit the rank of $\bar{\Gamma}$, the order of the basis functions are usually truncated so that

$$\lambda + \mu + \nu \leq R, \tag{3.24}$$

where $R$ is some integer. This condition limits the number of independent basis functions polarized along each axis to

$$N = \frac{(R + 3)!}{6(R!)}. \tag{3.25}$$

For $R = 2$, $N = 10$ and the $x^\lambda y^\mu z^\nu$ basis functions are

$$\Phi = \begin{bmatrix} 1 & x & y & z & xy & xz & yz & x^2 & y^2 & z^2 \end{bmatrix}, \tag{3.26}$$

while for $R = 3$, $N = 20$

$$\Phi = \begin{bmatrix} 1 & \ldots & xyz & x^2 y & x^2 z & xy^2 & y^2 z & xz^2 & yz^2 & x^3 & y^3 & z^3 \end{bmatrix}. \tag{3.27}$$

Each basis function can be polarized along one of three different axes, so $\vec{a}$ is a $3N$ component vector and $\bar{\Gamma}$ and $\vec{E}$ are $3N \times 3N$ matrices.

### 3.3.3.1 Symmetry

The $\bar{\Gamma}$ matrix can be reduced by considering the symmetry of the problem. $\alpha$-BBO belongs to the $\bar{3}m$ point group that has a three-fold symmetry around the $z$-axis $C_3(z)$. However, both the
Legendre Polynomials and the rectangular parallelepiped cut crystal do not share this three-fold symmetry. The highest symmetry shared between the $\bar{3}m$ space group, the Legendre Polynomials, and a crystallographically cut, parallelepiped crystal is the $C_{2h}$ point group with the $x$-axis replacing the $z$-axis as the axis of highest symmetry. Table 3.1 lists the irreducible representations (irreps) of the symmetry operations for $C_{2h}$. Every eigenvector $\bar{a}_n$ belongs to one of the irreps, and eigenvectors from different irreps are orthogonal. This allows the $\bar{\Gamma}$ matrix to be reduced to a diagonal matrix with four blocks along the diagonal, one for each irrep of $C_{2h}$. Also, there will be no degenerate eigenfrequencies because every irrep of $C_{2h}$ is one-dimensional [21]. The $\bar{\Gamma}$ matrix can be further reduced if both the crystal cut and the basis functions are adjusted to match the symmetry of the crystal. For trigonal crystals, this requires a hexagonally cut crystal and a complicated basis set in cylindrical coordinates [54].

The basis functions $\bar{\Phi}$, the Legendre Polynomials, need to be sorted into their respective irreps so that $\bar{\Gamma}$ will be a block diagonal matrix. There are four different symmetry operations in the $C_{2h}$ point group: identity $E$, two-fold rotation around the $x$-axis $C_2(x)$, inversion $i$, and the YZ mirror plane $\rho_{yz}$. If a basis function undergoes a symmetry operation and the sign of the function remains unchanged, then the function is symmetric with respect to the symmetry operation and is represented by a 1 in the irrep. If the sign of the function changes, then the function is antisymmetric with respect to the symmetry operation and is represented by a $-1$ in the irrep. The basis functions can be sorted into the proper irreps by determining how the basis function transforms with respect to each symmetry operator.

Table 3.1: Representations of $C_{2h}$ point group. $E$ identity, $C_2(x)$ 2-fold rotation, $i$ inversion, $\rho_{yz}$ mirror reflection in $x$.

<table>
<thead>
<tr>
<th></th>
<th>$E$</th>
<th>$C_2(x)$</th>
<th>$i$</th>
<th>$\rho_{yz}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_g$</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$A_u$</td>
<td>1</td>
<td>1</td>
<td>-1</td>
<td>-1</td>
</tr>
<tr>
<td>$B_g$</td>
<td>1</td>
<td>-1</td>
<td>1</td>
<td>-1</td>
</tr>
<tr>
<td>$B_u$</td>
<td>1</td>
<td>-1</td>
<td>-1</td>
<td>1</td>
</tr>
</tbody>
</table>

Table 3.2: Parity of the Legendre polynomial basis functions of order $\lambda$, $\mu$, and $\nu$ for the four mode groups $A_g$, $A_u$, $B_g$, and $B_u$. $E$: even, $O$: odd.

<table>
<thead>
<tr>
<th>Direction of Displacement</th>
<th>$A_g$</th>
<th>$A_u$</th>
<th>$B_g$</th>
<th>$B_u$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\hat{x}$</td>
<td>$\lambda$</td>
<td>$\mu + \nu$</td>
<td>$\lambda$</td>
<td>$\mu + \nu$</td>
</tr>
<tr>
<td>$\hat{y}$</td>
<td>$\lambda$</td>
<td>$\mu + \nu$</td>
<td>$\lambda$</td>
<td>$\mu + \nu$</td>
</tr>
<tr>
<td>$\hat{z}$</td>
<td>$\lambda$</td>
<td>$\mu + \nu$</td>
<td>$\lambda$</td>
<td>$\mu + \nu$</td>
</tr>
</tbody>
</table>
The identity operator $E$ leaves a function unchanged, so it is represented by a 1 for all irreps. The symmetry operator $\rho_{yz}$ transforms $x \rightarrow -x$ while leaving $y$ and $z$ unchanged. For Legendre polynomials, even and odd order polynomials correspond to even and odd functions, respectively \[ P_{\text{even}}(x_i) = P_{\text{even}}(-x_i) \quad \text{and} \quad P_{\text{odd}}(x_i) = -P_{\text{odd}}(-x_i) \]. Hence, an $\hat{x}$-polarized Legendre basis function with $\lambda = \text{odd}$ will transform as $P_{\text{odd}}(x)P_{\mu}(y)P_{\nu}(z)\hat{x} \xrightarrow{\rho_{yz}} -P_{\text{odd}}(-x)P_{\mu}(y)P_{\nu}(z)(-\hat{x})$, which is symmetric with respect to $\rho_{yz}$. This example can be derived from Tables 3.1 and 3.2.

From Table 3.1 we see that both the $A_u$ and $B_g$ irreps are symmetric with respect to the $\rho_{yz}$ operation, and Table 3.2 shows that $\lambda$ must be odd for $\hat{x}$-polarized basis functions in the $A_u$ and $B_g$ irreps. Tables 3.1 and 3.2 also show that for $\hat{y}$ and $\hat{z}$-polarized basis functions, $\lambda$ must be even to be symmetric with respect to the $\rho_{yz}$ symmetry operator, as Fig. 3.9 illustrates.

For the $C_2(x)$ symmetry operator, $y$ and $z$ change signs, but $x$ remains unchanged. As result an $\hat{x}$-polarized Legendre polynomial will transform as $P_{\lambda}(x)P_{\mu}(y)P_{\nu}(z)\hat{x} \xrightarrow{C_2(x)} P_{\lambda}(x)P_{\mu}(-y)P_{\nu}(-z)\hat{x}$, which will be symmetric or antisymmetric if $\mu + \nu = \text{even}$ or $\mu + \nu = \text{odd}$, as illustrated in Fig. 3.9. Once again further examples can be derived from Tables 3.1 and 3.2. For instance for $\hat{y}$ and $\hat{z}$-polarized functions, $\mu + \nu = \text{odd}$ or $\mu + \nu = \text{even}$ for symmetric or antisymmetric functions with respect to the $C_2(x)$ operation.

The final symmetry operator to consider is the inversion operator $i$ that is a reflection through the origin, and thus changes the sign of each coordinate: $P_{\lambda}(x)P_{\mu}(y)P_{\nu}(z)u_i \xrightarrow{i} P_{\lambda}(-x)P_{\mu}(-y)P_{\nu}(-z)(-u_i)$. Therefore, Legendre polynomials for any polarization with $\lambda + \mu + \nu = \text{odd}$ will be symmetric for the inversion operator $i$.

By understanding how the Legendre polynomials transform with respect to the four symmetry operators of $C_{2h}$, we can sort the basis functions into the appropriate irreps. For instance for the $A_g$ irrep, the basis functions need to be symmetric with respect to each symmetry operator. $E$ puts no restriction on the basis functions, but $\rho_{yx}$ and $C_2(x)$ require that $\lambda = \text{odd}$ and $\mu + \nu = \text{even}$ for $\hat{x}$-polarized functions, and $\lambda = \text{even}$ and $\mu + \nu = \text{odd}$ for both $\hat{y}$ and $\hat{z}$-polarized functions. For the last symmetry operator $i$, $\lambda + \mu + \nu$ must be odd, which agrees with the restrictions of $\rho_{yz}$ and $C_2(x)$. Table 3.2 lists the parity of each polarization for the Legendre polynomials required for the
Figure 3.9: Illustration of the symmetric and antisymmetric modes for different polarizations for the YZ mirror plane $\rho_{yz}$ and the 2-fold rotation around the x-axis $C_2(\hat{x})$ symmetry operators.

four different irreps of $C_{2h}$.

The names of the four groups of $C_{2h}$ come from group theory where $A$ and $B$ are one-dimensional representations that give a 1 or -1 to each rotation $C_n$ about the principal axis. When the inversion operator $i$ appears, $g$ or $u$ (gerade-even, ungerade-odd) are added for irreps that assign a 1 or -1 for $i$. With this knowledge, the similarities and differences between the four irreps for $C_{2h}$ are easily understood.

If the basis functions that comprise $\Phi$ are sorted so that functions that belong to $A_g$ come first, followed by the basis functions that belong to $A_u$, $B_g$, and then $B_u$, then the resulting $\tilde{\Gamma}$ matrix will be block diagonalized as in Fig. 3.11a. With $\tilde{\Gamma}$ block diagonalized, the eigenvalue problem
of Eq. 3.20 simplifies by solving for the eigenvalues of four smaller matrices instead of one larger matrix. If we assume that for an $N \times N$ matrix that each block is of size $N/4$, then the number of calculations required to fill the elements of the $\bar{\Gamma}$ matrix reduces by a factor of $\frac{1+N}{1+N/4}$. Here I am only considering filling out the upper or lower triangle of the symmetric $\bar{\Gamma}$ matrix. The computation time required for calculating the eigenvalues of a matrix scales as $N^3$, so calculating the eigenvalues of four smaller matrices is 16 times faster $\left(4 \left(\frac{N}{4}\right)^3 = \frac{N^3}{16}\right)$. Thus, reducing the $\bar{\Gamma}$ matrix greatly reduces the computation time required for RUS. However, the simplifications allowed by symmetry only apply to parallelepiped crystals cut along the XYZ axes. If the crystal is cut at a rotated angle, then the symmetry of the problem is violated and the $\bar{\Gamma}$ matrix can no longer be block diagonalize. RUS optimizations that refine the orientation of the crystal take significantly more time to converge because the crystal is assumed to not be properly cut along the crystallographic axes.

An eigenvector from the RUS calculations describes the deformation of the crystal for a given eigenmode as the weighted sum of the basis functions. Figure 3.11b displays four different exaggerated illustrations of a static deformation, one for each irrep, of static deformations in $\alpha$-BBO. The actual modes periodically oscillate at the resonant frequency for that eigenmode. Each image in Fig. 3.11b reveals the symmetry of its respective irrep. The $B_g$ mode, for instance, clearly demonstrates asymmetry and symmetry with respect to the $C_2(x)$ and $\rho_{yz}$ operators. Unfortunately, the symmetry with respect to inversion $i$ is difficult to see in this image. With the deformations displaying the symmetry of their respective irrep, RUS is sometimes supplemented with a scanning laser vibrometer that measures surface deformations to unambiguously identify the resonant modes. Mode identification simplifies the optimization algorithm, but also may be necessary if no prior information about the elastic stiffness coefficients is known [55].

It is worth noting that there are six independent eigenvectors that will have a resonance frequency $\omega = 0$ Hz, which are always found by the generalized eigensolution. These eigenmodes are associated with uniform translations (one for each axis direction) and uniform rotations (one about each axis). A uniform displacement of the molecules causes the restoring forces to vanish
Figure 3.10: (a) Illustration of the $1365 \times 1365 \Gamma$ matrix for a 12th order RUS calculation with our $\alpha$-BBO specimen. The black dots in the image represent non-zero values. b) Exaggerated illustrations of the deformations of our $\alpha$-BBO specimen for four different eigenmodes.

because there is no deformation in the crystal, so the crystal can not create an oscillatory motion.

3.3.4 $\alpha$-BBO Calculations

The RUS calculations are more accurate for larger $R$, but the computation time increases as the $\Gamma$ matrix grows in size. I found the computation time to be nominally proportional to $R^5$. $R$ is chosen as to balance the computation time with the accuracy of the RUS calculations. Figure 3.11 shows how both the computation time and accuracy of an RUS computation depend on the maximum Legendre order $R$. In RUS literature $R$ is usually set to 10, but with modern computers higher order Legendre polynomials can be used for RUS optimizations. I chose to work with $R = 12$ because the largest percent error for the first 42 resonance frequencies when compared to $R = 15$...
Figure 3.11: (a) Computation time on a Toshiba Satellite laptop for RUS calculation as a function of max Legendre order \( R \). The dashed-black line represents an \( R^5 \) curve. b) Percent difference of RUS calculation for various modes for different \( R \) compared to \( R = 15 \).

is less than 0.28\%, and an optimization run converges overnight.

Figure 3.11b reveals a couple of note-worthy facts about the RUS computation. First the accuracy of the \( A_g \) and \( B_g \) modes do not improve when \( R \) increases from an odd value to an even value. Similarly the \( A_u \) and \( B_u \) modes do not increase when \( R \) increases from an even value to an odd value. This is because the inversion operator \( i \) requires that the sum of the Legendre polynomials to be odd or even for polynomials that are symmetric or antisymmetric from an inversion \( i \), respectively. Therefore, any basis function where the Legendre orders sum to an odd value will belong to either the \( A_g \) or \( B_g \) irrep, but can not possibly belong to the \( A_u \) or \( B_u \) irreps. In other words, \( R \) increasing from an odd value to an even value provides no new basis functions to the RUS calculation that could belong to the \( A_g \) or \( B_g \) irreps. The second fact that Fig. 3.11b reveals is that in general the accuracy of the RUS calculation decreases for larger frequencies. However, there are few exceptions to this rule as seen with \( B^{14}_g \) and \( B^{11}_g \), where the superscript refers to the \( n^{th} \) smallest eigenmode of a given irrep.

With my RUS experiment, I measured 42 resonance frequencies and optimized the elastic
Figure 3.12: Flowchart of the optimization process for RUS in which the elastic stiffness coefficients are refined until the error between the measured and calculated resonance spectra is minimized.

coefficients to these measured resonances using the MATLAB Nelder-Mead optimizer. I assumed that I measured the first 42 resonance frequencies without missing a single resonance. Had I missed a resonance frequency, my code would not have converged correctly. Figure 3.12 shows a flowchart of my optimization algorithm in which I refine the stiffness coefficients until the optimizer minimizes the error between the calculated and measured resonance frequencies. The first row of Table 3.3 lists the RUS measured elastic coefficients for $\alpha$-BBO. The RMS percent error for the calculated versus measured frequencies is 0.29%.

The crystal I received from Newlight Photonics was mislabeled with the X and Y axes confused. Fortunately, the RUS optimizer could only converge to elastic-stiffness coefficients that resulted in an RMS percent error of 0.86%, which is large for RUS. In order to decrease the error, I optimized the orientation of the crystal by adding rotation angles to the optimization loop in addition to the $C_{IJ}$. As mentioned previously, optimizing the orientation of the crystal does not allow for the $\tilde{\Gamma}$ matrix to be reduced because the symmetry of the problem is violated for a crystal not cut along the crystallographic axes. As a result, the RUS computation and optimization time increases significantly, so I sacrificed accuracy for speed and set $N = 9$ for this optimization. The optimizer suggested the X and Y confusion, so I redid the optimization but switched the dimensions of the X and Y axes. Fortunately the crystal was cut with asymmetric dimensions, otherwise I would not have been able to differentiate the X and Y axes with RUS. With the use of a laser
vibrometer, I may have been able to distinguish the X and Y axes by looking at the symmetry of the deformations. When I later performed the SBD measurements, I was able to confirm the X and Y axes confusion by observing the cross-section that only had two visible momentum surfaces, which is required to be the YZ cross-section for trigonal crystals.

Figure 3.13 illustrates how the eigenfrequencies change as the orientation of the crystal cut is rotated around the X, Y, and Z axes for the 8.12×6.08×7.10 mm specimen of α-BBO. Even though the eigenfrequencies vary a noticeable amount for a 90° rotation about the Z axis, this does not...
mean that the RUS code should not be able converge when confusing the X and Y axes. The fact that the optimizer was unable to accurately converge with the X and Y axes confused is probably a reflection of the limitations of the symmetry of crystal, a point better understood by looking at the SBD patterns.

The eigenfrequencies in Fig. 3.13 vary significantly around the X and Y axes, but not around the Z-axis. This can be explained by looking at the SBD patterns. For instance, the XY SBD is very symmetric with little change in the SBD as you rotate around the Z-axis. If the crystal is cut at an angle rotated with respect to the Z-axis, the decomposition of the RUS modes into the Fourier eigenmodes will change. However, the change in the resonant frequencies will not significantly change as the XY SBD is very symmetric. The XZ and YZ SBD patterns on the other hand, are very asymmetric and any change to the crystal cut around the X or Y axes will greatly change the Fourier decomposition into the acoustic eigenmodes and thus change the measured resonance frequencies.

### 3.3.5 β-BBO Calculations

For the β-BBO calculations, the piezoelectricity must be included when determining the resonant frequencies. The piezoelectricity causes a stiffening of the elastic tensor that causes a positive shift in the resonant frequencies. Failure to include the piezo-electric effect in the RUS calculations will increase the error of the measurements.

To include the piezoelectric effect in the RUS calculations, the electric potential $\phi$ must be expanded with an orthogonal basis set, as is done with displacement field $u$. Once again, the symmetry of the crystal allows for a simplification of the calculations. Ohno and Ogi provide a detailed analysis including the piezoelectric effect in trigonal crystals and provide sample calculations when excluding and including the piezoelectric effect [56, 57]. My own calculations agreed with those presented by Ohno and Ogi, confirming the accuracy of my RUS piezoelectric code.

Despite the accuracy of my code, I was unable to successfully measure the stiffness coefficients of β-BBO with RUS because of the defects in the specimen. I optimized the elastic and piezoelectric
coefficients to 40 measured resonance frequencies, but the RUS optimizer was never able to converge. In addition, the difference of measured resonance spectrum with the calculated resonance spectrum using the stiffness coefficients measured from SBD exceeded 10% for multiple frequencies. I believe that the defects of the specimen caused such large shifts in the resonance spectrum making the optimization of the elastic coefficients impossible.

3.3.6 RUS Error Analysis

Calculation of the resonance frequencies of a specimen depends on the specimen shape, orientation, size, density and any external loading. Any deviation from an ideal specimen will cause a disagreement in the measured and calculated resonance frequencies; therefore, great care is taken in specimen preparation, and precise knowledge of the specimen dimensions and density is required. Unfortunately, $\alpha$-BBO is a soft crystal (Mohs hardness of 4), which allowed the crystal to be easily scratched and rounded by the experimental transducers. The effect this had on the measured resonance spectrum is difficult to quantify.

To determine the error bars for my RUS measurements, I varied the elastic-stiffness coefficients until the change in the calculated resonance spectrum exceeded experimental error. The average, absolute percent error of the calculated versus measured frequencies of 0.29% provided a metric of the experimental errors. As with SBD, I found the shear elastic coefficients to have a smaller error than the longitudinal coefficients, and the diagonal coefficients to have a smaller error than the off-diagonal coefficients. I also found coupling between $C_{11}$ and $C_{12}$ and $C_{33}$ and $C_{13}$ elastic coefficients. This similarity between the RUS and SBD error analysis is not surprising because the resonant modes can be decomposed into a superposition of plane waves; therefore, the sensitivity of the resonance frequencies depend on the sensitivity of the plane waves that compose the resonance mode.
Table 3.3 lists the measured elastic-stiffness coefficients for α-BBO from RUS and SBD measurements as well as previously reported results from pulse-echo measurements [14]. The SBD measurements have smaller error bars than the RUS measurements, but both measurements agree quite well with each other. Interestingly all of the measured elastic coefficients form RUS are larger than the measured values of SBD, which may be a result of the loading of the crystal from the transducers.

The prior pulse-echo measurements concur with our results except for the $C_{44}$ and $C_{13}$ elastic coefficients. The measured value of the $C_{44}$ from the pulse-echo only differs from my values by 0.8 GPa, but the reported error bar from the pulse-echo experiment is only 0.2 GPa. The $C_{44}$ was determined by their measurement of the acoustic velocities along the X and Y axes with $\hat{z}$-polarized light, which they measured with the same velocity of $1219 \pm 25$ m/s. According to the elastic coefficients from the SBD experiment, they should have measured $V_x = 1299$ m/s and $V_y = 1219$ m/s instead of the same velocity. The analytic solutions for the acoustic velocities for these two directions are

$$V_x^2 \rho = \frac{1}{2} \left( C_{11} + C_{44} - \sqrt{C_{11}^2 + 4C_{14}^2 - 2C_{11}C_{44} + C_{44}^2} \right)$$

$$V_y^2 \rho = \frac{1}{4} \left( C_{11} - C_{12} + 2C_{44} - \sqrt{C_{11}^2 - 2C_{11}C_{12} + C_{12}^2 + 16C_{14}^2 - 4C_{11}C_{44} + 4C_{12}C_{44} + 4C_{44}^2} \right).$$

Both solutions can be approximated as $V^2 \rho = C_{44}$, which results in the reported $C_{44} = 5.6$ GPa. If instead $C_{44}$ is solved for when using the complete analytic solutions and their reported stiffness values for the other stiffness coefficients, then the calculated stiffness coefficients for $C_{44}$ is 6.6 and 5.8 GPa for the X and Y measurements, respectively. Therefore, the 0.2 GPa error bar seems to
Table 3.4: Measured elastic coefficients in units of GPa for $\beta$-BBO using SBD and pusle-echo.

<table>
<thead>
<tr>
<th></th>
<th>$C_{11}$</th>
<th>$C_{33}$</th>
<th>$C_{44}$</th>
<th>$C_{12}$</th>
<th>$C_{13}$</th>
<th>$C_{14}$</th>
<th>$C_{66}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>SBD</td>
<td>$123\pm4$</td>
<td>$52.1\pm1.2$</td>
<td>$3.40\pm0.02$</td>
<td>$63\pm3$</td>
<td>$26\pm2$</td>
<td>$-3.67\pm0.04$</td>
<td>$31.2\pm1.4$</td>
</tr>
<tr>
<td>Pusle-echo [17]</td>
<td>$123.8$</td>
<td>$53.3$</td>
<td>$7.8$</td>
<td>$60.3$</td>
<td>$49.4$</td>
<td>$12.3$</td>
<td>$31.8$</td>
</tr>
</tbody>
</table>

Table 3.5: Measured piezoelectric coefficients in units of $10^{-12}$ C/N for $\beta$-BBO.

<table>
<thead>
<tr>
<th></th>
<th>$d_{15}$</th>
<th>$d_{22}$</th>
<th>$d_{31}$</th>
<th>$d_{31}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>SBD</td>
<td>$1.25\pm.05$</td>
<td>$-0.3\pm0.15$</td>
<td>$-0.9\pm0.2$</td>
<td>$4.1\pm0.3$</td>
</tr>
<tr>
<td>[58]</td>
<td>$1.26$</td>
<td>$-0.3$</td>
<td>$-0.85$</td>
<td>$4.0$</td>
</tr>
</tbody>
</table>

The pulse-echo measurement for the $C_{13}$ coefficient, on the other hand, disagrees with my measurements by 32 GPa, but with a reported error bar of 11 GPa. I believe that this large difference in the measured $C_{13}$ coefficient could be a result of acoustic walk-off in the pulse-echo measurement. The $C_{13}$ measurement was made with with an acoustic wave launched at $\phi = \frac{\pi}{2}$ and $\theta = \frac{\pi}{4}$. Using the stiffness coefficients measured with SBD, I calculated a 31° acoustic walk-off for this mode. Such a large acoustic walk-off could cause the acoustic wave to bounce off the side walls during the pulse-echo, resulting in significant errors. Without knowing the specific details of the measurement, the size and geometry of the specimen and transducer, and their reason for reporting such large error bars, I am not sure if acoustic walk-off is responsible for the large reported value of $C_{13}$.

Tables 3.4 and 3.5 compares the measured stiffness and piezoelectric coefficients for $\beta$-BBO to previously published values. My stiffness measurements agree with the pulse-echo measurements by Eimerl [17], except for the $C_{44}$, $C_{13}$, and $C_{14}$ coefficients. Eimerl does not give explicit details about his pulse-echo measurements, but he does mention that the geometries of his pulse-echo measurements were based on the geometries suggested by R.B. Giekse [59]. Giekse suggests measuring the $C_{44}$ coefficient by launching a pure-shear mode along the Z-axis of the crystal because this mode only depends on the $C_{44}$ coefficient in trigonal crystals. What Giekse fails to consider is that the Z-axis, as required by trigonal symmetry, is an acoustic concical refraction direction. Therefore, the acoustic wave will diffract into a cone of acoustic waves that could greatly complicate
the pulse-echo measurement. If Eimerl chose this geometry, then his $C_{44}$ measurement would be fraught with potential errors that could lead to an incorrect measurement. Any inaccuracies in the $C_{44}$ measurement will cascade into the $C_{13}$ and $C_{14}$ measurements because these measurements assume that $C_{44}$ is known.

My piezoelectric values agree with the piezoelectric resonance method measurements of R. Guo and A. S. Bhalla [58]. Guo and Bhalla do not provide any error bars, so I am not certain which measurement set is more accurate.

### 3.5 SBD versus RUS

Both SBD and RUS are very similar experiments because they both measure the eigenvalues of a Lagrangian in order to optimize the elastic-stiffness coefficients. However, there are important differences between the two experiments. For example, RUS requires a numerical calculation for the eigenvalues that is computationally expensive, while SBD can use simple analytic solutions for specific crystal symmetries and cross-sections. Numerical solutions may be required for SBD, but numerically solving the Christoffel equation for a simple cross-section is relatively easy. The biggest consequence of this difference is that RUS requires hours for optimizing the elastic coefficients while SBD requires minutes. I also found that the SBD optimization was more likely to converge to a local minimum, but was easy to identify by comparing the solutions to the data. For RUS, it is difficult to visually disambiguate a local minimum from the global minimum, which is a consequence of RUS being a less visually informative experiment. Visually gaining information from a resonance spectrum is very difficult, but the SBD patterns display the crystal symmetry and anisotropy in a visually pleasing way. For example, SBD can easily distinguish the orientation of a crystagraphically cut, trigonal crystal by carefully analyzing the measured SBD patterns. The Z-axis can be distinguished because the XY cross-section must repeat its pattern every 60° to obey the crystal symmetry. The XZ and YZ cross-sections can be distinguished because one of the acoustic modes of the YZ cross-section is not visible from SBD due to symmetry. Trigonal symmetry puts no such restriction on the XZ cross-sections, so all three modes are visible as seen
in Fig. 3.6. With RUS, I can only distinguish the X, Y, and Z axes by optimizing the stiffness coefficients with the different orientations and comparing the resulting errors. This takes a few days because each optimization run takes about seven hours.

RUS also requires precise knowledge of the shape, orientation, and size of the specimen. SBD, on the other hand, assumes that the specimen is infinitely large, so the size and shape of the crystal are of no consequence, but the correct orientation of the crystal is required. With RUS, any defects to the crystal will cause a change to the measured resonance spectrum. With the $\beta$-BBO specimen, the defects were so large that the RUS algorithm was unable to converge. The SBD measurement, despite the defects, was still able to measure the stiffness coefficients. The defects, in fact, might actually help populate the spectrally rich acoustic field, creating a better data set.

Missing data are also very problematic for RUS and may be very difficult to identify if a datum is missing and where it may be missing. For SBD, missing data will only increase the error bars of the measurements and should be easy to identify if enough data are measured.

RUS can better handle smaller specimens with dimensions on the order of a few mm. For SBD the size of the specimen determines the diameter of the incident optical beam. Smaller beam diameters increase the size of the diffraction limited spot size in the optical Fourier plane; thus, increasing the experimental uncertainty of the measured SBD pattern. In contrast the resolution for RUS does not decrease for a small specimen, but the resonance frequencies are larger for smaller specimens. For both experiments, a larger specimen increases the loading on the specimen, which may result in larger experimental errors. These experimental errors, however, can be eliminated. In RUS, the resonance width is affected by the damping of the resonances, so additional information about the internal friction can be gained [60]. In SBD, the thickness of the crystal determines the width of the diffracted pattern and needs to be adequately sampled by the detector pixels.
Chapter 4

Photoelastic Coefficients

An artificial birefringence or refractive index perturbation can be created in a material by applying a mechanical stress. The magnitude and direction of the induced birefringence is determined by the unitless, fourth-rank photoelastic tensor $p_{ijkl}$ and the fourth-rank elastic-stiffness tensor $C_{ijkl}$. Determining the induced modification of the dielectric tensor is vital for designing acousto-optic (AO) devices because the magnitude and direction of the artificial birefringence governs the diffraction efficiency of the device. As shown in Chapter 5, the effective photoelastic coefficient can demonstrate a larger anisotropy than the acoustic slowness. Therefore, accurate measurements of the photoelastic coefficients are required to find the AO geometry with the largest diffraction efficiency.

In this chapter, a brief overview of the photoelastic tensor $p_{ijkl}$ is provided before discussing the different types of experiments for measuring $p_{ijkl}$. Both the Dixon and the Schaefer-Bergmann diffraction (SBD) experiments are discussed in detail with measurements of fused silica and $\alpha$-BBO as examples.

4.1 The Photoelastic Tensor

When a stress $T_{ij}$ is applied to a material, the resulting symmetric and antisymmetric gradients of the displacement vector, $S_{kl}$ and $\omega_{kl}$, are determined by the elastic stiffness tensor $C_{ijkl}$. As discussed in section 2.3.1, a non-zero $S_{kl}$ and $\omega_{kl}$ will change the permeability tensor $\eta_{ij}$:

$$\Delta\eta_{ij} = p_{ijkl}S_{kl} + P_{ij(kl)}\omega_{kl}.$$  (2.56 revisited)
Here $p_{ijkl}$ and $P_{ijkl}$ are the symmetric and antisymmetric tensors of the general photoelastic tensor $P_{ijkl}$. The antisymmetry of $P_{ijkl}$ only exists in the $k$ and $l$ coefficients because $\eta_{ij}$ must remain symmetric even after the material deformation. Excluding triclinic and monoclinic crystals, $P_{ijkl}$ is defined as

$$P_{ijkl} = -\frac{1}{2} \left( \frac{1}{n_i^2(\lambda)} - \frac{1}{n_j^2(\lambda)} \right) (\delta_{ik}\delta_{jl} - \delta_{jk}). \quad (4.1)$$

Notice how $P_{ijkl}$ only depends on the optical-wavelength-dependent index of refraction of the material $n_i(\lambda)$, so no measurements are required for $P_{ijkl}$ if $n_i(\lambda)$ is known. However, the contribution of $P_{ijkl}$ must be considered when measuring $p_{ijkl}$. Unfortunately there is no explicit definition for $p_{ijkl}$, so it must be measured with one of the experiments discussed below.

### 4.2 Measurements

There are several different techniques for measuring the photoelastic tensor of material, but they all fall under three different categories: 1) applying a static load and measuring the change in the birefringence, 2) measuring Brillouin scattering, and 3) measuring acousto-optic scattering [61, 62]. Below I briefly review the first two methods, but discuss in detail the third method.

The first technique does not directly measure the photoelastic tensor, but instead measures the fourth-rank piezo-optic tensor $q_{ijkl}$. The peizo-optic tensor describes the linear change in the birefringence of a material from an applied stress $T_{ij}$:

$$\Delta \eta_{ij} = q_{ijkl} T_{kl} \quad (4.2)$$

Here the antisymmetric contribution has been ignored. The photoelastic and piezo-optic tensors are related through the elastic-stiffness tensor:

$$p_{ijkl} = q_{ijkl} C_{klmn}. \quad (4.3)$$

Equation 4.2 shows how the piezo-optic coefficients $q_{ijkl}$ can be determined by measuring the change in birefringence from a known load. The change in birefringence can be measured using interferometers to measure fringe shifts or changes in the polarization of light [61]. The piezo-optic coefficients
are usually on the order of $10^{-12} \text{ N/m}^2$, so a large load ($\sim 1000 \text{ N}$) is required to accurately measure the artificial birefringence. To ensure that the loads do not push the sample beyond the elastic limit, the birefringence of the sample should be measured both before and after the load is applied. In addition, the experiments should measure the change in the birefringence in different parts of the crystal to ensure that the sample is uniformly stressed \cite{62}. When computing the change in the birefringence, the deformation of the specimen and the rotation of the index ellipsoid must be included in the calculations.

In the Brillouin scattering experiment, thermally induced acoustic waves diffract incident optical light. The elastic coefficients of the material can be determined by the frequency shift of the Brillouin lines while the photoelastic coefficients can be determined by the intensity of the Brillouin lines \cite{61, 62, 63, 64}. However, the light is weakly scattered and needs to be measured with high resolution, which presents a host of practical experimental difficulties \cite{65}.

The third method measures the photoelastic coefficients with acousto-optic diffraction. Chapter 3 discusses how to measure the elastic-stiffness tensor of a material by studying the shape the Schaefer-Bergmann diffraction (SBD) pattern. This chapter discusses how to measure the photoelastic tensor by measuring the intensity of the diffracted light in the SBD. Assuming perfect phase matching along the SBD pattern, the intensity of the diffracted light $I_d$ from an acoustic wave is determined by

$$I_d = \sin^2 \left[ \frac{\pi}{\lambda_0 (\cos \theta_i \cos \theta_d)^{1/2}} \sqrt{\frac{n_d^3 n_i^3 p^2 P_a L^2}{\rho V_a^3}} \right] I_i. \quad (4.4)$$

Here $\lambda_0$ is the optical wavelength, $n_d$ and $n_i$ are the indices of refraction of the diffracted and incident light, $p$ is the effective photoelastic coefficient, $\rho$ is the material density, $V_a$ is the acoustic velocity, $I_i$ is the intensity of the incident light, $P_a$ is the acoustic power density, and $L$ is the length of the optical and acoustic beam overlap. For the photoelastic measurements, I assume that $\cos \theta_i \simeq \cos \theta_d \simeq 1$ and that the diffraction efficiency is so small that the $\sin^2$ term can be dropped, reducing Eq. 4.4 to

$$I_d = \frac{\pi^2 n_d^3 n_i^3 p^2 P_a L^2}{\lambda_0^2 \rho V_a^3} I_i. \quad (4.5)$$
The effective photoelastic coefficient is determined by the incident and diffracted electric flux polarizations \( \hat{d}^{(i)}_i \) and \( \hat{d}^{(d)}_j \), the photoelastic tensor \( p_{ijkl} \), and the unit strain tensor \( \hat{S}_{kl} \) from the acoustic plane wave:

\[
p = \hat{d}^{(i)}_i \ p_{ijkl} \hat{S}_{kl} \hat{d}^{(d)}_j.
\]  

Equations 4.5 and 4.6 show how the diffracted light intensity depends on the photoelastic tensor \( p_{ijkl} \). In a given geometry, all of the variables in Eq. 4.5 are usually known except for the acoustic power density \( P_a \), even though a specific RF power is applied to the device, because the conversion of the transducer electrical power to acoustic power into the crystal is unknown. Below I discuss two different measurements, SBD and the Dixon method, that successfully eliminate the acoustic power density term \( P_a \) to measure the photoelastic coefficients with acousto-optic diffraction.

### 4.2.1 Schaefer-Bergman Measurement

In 1938 Hans Mueller proposed three different methods, which he labeled ‘A’, ‘B’, and ‘C’, for measuring the relative ratios of the photoelastic constants in either isotropic materials or cubic crystals with the acoustic wave propagating along a principal axis of the crystal [66]. In method A, a specimen is placed between two crossed polarizers. The polarizers are then rotated until the diffracted light is no longer visible for both longitudinal and shear acoustic waves. By noting the angle between the polarization of the incident light with the x-axis of the crystal, the relative ratios of the \( p_{44} \) to \( (p_{11} - p_{12}) \) can be determined for cubic crystals. In method B, linearly polarized light is diffracted by an acoustic wave in a specimen. A polarizer placed after the specimen is then rotated until the polarizer completely blocks the diffracted light. The angle of the polarizer measures the polarization of the diffracted light, from which the relative ratios of the photoelastic coefficients can be determined. For the final method, method C, Mueller proposes measuring the intensity of the acousto-optically diffracted light from two different incident beams with orthogonal polarizations. The ratio of the photoelastic coefficients are then determined from the ratio of the intensities of the diffracted light.

Mueller’s methods B and C have been applied by Bergman and Fues (1936) and Gates and
Heidemann (1956) to study the photoelastic constants of glasses [67, 68]. Narasimhamurty (1955) was the first to apply Mueller’s methods to study uniaxial and biaxial crystals [69]. In all of these experiments, only a single diffraction spot was considered at a time. Below I present a modified version of method C for measuring the ratios of the photoelastic coefficients by the measuring the intensity of the entire SBD pattern. Additionally I present my measurements of optically isotropic fused silica and the uniaxial crystal $\alpha$-BBO.

4.2.2 Schaefer-Bergman Experiment

Figure 4.1 illustrates the SBD experiment for measuring photoelastic coefficients. As with the SBD experiment for measuring the elastic-stiffness constants (see Chapter 3), I temporarily bond the specimen to a fused silica reference cell that has an attached piezoelectric transducer. I then launch acoustic waves into the fused silica using either a longitudinal or shear transducer. I launch a single frequency acoustic wave ($\sim$30 MHz) that after several reflections and scatterings fills the crystals with acoustic plane-wave components with a variety of polarizations and propagation directions. Surprisingly, nearly every possible direction of propagation and acoustic polarization of the acoustic waves can be excited, albeit with varying acoustic power in each plane-wave component.

An incident, collimated 532 nm optical beam with either vertical or horizontal polarization passes through an orthogonal face of the $\alpha$-BBO crystal to interact with the diffuse spectrally rich acoustic field. The individual acoustic plane-wave components acousto-optically diffract the incident optical wave, and only the Bragg matched components that conserve momentum will produce substantial diffraction. A Fourier transform lens maps the angles of propagation of the diffracted light onto a CCD camera to record the resulting SBD pattern. A polarizer after the crystal determines the polarization of the diffracted light detected by the CCD camera.

The specimen does not need to be bonded to a reference cell for measuring the SBD; however, using the reference cell avoids the need to bond the transducer directly to the specimen. This bonding scheme allows for the measurement of different SBD cross-sections of the specimen without the difficulty of bonding multiple piezoelectric transducers. The transducer could be bonded to the
Figure 4.1: Illustration of the SBD experiment. A diffuse spectrally-rich resonating acoustic field diffracts the incident, collimated optical beam. A Fourier transform (FT) lens maps the diffraction onto a CCD camera; thus, recording the SBD. I then extract the intensity of the SBD pattern as function of the polar angle $\theta$ for each acoustic mode.

corner of the crystal to avoid the need of doing multiple transducer bonds, and could better excite all modes of the crystal akin to low frequency resonant ultrasound spectroscopy (RUS), providing better data sets. However, my experimental set-up also easily allows for Dixon measurements, which is discussed below.

I overlap the recorded SBD with a calculated SBD pattern, which is determined by the Christoffel equation and the measured stiffness coefficients. I optimize the center and rotation of the calculated SBD by maximizing the overlap of the calculated and measured SBD patterns. Next
I measure the intensity of the recorded SBD pattern as a function of the polar angle $\theta$ by integrating a 2D Gaussian with a 1-3 pixel width centered on the calculated SBD that is overlapped with the measured SBD pattern. The measured diffraction intensity $I_d(\theta)$ allows for the determination of the ratios of the photoelastic coefficients.

Unlike the SBD experiment for the measuring the elastic-stiffness coefficients, the input and output polarizations of the optical light needs to be known in order to determine the photoelastic coefficients. Also, camera saturation results in wasted data, where for the elastic-stiffness constants experiment saturation only increases the error bars for the data. The exposure time of the CCD camera needs to be critically adjusted so that the signal is maximized without losing too much data to saturation. Lastly, I had to average about 10 different SBD patterns separated by 1 kHz to fill in the SBD pattern. A single SBD pattern was so speckled that I was unable to effectively process the data. By varying the acoustic frequency, I was able to fill in the speckle and produce a SBD that I could effectively process. The variation of the radii of the SBD over 10 kHz is negligible when the AO frequency is centered around 30 MHz, which made measuring the intensity of the SBD pattern simple as I did not need to adjust the size of the calculated SBD pattern. The center frequency of these 10 different SBD patterns was critically adjusted until the SBD was brightest. I only took 10 different SBD patterns because taking more SBD patterns did not provide more information or fill in the speckle as the SBD patterns would either start to repeat itself or become increasingly more dim as separation from the center acoustic frequency increased.

4.3 Processing the Schaefer-Bergmann Patterns

The intensity of the SBD pattern $I_d(\theta)$ for low acoustic power is described by modifying equation 4.5 to have an angle dependence $\theta$:

$$I_d(\theta) = \frac{\pi^2 n_3^2 n_3^3 \rho (\theta) P_a(\theta) L^2}{\lambda_0^2 \rho V_a^3(\theta)} I_i,$$

(4.7)

Here the intensity of the diffracted beam $I_d(\theta)$ along the SBD varies as the effective photoelastic coefficient, acoustic power $P_a(\theta)$, and acoustic velocity $V_a(\theta)$ vary as a function of $\theta$. The effective
photoelastic coefficient is still calculated by Eq. 4.6, but now the strain and optical polarizations depend on the propagation direction of the acoustic wave, and thus \( \theta \) as well.

The photoelastic tensor of a sample can be determined as long as all of the other parameters from Eq. 4.7 are known. The indices of refraction \( n_d \) and \( n_i \) and the material density \( \rho \) can easily be measured, the acoustic velocity \( V_a \) can be calculated using the Christoffel equation if the elastic stiffness tensor \( C_{ijkl} \) is known, and \( L \) is a constant that depends on the crystal and optical beam size. However, \( P_a(\theta) \) varies randomly and unpredictably for the various plane-wave components propagating and resonating around the crystal, preventing the determination of \( p_{ijkl} \). I circumnavigate this problem by eliminating both \( P_a(\theta) \) from the calculations by measuring the SBD pattern for a certain cross-section multiple times with different input and output optical polarizations, which changes the photoelastic coefficients that contribute to \( p(\theta) \) but keeps all other parameters in Eq. 4.7 constant. I then take the ratios of the angular dependence of the diffraction efficiency for different SBD patterns:

\[
\frac{I_{d1}(\theta)}{I_{d2}(\theta)} = \frac{\frac{\pi^2 n_d^3 n_i^3 p_1^2(\theta)}{\rho V_a^2(\theta)}}{\frac{\pi^2 n_d^3 n_i^3 p_2^2(\theta)}{\rho V_a^2(\theta)}} = \frac{n_d^3 n_i^3 p_1^2(\theta)}{n_d^3 n_i^3 p_2^2(\theta)}.
\]

If \( p_1(\theta) \) and \( p_2(\theta) \) depend on different photoelastic coefficients \( p_{ijkl} \), then the relative magnitudes of these photoelastic coefficients can be determined. The relative signs of the photoelastic coefficients that belong to \( p_1(\theta) \) and \( p_2(\theta) \) independently can be determined as well, but the sign of the coefficients of \( p_1(\theta) \) with respect to \( p_2(\theta) \) cannot be determined because both terms are squared in Eq. 4.8.

Below I apply my method to fused silica and \( \alpha \)-BBO. For fused silica I solved for the analytic solutions of \( p(\theta) \). I am unable to express simple analytic solutions for BBO, but I am able to determine the photoelastic coefficients that contribute to a given cross-section, acoustic mode, and incident optical polarization.
Figure 4.2: (a) Plot of the analytic solutions for $p(\theta)$ for diffraction from the longitudinal acoustic wave in fused silica using the known values $p_{11} = 0.121$ and $p_{12} = 0.270$. (b) SBD pattern for $\hat{H}$-to-$\hat{H}$ optical diffraction using a longitudinal mode transducer, and in blue the measured intensity $I_d(\theta)$ of the diffraction (in arbitrary units) from the longitudinal acoustic mode.

### 4.3.1 Optically Isotropic Materials: with fused silica as an example

For the fused silica experiments, I measured three different SBD patterns with 1) $\hat{H} \rightarrow \hat{V}$ (horizontal-to-vertical), (2) $\hat{H} \rightarrow \hat{H}$, and 3) $\hat{V} \rightarrow \hat{V}$ polarization diffraction. For each of the three different cases, I have analytically solved for $p(\theta)$ by assuming that the incident and diffracted light are either purely $\hat{H}$ or $\hat{V}$ polarized. The longitudinal wave in fused silica is a pure longitudinal mode, so the acoustic polarization is simply equal to the normalized acoustic wavevector $\hat{K}_a(\theta)$.

The photoelastic tensor written in reduced subscript notation for fused silica is

$$p_{i,j} = \begin{pmatrix}
p_{11} & p_{12} & p_{12} & 0 & 0 & 0 \\
p_{12} & p_{11} & p_{12} & 0 & 0 & 0 \\
p_{12} & p_{12} & p_{11} & 0 & 0 & 0 \\
0 & 0 & 0 & p_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & p_{44} & 0 \\
0 & 0 & 0 & 0 & 0 & p_{44}
\end{pmatrix}.$$
Here \( p_{44} = \frac{1}{2}(p_{11} - p_{12}) \), so there are only two independent photoelastic coefficients to measure. With the optical and acoustic polarizations and the photoelastic tensor, \( p(\theta) \) can be solved for each of the three different cases:

\[
\begin{align*}
\hat{p}^{\hat{H}\hat{H}} &= p_{11}\cos(\theta)^2 + p_{12}\sin(\theta)^2, \\
\hat{p}^{\hat{V}\hat{V}} &= p_{11}\sin(\theta)^2 + p_{12}\cos(\theta)^2, \\
\hat{p}^{\hat{H}\hat{V}} &= \hat{p}^{\hat{V}\hat{H}} = [p_{11} - p_{12}]\cos(\theta)\sin(\theta).
\end{align*}
\] (4.9)

Here the superscripts refer to the incident and diffracted optical polarizations, \( \theta \) is an angle measured from the orientation of the \( \hat{H} \) axis, and \( p_{11} \) and \( p_{12} \) are the two independent photoelastic coefficients of fused silica. The three solutions are different from one another, but all depend on \( p_{11} \) and \( p_{12} \). Therefore, the relative ratio and sign of the two photoelastic coefficients can be determined by fitting to the ratio of two of the above solutions.

Figure 4.2a plots \( p^2(\theta) \) for each of the three different cases. For fused silica, the longitudinal acoustic velocity is constant regardless of the propagation direction, so \( I_d(\theta) \) from Eq. 4.5 only changes as \( p(\theta) \) and \( P_a(\theta) \) change. Figure 4.2b displays a SBD pattern for \( \hat{H} \rightarrow \hat{H} \) diffraction. The acoustic slowness surface for fused silica is two spheres of different radii with the smaller sphere corresponding to the longitudinal wave. On top of the SBD in Fig. 4.2b in blue, is a plot in arbitrary units \( I_d(\theta) \) of the longitudinal SBD. Comparing the measured data to \( \hat{p}^{\hat{H}\hat{H}}(\theta) \) reveals that \( P_a(\theta) \) is not constant throughout the crystal. Most of the acoustic power propagates along the horizontal axis, which is the direction that the transducer launches the acoustic wave. However enough acoustic power is scattered into other directions for the measurement of \( p_{ijkl} \).

With the acoustic power unevenly distributed in the acoustic modes, I am unable to determine the photoelastic coefficients by using Eq. 4.7 alone, so I must take the ratios of different SBD patterns (Eq. 4.8) to measure \( p_{ijkl} \).

Figure 4.3 illustrates the experimental procedure for determining the independent photoelastic coefficients of fused silica. The figure illustrates two different SBD patterns taken with identical experimental conditions, except for the input and output optical polarizations are different. To
Figure 4.3: (left to right) Experimental set-up for recording the SBD, resulting SBD patterns, measured intensity along the SBD from longitudinal acoustic mode, and the measured ratio of $I_{VV}(\theta)/I_{HH}(\theta)$ in blue with the fit in red and green.

determine the photoelastic coefficients, I divide the measured intensities of the measured SBD patterns (i.e. $I_{VV}(\theta)/I_{HH}(\theta)$). Doing so eliminates the acoustic power $P(\theta)$ contribution because $P(\theta)$ is the same for the two measurements. Figure 4.3 shows the resulting data of $I_{VV}(\theta)/I_{HH}(\theta)$ (blue) and the corresponding fit (red and green) using Eqs. 4.8 and 4.9. The red points represent points that did not contribute to the fitting routine because the data were either saturating the camera or below the noise floor.

Figure 4.4 shows the data and corresponding fits for the ratios of $I_{\hat{V}\hat{V}}$, $I_{\hat{H}\hat{H}}$, and $I_{\hat{H}\hat{V}}$ with respect to each other. The measured ratio of the photoelastic coefficients for fused silica was $p_{12}/p_{11} = 2.24$, which agrees with the known values to within 0.4% and with the correct sign.

For the shear wave in isotropic materials that result in the outer SBD circle, all of the possible
effective photoelastic coefficients $p(\theta)$ are proportional to $p_{44}$ alone:

$$p_{\hat{H}\hat{H}} = 2p_{44} \cos(\theta) \sin(\theta)$$

$$p_{\hat{V}\hat{V}} = -2p_{44} \cos(\theta) \sin(\theta)$$

$$p_{\hat{H}\hat{V}} = p_{\hat{V}\hat{H}} = p_{44} \left[ \sin(\theta)^2 - \cos(\theta)^2 \right].$$

No information about the relative magnitudes of the photoelastic coefficients can be gained with the acoustic-shear wave because the photoelastic coefficients cancel out when taking the ratio of the different SBD patterns. The solutions do predict that maximum diffraction for $p_{\hat{H}\hat{H}}$ occurs at $\theta = 45^\circ \pm n90^\circ$, where $n$ is an integer. Figure 4.2b corroborates this prediction with four peaks in the intensity $I_{\hat{H}\hat{H}}(\theta)$ at $\theta = 45^\circ \pm n90^\circ$ in the outer circle that represents the shear wave. The peaks are narrower than expected from Eq. 4.10, implying that there may be a diagonal resonance in this geometry.

4.3.2 Optically Uniaxial Crystals: with $\alpha$-BaB$_2$O$_4$ as an example

Measuring the photoelastic coefficients is more difficult for uniaxial crystals than for isotropic crystals, which is best understood by looking at the SBD experiment in momentum space. For a
negative uniaxial crystal like $\alpha$-BBO, the double-sheeted optical momentum surface is an ellipsoid inside a sphere with the two surfaces touching along the z-axis. The ellipsoid and the sphere are commonly referred to as the extraordinary and ordinary momentum surfaces, respectively. The momentum surfaces represent the allowed modes of propagation in the crystal. Conservation of momentum can prevent optical diffraction between the two modes when the surfaces are separated by more than the length of the acoustic wavevector $\overline{K}_a$, which limits the number of independent data sets.

Figure 4.5 illustrates three different SBD experiments in momentum space. The dimensions of the acoustic momentum surfaces have been greatly exaggerated ($f_a = 0.5$ GHz) for elucidative purposes. The first experiment, Fig. 4.5a, is in fused silica where the optical momentum surface is a single, degenerate surface, so the optical eigenpolarizations lie in a plane orthogonal to the propagation of the optical beam. Thus $I^V \hat{V}$, $I^V \hat{H}$, and $I^H \hat{H}$ of identical SBD patterns can be measured while satisfying Bragg matching.

The SBD experiment illustrated in Fig. 4.5b is in $\alpha$-BBO with $\hat{z}$-polarized light propagating along the x-axis. Because the light is $\hat{z}$-polarized, the light is an allowed mode of propagation on the extraordinary momentum surface. Acoustic waves will diffract the incident light if the incident momentum vector $\overline{k}_i$ and the acoustic momentum vector $\overline{K}_a$ add to an allowed mode of propagation:

$$|\overline{k}_d| = \frac{2\pi n_o}{\lambda} = |\overline{k}_i \pm \overline{K}_a|,$$

(ordinary)

$$\left|\left(\frac{1}{n_e}, \frac{1}{n_e}, \frac{1}{n_o}\right) \cdot \overline{k}_d\right| = \frac{2\pi}{\lambda} = |\overline{k}_i \pm \overline{K}_a|/n_e(\theta),$$

(extraordinary) (4.11)

which is commonly referred to as Bragg matching. Figure 4.5b illustrates Bragg matching by centering the acoustic momentum surface on $\overline{k}_i$, which represents the summation of $\overline{k}_i$ with all possible $\overline{K}_a$. As the figure shows, the displaced momentum surfaces of the longitudinal and the fast quasi-shear mode never intersect with the ordinary-optical momentum surface; therefore, no light can diffract to the ordinary momentum surface from these acoustic modes because conservation of momentum would be violated. The slow quasi-shear mode, on the other hand, does intersect with the ordinary momentum surface. However, the intersection is not a central slice of the acoustic wavevector.
Figure 4.5: SBD measurements in momentum space. Left and right columns show the geometry of the experiments and the resulting SBD patterns with $f_a=0.5$ GHz, $\lambda = 532$ nm. The blue, dotted line in the YZ cross-section is from slow-shear acoustic mode diffracting the incident light to the ordinary-optical momentum surface.

momentum surface so the acoustic power of the resulting SBD pattern cannot be normalized by using the slow-shear SBD patterns of the isotropic diffraction. Therefore, for light propagating along the x-axis, which results in a YZ SBD pattern, only two independent data sets can be measured:
Figure 4.6: (top row) Experimental orientation of the BBO crystal, polarization of the transducer, and polarization of the incident and measured light for the YZ SBD patterns. (bottom row) Measured SBD patterns.

1) $\hat{y}$-to-$\hat{y}$ and 2) $\hat{z}$-to-$\hat{z}$ optical polarization diffraction. If the $\hat{x}$-propagating incident light were any other polarization, the data set would simply be a linear combination of the aforementioned two data sets.

The SBD experiment illustrated in Fig. 4.5c is for optical light propagating along the z-axis (also known as the optic axis). The z-axis is a point of degeneracy for uniaxial crystals without optical activity. For this SBD experiment, the two surfaces are so close that the incident optical light can diffract to either surface. Therefore, similar to the optically isotropic measurements, three independent data sets can be taken.

Figure 4.6 illustrates the experimental set-up and the measured SBD for the YZ cross-section
of α-BBO. As previously mentioned and illustrated in momentum space in Fig. 4.5b, the only two independent data sets for this experimental orientation are \( \hat{y} \rightarrow \hat{y} \) and \( \hat{z} \rightarrow \hat{z} \) optical diffraction. For both measurements, the orientation of the crystal and the acoustics within the crystal were identical. I used a shear transducer attached to the fused silica with the particle displacement along the y-axis to better populate the visible shear mode. Both the half-wave plate and the polarizer are adjusted so that the correct optical diffraction is measured. For both data sets, I measured the intensity of the diffracted light as function of the polar angle \( \theta \) along the two different visible acoustic modes in the SBD. The first two columns of Fig. 4.7 show the measured intensity of the different acoustic modes in the different data sets, and the third column shows in blue the ratio of the diffracted intensity of the \( \hat{y} \rightarrow \hat{y} \) with respect to the diffracted intensity of \( \hat{z} \rightarrow \hat{z} \) \( (I(\hat{y}\hat{y})/I(\hat{z}\hat{z})) \).

The photoelastic tensor written in reduced subscript notation for α-BBO is

\[
p_{IJ} = \begin{pmatrix}
p_{11} & p_{12} & p_{13} & p_{14} & 0 & 0 \\
p_{12} & p_{11} & p_{13} & -p_{14} & 0 & 0 \\
p_{31} & p_{31} & p_{33} & 0 & 0 & 0 \\
p_{41} & -p_{41} & 0 & p_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & p_{44} & p_{41} \\
0 & 0 & 0 & 0 & p_{14} & (p_{11} - p_{12})/2
\end{pmatrix}.
\]

Inserting this photoelastic tensor into Eq. 4.6 for the YZ cross-section experiment and allowing the strain tensor to be arbitrary results in

\[
p_{IJ}S_I = [p_{31}, p_{31}, p_{33}, 0, 0, 0] S_I,
\]

\[
p_{IJ}S_I = [p_{12}, p_{11}, p_{13}, -p_{14}, 0, 0] S_I,
\]

for \( \hat{z} \rightarrow \hat{z} \) and \( \hat{y} \rightarrow \hat{y} \) polarized light diffraction, respectively. These two rows of \( p_{IJ} \) reveal which photoelastic coefficients can be measured and whether or not the sign can be measured as well. For \( \hat{z} \)-polarized light propagating along \( \hat{x} \) and with acoustic waves in the YZ plane, the relative magnitudes and signs of \( p_{31} \) and \( p_{33} \) can be measured. For the \( \hat{y} \)-polarized light propagating along \( \hat{x} \), the relative magnitudes and signs of \( p_{11}, p_{13}, \) and \( p_{14} \) can be measured. The photoelastic coefficient
Figure 4.7: Measured intensity along the SBD for the longitudinal and shear modes in α-BBO for the YZ SBD patterns. The right column shows the data (blue) and fit (green and red) for $I(\hat{y}\hat{y})/I(\hat{z}\hat{z})$. The red diamonds represent points that do not contribute to the fit because the data for that point either saturated the camera or were below the noise floor.

$p_{12}$ cannot be measured because this would require acousto-optic diffraction from an x-propagating longitudinal acoustic wave, and the YZ experiment only measures acoustic waves propagating in the YZ plane. By taking the ratio of the two different SBD data sets (e.g. $I(\hat{y}\hat{y})/I(\hat{z}\hat{z})$), the relative magnitudes of 5 of the 8 independent photoelastic coefficients can be determined. However, the relative signs of $p_{31}$ and $p_{33}$ with respect to $p_{11}$, $p_{13}$, and $p_{14}$ cannot be determined because there is no overlap of these photoelastic coefficients in the two data sets. The green and red points in the third column of Fig. 4.7 represent the fit to the data using Eq. 4.8. The red points represent
Figure 4.8: (top row) Experimental orientation of the BBO crystal, polarization of the transducer, and polarization of the incident and measured light for the XZ SBD patterns. (bottom row) Measured SBD pattern.

points that did not contribute to the fitting routine because the data were either saturating the camera or below the noise floor.

The XZ cross-section measurement is very similar to the YZ measurement. With the XZ cross-section, the relative magnitudes of \( p_{31}, p_{33}, p_{11}, p_{13}, \) and \( p_{14} \) can be measured, but not the relative signs of \( p_{31} \) and \( p_{33} \) with respect to \( p_{11}, p_{13}, \) and \( p_{14} \). Unlike the YZ cross-section, the three different acoustic modes can be measured. However, only one of the shear modes is visible for a given angle because whichever mode is more \( \hat{y} \)-polarized is nearly invisible. I used a shear transducer polarized along the \( \hat{y} \)-axis, as illustrated in Fig. 4.8, to help make the \( \hat{y} \)-polarized shear mode for visible, but this did not improve the visibility enough to provide useful data. To clearly
Figure 4.9: Measured intensity along the SBD for the longitudinal and shear modes in $\alpha$-BBO for the XZ SBD patterns. The right column shows the data (blue) and fit (green and red) for $I^{XX}(\theta)/I^{ZZ}(\theta)$. The red diamonds represent points that do not contribute to the fit because the data for that point either saturated the camera or were below the noise floor.
measure both shear modes, I would have to saturate the camera, which would destroy useful data in the process. In Fig. 4.8, the black circles in the XZ SBD pattern label points of acoustic conical refraction. The acoustic conical refraction points mark a transition of which acoustic mode is more visible, so I mark these points to emphasize this transition. Figure 4.9 illustrates the measured intensity for the three acoustic modes for the two different data sets and the corresponding intensity ratios and fits.

For the XY measurements, the diffracted light was so weak compared to the DC, undiffracted light that the DC spot would saturate the CCD camera, even with a DC beam block, if I did not cross the polarizers. Therefore, I measured the AO diffraction of $\hat{x} \rightarrow \hat{y}$ and $\hat{y} \rightarrow \hat{x}$ polarized light. With these measurements, I was able to measure the $p_{11}, p_{12}, p_{14}, p_{41},$ and $p_{44}$ coefficients. The $p_{41}$ and $p_{44}$ are only measurable with the $\hat{z}$-polarization component of the diffracted light (the incident light does not have a $\hat{z}$-polarization component because it propagates along the $z$-axis). To solve for the $\hat{z}$-polarization component of the diffracted light, I numerically solved for the intersection of the acoustic momentum surface centered on $(0, 0, n_\omega k_0)$ of the optical momentum surface. However with the small AO frequencies, the $\hat{z}$-polarization component of the diffracted light is so small that the error bars of $p_{41}$ and $p_{44}$ are large. If I were to use a larger AO frequency, then the angle of the diffracted light would increase along with the $\hat{z}$-polarization component of the diffracted light. However, approximating the ordinary and extraordinary momentum surface as having the same shape for the data analysis breaks down for larger AO frequencies, making the results invalid.

Figure 4.10 illustrates the experimental set-up for the XY measurement. I used a longitudinal transducer along the $y$-axis of the BBO crystal to make the slow-shear acoustic mode more evenly populated with acoustic power. I only measured the intensity of the slow-shear acoustic mode because the fit to the faster acoustic modes were not nearly as sensitive to changes in the photoelastic coefficients as the slow-shear mode. The bottom row shows the measured intensities of the slow-shear mode for both SBD measurements along with their respective ratio. The results of the SBD measurements are listed in Table 4.1.
Figure 4.10: Experimental set-up and resulting measured SBD pattern for the XY cross-section. The bottom row illustrates the measured intensity of the slow-shear acoustic mode and the ratio of the measured intensities with the fit in green.
4.3.3 Dixon Method

The Dixon experiment [70] is a well established method in which the crystal to be measured is bonded to a reference crystal that is usually fused silica. Bonded to the reference cell is a transducer that launches a $\sim 1\mu s$ long acoustic pulse that diffracts the incident optical beam at different times and locations. Figure 4.11 illustrates the Dixon method with the horizontal axis in dimension of time to show the evolution of the acoustic pulse (red). Therefore, the slope of the acoustic pulse in the figure represents the velocity of the pulse. For instance, acoustic waves travel slower in BBO than in fused silica, so the slope of the acoustic pulse is flatter in BBO. As the acoustic pulse traverses through the crystals, the acoustic pulse will lose power as it is absorbed by the crystals and will split by reflecting and transmitting at the interface of the two crystals. The thickness of the line representing the acoustic pulse in Fig. 4.11 illustrates the acoustic power and can be seen.
Figure 4.12: Illustration of the propagation for each pulse in the Dixon calculation.

to diminish with propagation and each coupling.

The acoustic pulse diffracts light every time it passes through an incident optical beam. The diffracted light is measured by photo-detectors and are displayed on an oscilloscope. The magnitude of the diffracted light decreases with every successive pulse as the power of the acoustic pulse diminishes. However, the intensity of diffracted light from the reference sample may be larger despite having less acoustic power in the pulse if the effective photoelastic constant is sufficiently larger than the effective photoelastic constant of the reference \((p^{(s)} > \rho^{(r)})\). Figure 4.11 illustrates such a case.

Dixon showed that the square root of the ratio of the product of the diffracted light intensities of the first two pulses in the sample to be measured, \(I_{s1}^s \) and \(I_{s2}^s\), and the first diffracted pulse of the reference sample, \(I_{1r}^r\), with the diffracted light from the pulse to traverse both samples and return to fused silica, \(I_{rn}^r\), is proportional to the ratio of the respective effective photoelastic coefficients:

\[
\frac{I_{s1}^s I_{s2}^s}{I_{1r}^r} = \frac{\left(\rho \nu_3^{(s)/\rho V_a^3}\right)^{s}}{\left(\rho \nu_3^{(r)/\rho V_a^3}\right)}.
\]  

Equation 4.12 shows that the Dixon method eliminates the acoustic power density term \(P_a\), allowing for the calculation of the photoelastic coefficient.

The accuracy of the Dixon method comes from the elimination of the acoustic power term
$P_a$ and any linear acoustic loss terms associated with coupling loss of the piezoelectric transducer $C$, acoustic reflection $R$, transmission $T$, and absorption from traveling a distance $d$. Figure 4.12 illustrates the propagation path of the acoustic pulse of the four terms of the diffracted light in Eq. 4.12. Below I provide the loss $L$ of the acoustic power for each term:

$$L_1^r = Cd_1,$$
$$L_n^r = Cd_1d_2Td_3d_4Rd_4d_3Td_2,$$
$$L_1^s = Cd_1d_2Td_3,$$
$$L_2^s = Cd_1d_2Td_3d_4Rd_4.$$  \hspace{1cm} (4.13)

Multiplying the terms from Eq. 4.13 as they appear in Eq. 4.12, we see how the loss terms are eliminated:

$$\frac{L_1^s L_2^s}{L_1^r L_n^r} = \frac{(Cd_1)(Cd_1d_2Td_3d_4Rd_4d_3Td_2)}{(Cd_1d_2Td_3)(Cd_1d_2Td_3d_4Rd_4)} = 1 \hspace{1cm} (4.14)$$

With the aide of the above equation, the choice of the terms from Eq. 4.12 can be understood. Equation 4.12 does not include any loss from the optical beam from reflection. The inclusion of this optical loss may be necessary if the difference of the indices of refraction of the two samples is large since

$$R_{FS} = \left(\frac{n_{FS} - 1}{n_{FS} + 1}\right)^2,$$
$$R_{BBO} = \left(\frac{n_{o,e} - 1}{n_{o,e} + 1}\right)^2.$$  \hspace{1cm} (4.15)

To measure different photoelastic coefficients, the polarization and propagation direction of the optical beam must vary in the sample. Each measurement can measure the magnitude of the effective phototelastic coefficient $p$. The relative sign of the photoelastic coefficients can be measured if the effective phototelastic coefficient is a combination of more than one independent coefficients. With the crystographically cut $\alpha$-BBO crystal and longitudinal and shear transducers bonded to the fused silica crystals, I was able to measure 7 of the 8 independent coefficients, all but $p_{44}$, and the relative sign of $p_{11}$ with respect to $p_{12}$ and $p_{12}$ with respect to $p_{13}$. Table 4.1 lists the measured results.

### 4.4 Results

Table 4.1 lists the measured photoelastic coefficients from the SBD and Dixon experiments, as well as the photoelastic coefficients calculated from the piezo-optic coefficients measured by
Table 4.1: Measured photoelastic coefficients of $\alpha$-BBO using SBD, the Dixon method, and static stress.

<table>
<thead>
<tr>
<th></th>
<th>SBD</th>
<th>Dixon</th>
<th>Stress [14]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$p_{11}$</td>
<td>0.12±.02</td>
<td>+[0.12] ± .012</td>
<td>0.27±.14</td>
</tr>
<tr>
<td>$p_{33}$</td>
<td>[1+0.23] ± .02</td>
<td>[0.25] ± .02</td>
<td>-0.21±.10</td>
</tr>
<tr>
<td>$p_{44}$</td>
<td>0.2 ±0.2</td>
<td>-</td>
<td>-0.18±.09</td>
</tr>
<tr>
<td>$p_{12}$</td>
<td>0.23±.03</td>
<td>[0.20] ± .016</td>
<td>0.16 ±14</td>
</tr>
<tr>
<td>$p_{13}$</td>
<td>0.22±.02</td>
<td>[0.22] ± .012</td>
<td>-0.08±.09</td>
</tr>
<tr>
<td>$p_{14}$</td>
<td>-0.07±.003</td>
<td>[0.005] ± .009</td>
<td>-0.09±.02</td>
</tr>
<tr>
<td>$p_{31}$</td>
<td>[1+0.20] ± .02</td>
<td>[0.21] ± .014</td>
<td>0.07 ±15</td>
</tr>
<tr>
<td>$p_{41}$</td>
<td>-1±1.3</td>
<td>.02 ± .013</td>
<td>-0.14-0.24</td>
</tr>
</tbody>
</table>

Martynyuk-Lototska et al. [14] and my measured elastic-stiffness coefficients with SBD. For the SBD results, I set all the ratios relative to $p_{11} = 0.12$ for ease of comparison. The SBD results agree with the results from the established Dixon experiment, but not with the results measured from Martynyuk-Lototska.

4.5 Conclusion

I successfully demonstrated a new technique for measuring the photoelastic coefficients with SBD using both fused silica and $\alpha$-BBO as examples. For the SBD measurements, I had to average the SBD patterns over ten different frequencies to fill in the speckle of the SBD. A better transducer bonding scheme could better excite the acoustic modes to avoid the need for averaging the SBD patterns, and improve the data overall. My experimental set-up, however, allows for Dixon measurements that measure the absolute magnitude of the photoelastic coefficients and SBD measurements that allow for measurements of the elastic-stiffness coefficients and the relative ratios of the photoelastic coefficients. In addition, I can easily measure various crystals with my temporary bonding scheme.
Chapter 5

Maximum Acousto-Optic Figure of Merit Graphical Representation

The previous two chapters focus on measuring the necessary physical properties of a crystal for designing an acousto-optic (AO) device. This chapter and the next discuss how to design an AO device assuming that all of the required physical parameters are known. This chapter presents a graphical representation for discovering AO interaction geometries that maximize the diffraction efficiency of an AO device at a fixed optical wavelength \( \lambda \) and AO frequency \( f_a \) for isotropic and uniaxial media. Previously devices have been primarily designed by evaluating the AO interaction along principle symmetry axes and looking for good performance characteristics. Most interaction geometries are not evaluated, which likely leaves the optimum geometry unexplored. My algorithm works by sweeping through all possible Bragg matched geometries and for each geometry calculating the AO figure of merit \( M_2 \), which is proportional to the AO diffraction efficiency. I illustrate the results with \( M_2 \) surfaces that display the maximum \( M_2 \) value as a function of the acoustic wave propagation direction \( \hat{m} \) for the three different acoustic modes. These \( M_2 \) surfaces will change as \( \lambda \) and \( f_a \) change, which will be discussed in this chapter.

I applied this \( M_2 \) graphical representation to isotropic crystals Potassium Bromide (KBr) and Gallium Arsenide (GaAs), and uniaxial crystal Lithium niobate (LiNbO\(_3\)). Appendix B presents the \( M_2 \) graphical representations to Tellurium Dioxide (TeO\(_2\)), Potassium Dideuterium (KDP), and Lead Molybdate (MoO\(_4\)Pb). The results show that the AO diffraction efficiency is very anisotropic with respect to the propagation direction of the acoustic wave with the peak values sometimes occurring at complicated, off-axes angles for the acoustic and optical waves.
5.1 Acousto-optic figure of merit

There are several different types of acousto-optic figures of merit (AOFM), but this chapter focuses on the $M_2$ AOFM which is a metric for the diffraction efficiency of an AO deflector in a given material [71, 72]. This graphical method for finding the maximum $M_2$ value, however, can be applied to any other AOFM.

The $M_2$ AOFM is defined as

$$M_2 = \frac{n_i^3(\lambda)n_d^3(\lambda)p^2}{\rho V_a(\phi, \theta)^3}.$$  \hspace{1cm} (5.1)

Here $n_i(\lambda)$ and $n_d(\lambda)$ are the index of refraction of the incident and diffracted light, $\rho$ is the medium density, $V_a(\phi, \theta)$ is the acoustic wave velocity, and $p$ is the effective photoelastic coefficient. Chapter 2 discusses how to determine both $V_a(\phi, \theta)$ and $p$.

In a piezoelectric material, the strain from an acoustic wave can induce a change in the electric field, which will then change the index of refraction through the electro-optic effect. This effect modifies the photoelastic tensor by

$$\tilde{p}_{ijkl} = p_{ijkl} - \frac{r_{ijp}m_pm_qe_{qkl}}{\epsilon_{jk}m_jm_k}.$$  \hspace{1cm} (5.2)

Here $p_{ijkl}$ is the fourth rank photoelastic tensor and $r_{ijp}$ is the third rank electro-optic tensor. Similarly, the piezoelectric effect stiffens the elastic-stiffness coefficients $C_{ijkl}$, thus increasing the acoustic velocity of the acoustic wave $V_a$:

$$\left( C_{ijkl} + \frac{e_{ijp}m_pm_qe_{qkl}}{\epsilon_{jk}m_jm_k} \right)m_jm_kU_i = \rho V_a^2 U_i.$$  \hspace{1cm} (5.3)

Here $m_i$ is the propagation direction of the acoustic wave, $C_{ijkl}$ is the forth rank elastic-stiffness tensor, $e_{ijp}$ is the third rank piezoelectric stress tensor, $\epsilon_{ij}$ is the second rank relative permittivity tensor, and $U_i$ is the particle displacement of the acoustic wave. For piezoelectric media, the change to both $V_a$ and $p$ from the piezoelectric effect must be accounted for to accurately calculate $M_2$. 
5.2 Bragg matching

The figure of merit calculation depends on the geometry of the acoustic wave and the incident and diffracted optical waves. I restrict the search to geometries that are exactly Bragg matched. Below I discuss my motivation for considering only Bragg matched geometries and how I solve for the Bragg matched geometries in a given medium.

In AO devices the acoustic wave diffracts the incident light by exchanging momentum with the incident light:

$$\vec{k}_d = \vec{k}_i \pm \vec{K}_a.$$  

Here $\vec{K}_a$ is the acoustic wavevector, $\vec{k}_d$ and $\vec{k}_i$ are the diffracted and incident light wavevectors respectively, and the $+$ and $-$ signs correspond to Doppler up-shifting and down-shifting interactions with optical angular radian frequencies $\omega_d = \omega_i \pm \Omega$. Momentum mismatch $\Delta \vec{k}$ of the AO interaction is often considered to arise when the wavevectors of the incident light and the acoustic wave do not sum to a wavevector that is a solution of the optical wave equation. In a finite medium, an alternative viewpoint is to ascribe a momentum uncertainty $\Delta k = \frac{2\pi}{L}$, due to the finite length of the interaction that corresponds to the width of the distribution of Fourier components of the acoustic wave along the direction of optical propagation in momentum space [22, 31]. The intensity diffraction efficiency $\eta$ scales as

$$\eta \sim \text{sinc}^2 \left( \frac{\Delta k L}{2\pi} \right),$$  

where $L$ is the AO interaction length. As Eq. 5.4 shows, the largest diffraction efficiency $\eta$ occurs when the wavevectors are Bragg matched ($|\Delta k| = 0$). For my $M_2$ calculations, I only consider Bragg matched AO geometries because I am searching for the AO geometry with the largest diffraction efficiency. I solve for Bragg matched geometries by using $k$-surfaces as a geometrical tool.

Figure 5.1a illustrates in momentum space a Bragg matched AO interaction in the XZ cross-section of a negative uniaxial crystal. An acoustic wave, represented by the green momentum vector $\vec{K}_a$, deflects an incident optical beam $\vec{k}_i$ from the extraordinary optical momentum surface to create the diffracted beam $\vec{k}_d$ on the ordinary momentum surface. The AO interaction illustrated in Fig.
Figure 5.1: (a) Bragg matching acousto-optic interaction illustrated as a $\vec{k}$-vector triangle in a 2D plane. (b) Solving for the Bragg matching geometry by solving for the intersection of the diffracted $\vec{k}$-surface (blue) with the displaced, incident $\vec{k}$-surface (dashed red) displaced by a given acoustic wavevector $\vec{K}_a$ (green). (c) 3D illustration of the intersection of a sphere (blue) with a displaced ellipsoid (red), showing the set of Bragg matched solutions.

5.1a is perfectly Bragg matched because the acoustic wavevector $\vec{K}_a$ and the incident wavevector $\vec{k}_i$ sum to a diffracted wavevector $\vec{k}_d$ that lies exactly on the diffracted $\vec{k}$-surface.

To solve for a Bragg matched geometry, I first solve for the incident and diffracted $\vec{k}$-surfaces. Next I displace the incident $\vec{k}$-surface by the acoustic wavevector $\vec{K}_a$. The intersections of the displaced incident $\vec{k}$-surface with the diffracted $\vec{k}$-surface are possible solutions for the perfectly Bragg matched diffracted wavevector $\vec{k}_d$ for the given $\vec{K}_a$. The incident wavevector $\vec{k}_i$ is easily solved for by subtracting the acoustic wavevector $\vec{K}_a$ from the diffracted wavevector $\vec{k}_d$. Figure 5.1b illustrates a 2D cross-section of the incident $\vec{k}$-surface being displaced by the acoustic wavevector $\vec{K}_a$, and the solution for the exact Bragg matched diffraction occurring where the displaced $\vec{k}$-surface intersects with the non-displaced, diffracted $\vec{k}$-surface. For this 2D cross-section, there are two different points of intersection and therefore two different solutions. In 3D the locus of solutions forms a curve known as the Bragg degeneracies.

The advantage of displacing the optical momentum surfaces to solve for the Bragg matched geometries is that the intersection of the acoustic momentum surface with an optical momentum surface does not need to be solved. The acoustic momentum surfaces are more complicated and
rarely have an analytic solution for the three-dimensional surface. Therefore, analytic solutions of the intersection of the acoustic and optical momentum surfaces do no exist. Recently, the intersection of acoustic and optical momentum surfaces has been solved for numerically [73] and can be viewed as an alternative approach to the algorithm presented here, but their algorithm is computationally more expensive.

For uniaxial crystals the incident optical wave can diffract in four different ways: 1) $\hat{o} \rightarrow \hat{o}$, 2) $\hat{e} \rightarrow \hat{e}$, 3) $\hat{e} \rightarrow \hat{o}$, and 4) $\hat{o} \rightarrow \hat{e}$. Here $\hat{o}$ and $\hat{e}$ refer to ordinary and extraordinary optical polarizations of the incident and diffracted waves. Cases 3 and 4 result in identical $M_2$ calculations, so I will only focus on cases 1-3 even though $\hat{e} \rightarrow \hat{o}$ and $\hat{o} \rightarrow \hat{e}$ have different AO bandwidths referred to as tangential matching for conventional wideband birefringent diffraction and anti-tangential matching for narrow band diffraction [72, 71]. To solve for the Bragg matched geometries in three dimensions for $\hat{o} \rightarrow \hat{o}$, $\hat{e} \rightarrow \hat{e}$, and the very important anisotropic polarization-switching $\hat{e} \rightarrow \hat{o}$ AO diffraction, the intersections of a sphere with a sphere, an ellipsoid with an ellipsoid, and an ellipsoid with a sphere need to be solved. Such quartic intersection problems have been solved numerically in a very general context for computer graphics [74, 75, 76], but I have solved for the special cases of optical momentum surfaces for isotropic and uniaxial crystals.

5.3 Solving for Intersecting Quadrics

I solve for AO Bragg matched geometries by solving for the intersection of two optical momentum surfaces with one momentum surface displaced from the origin by the acoustic wavevector $\vec{K}_a$. To solve for the intersections, I first represent the surfaces in tensor form. Next I find a degenerate, simpler surface that shares the intersection of the two momentum surfaces. Lastly, I solve for the intersection of the non-displaced momentum surface with the degenerate solution.

A three-dimensional ellipsoid can be written algebraically as

$$Ax^2 + By^2 + Cz^2 + 2Dxy + 2Eyz + 2Fxz + 2Gx + 2Hy + 2Jz + K = 0, \quad (5.5)$$

Here $A, B, C, D, E, F, G, H, J,$ and $K$ are all constants. The constants $A, B,$ and $C$ describe
the quadratic curvatures along the x, y, and z axes, and $D$, $E$, and $F$ are related to the tilts in the x, y, and z planes. $G$, $H$, and $J$ create shifts in the x, y, and z directions. The shifts are given by

$$
\begin{pmatrix}
A & D & F \\
D & B & E \\
F & E & C \\
\end{pmatrix}^{-1}
\begin{pmatrix}
G \\
H \\
J \\
\end{pmatrix} =
\begin{pmatrix}
x_d \\
y_d \\
z_d \\
\end{pmatrix}.
$$

(5.6)

Lastly, $K$ is a scaling factor that helps determine the size of the surface.

Equation 5.5 can be written in tensor form, commonly known as the augmented quadratic form,

$$
\bar{X} \bar{S} \bar{X}^T = 0,
$$

(5.7)

where,

$$
\bar{X} = \begin{pmatrix} x & y & z & 1 \end{pmatrix}, \quad \bar{S} = \begin{pmatrix}
A & D & F & G \\
D & B & E & H \\
F & E & C & J \\
G & H & J & K \\
\end{pmatrix}.
$$

(5.8)

Most of my analysis uses the tensor form, but understanding the connection between the algebraic and tensor forms is useful.

### 5.3.1 Degenerate Surfaces

Given two intersecting quadrics described by matrices $\bar{S}_1$ and $\bar{S}_2$, any point $\bar{X}_p$ on their intersection must satisfy both $\bar{X}_p \bar{S}_1 \bar{X}_p^T = 0$ and $\bar{X}_p \bar{S}_2 \bar{X}_p^T = 0$. Any linear combination of $\bar{S}_1$ and $\bar{S}_2$ will satisfy $\alpha \bar{X}_p \bar{S}_1 \bar{X}_p^T + \beta \bar{X}_p \bar{S}_2 \bar{X}_p^T = 0$, which can be written as

$$
\bar{X}_p (\bar{S}_1 + \gamma \bar{S}_2) \bar{X}_p^T = 0,
$$

(5.9)

Here $\gamma = \beta/\alpha$. I can solve for $\gamma$ such that the determinant of the term in the parenthesis equals zero:

$$
|S_1 + \gamma S_2| = 0.
$$

(5.10)
Solving Eq. 5.10 for $\gamma$ will result in a degenerate quadric, $\bar{S}_D = \bar{S}_1 + \gamma \bar{S}_2$, that will share the intersection of $\bar{S}_1$ and $\bar{S}_2$. Using the degenerate surface $\bar{S}_D$ to solve for the intersection of $\bar{S}_1$ or $\bar{S}_2$ simplifies the process because the more complicated surface $\bar{S}_1$ or $\bar{S}_2$ can be replaced with $\bar{S}_D$.

### 5.3.2 Ellipsoid-Ellipsoid Intersections

To solve for $\hat{o} \rightarrow \hat{o}$ and $\hat{e} \rightarrow \hat{e}$ diffraction, the intersection of two identical ellipsoids with one ellipsoid displaced from the origin needs to be solved. The ellipsoid at the origin $\bar{S}_{EO}$ and the ellipsoid displaced from the origin $\bar{S}_{ED}$ are described by

\[
\bar{S}_{EO} = \begin{pmatrix}
1/n_e^2 & 0 & 0 & 0 \\
0 & 1/n_e^2 & 0 & 0 \\
0 & 0 & 1/n_o^2 & 0 \\
0 & 0 & 0 & -1
\end{pmatrix}, 
\]

\[\tag{5.11}\]

\[
\bar{S}_{ED} = \begin{pmatrix}
1/n_e^2 & 0 & 0 & -x_d/n_e^2 \\
0 & 1/n_e^2 & 0 & -y_d/n_e^2 \\
0 & 0 & 1/n_o^2 & -z_d/n_o^2 \\
-x_d/n_e^2 & -y_d/n_e^2 & -z_d/n_o^2 & -1 + \frac{x_d^2+y_d^2}{n_e^2} + \frac{z_d^2}{n_o^2}
\end{pmatrix}
\]

\[\tag{5.12}\]

Here $x_d$, $y_d$, and $z_d$ are the displacements of the ellipsoid from the origin in the x, y, and z directions, respectively. These displacements are related to the components of the acoustic wavevector that diffracts the incident optical beam by $x_d = K_z A \lambda / 2\pi$, $y_d = K_y A \lambda / 2\pi$, and $z_d = K_z A \lambda / 2\pi$, where $\lambda$ is the incident light wavelength and the scaling factor $\lambda / 2\pi$ transforms $\bar{K}_a$ from momentum space to optical index space. The ellipsoids have two radii of equivalent length, which is the case for the extraordinary momentum surfaces in uniaxial crystals. The momentum surface for the ordinary polarization and isotropic crystals reduces to a sphere (except for gyrotropic media, but even in this case a spherical approximation is often used [25]).

Solving Eq. 5.10 for $\bar{S}_{EO}$ and $\bar{S}_{ED}$ results in a solution where $\gamma = -1$, which creates the
Figure 5.2: Pictures of an ellipsoid (green) displaced by \( x_d = 0.1, y_d = 0.05, \) and \( z_d = 0.21 \) intersecting with an identical ellipsoid (blue) centered at the origin \( (n_e = 1.5, n_o = 1.9) \). (a) Initial problem, (b) planar degenerate surface with the two ellipsoids, and (c) solutions of the intersection.

The degenerate surface is given by:

\[
\vec{S}_D = \begin{pmatrix}
0 & 0 & 0 & \frac{x_d}{n_e^2} \\
0 & 0 & 0 & \frac{y_d}{n_e^2} \\
0 & 0 & 0 & \frac{z_d}{n_o^2}
\end{pmatrix},
\]

(5.13)

I can rewrite \( \vec{S}_D \) in algebraic form:

\[
\frac{x_d}{n_e^2} x + \frac{y_d}{n_e^2} y + \frac{z_d}{n_o^2} z = \frac{1}{2} \left( \frac{x_d^2 + y_d^2}{n_e^2} + \frac{z_d^2}{n_o^2} \right).
\]

(5.14)

Since there are no quadratic terms (the first three diagonal components of \( \vec{S}_D \) equal 0) in Eqs. 5.13 and 5.14, the degenerate surface \( \vec{S}_D \) from the intersection of the two ellipsoids \( \vec{S}_{EO} \) and \( \vec{S}_{ED} \) is a plane. This plane intersects with the two ellipsoids and cuts out a planar ellipse. Figure 5.2 illustrates the two ellipsoids \( \vec{S}_{EO} \) and \( \vec{S}_{ED} \) with the corresponding degenerate surface \( \vec{S}_D \).

I solve for the intersection of two ellipsoids by first solving for the special case of a sphere intersecting with a sphere that is identical in dimensions, but displaced from the origin by \( \vec{d} = (x_d, y_d, z_d) \). The resulting degenerate surface is a plane described by the vector \( \vec{p} = \frac{1}{\sqrt{x_d^2 + y_d^2 + z_d^2}} (x_d, y_d, z_d) \) and displaced from the origin by \( \vec{d}_o = \frac{|\vec{d}|}{2} \vec{p} \) (halfway between the centers of the two spheres). The inter-
section of the two spheres is a circle with a radius of \( r = \sqrt{1 - \frac{x_d^2 + y_d^2 + z_d^2}{4n_e^2}} \). I calculate the solutions by parametrically sweeping over the circle:

\[
\vec{c}(s) = r(\hat{r}_1 \cos s + \hat{r}_2 \sin s) + \vec{d}_o. 
\]  (5.15)

Here \( \vec{c}(s) \) is the solution of the intersection, \( s \) is a parametric variable, and \( \hat{r}_1 \) and \( \hat{r}_2 \) are two orthogonal vectors that span the plane \( \vec{S}_{ED} \).

In momentum space, the intersection describes a set of Bragg matched diffracted optical wavevectors \( \vec{k}_d(s) \) for a given acoustic wavevector \( \vec{K}_a = \vec{d} \). The incident wavevectors \( \vec{k}_i(s) \) are solved for by taking the difference of the diffracted and acoustic wavevectors: \( \vec{k}_i(s) = \vec{k}_d(s) - \vec{K}_a \).

The Bragg matched geometries are Bragg degenerate and can all be addressed by rotating an AO device about the acoustic wavevector \( \vec{K}_a \).

I can extrapolate the intersecting spheres solution to intersecting ellipsoids by applying appropriate scaling factors to transform the ellipsoids to spheres, solve for the intersection of the two spheres, and then back transform the solutions to the original coordinate system. This solution scheme only works because the ellipsoids are equivalent in dimension and are not rotated with respect to each other or the principal axes. The displacement of \( \vec{S}_{ED} \) does not create any issues, although \( \vec{K}_a \) must be appropriately scaled.

By scaling the \( x, y, \) and \( z \) axes by \( \frac{1}{n_e}, \frac{1}{n_e}, \) and \( \frac{1}{n_o} \) respectively, the ellipsoids can be transformed to spheres. With this transformation, the displacement of the plane from the origin \( \vec{d}_o \) and the radius of the intersecting circle \( r \) become

\[
\vec{d}_o = \frac{1}{2}(\frac{x_d}{n_e} \frac{y_d}{n_e} \frac{z_d}{n_o}), 
\]  (5.16)

\[
r = \sqrt{1 - \frac{x_d^2/n_e^2 + y_d^2/n_e^2 + z_d^2/n_o^2}{4}}. 
\]  (5.17)

Equation 5.15 can now be applied to solve for the intersection \( \vec{c}(s) \). These solutions are then back transformed by multiplying each point by the inverse of the original scaling factor: \( (n_e, n_e, n_o) \).

The intersecting ellipsoids represent Bragg matched solutions for \( \vec{e} \rightarrow \vec{e} \) diffraction in uniaxial crystals. In addition, the solutions can work for gyrotropic crystals if the momentum surfaces are
approximated as ellipsoids (see Appendix B). Figure 5.2 illustrates the solution of the intersection in magenta for two ellipsoids equivalent in dimension but with one of the ellipsoids displaced from the origin.

5.3.3 Sphere-Ellipsoid Intersection

For \( \hat{o} \rightarrow \hat{e} \) or \( \hat{e} \rightarrow \hat{o} \) optical diffraction, the intersection of a sphere centered at the origin with an ellipsoid displaced from the origin must be solved. The ellipsoid is defined by Eq. 5.12 and the sphere by

\[
\bar{S}_{SO} = \begin{pmatrix}
\frac{1}{n_o^2} & 0 & 0 & 0 \\
0 & \frac{1}{n_o^2} & 0 & 0 \\
0 & 0 & \frac{1}{n_o^2} & 0 \\
0 & 0 & 0 & -1
\end{pmatrix}.
\]

This special case where both \( \bar{S}_{SO} \) and \( \bar{S}_{ED} \) share the same \( A \) and \( B \) coefficients \( (1/n_o^2) \) is the relevant case for uniaxial crystals, but much simpler than the general sphere-ellipsoid intersection problem. As before, I solve for the degenerate surface (Eq. 5.10). One solution to Eq. 5.10 is \( \gamma = -\frac{n_e^2}{n_o^2} \), which results in the following degenerate surface:

\[
\bar{S}_D = \begin{pmatrix}
0 & 0 & 0 & x_d/n_o^2 \\
0 & 0 & 0 & y_d/n_o^2 \\
x_d/n_o^2 & y_d/n_o^2 & \frac{z_d n_e^2}{n_o^4} & \frac{z_d n_e^2}{n_o^4} - \frac{n_e^2 x_d^2 - y_d^2}{n_o^4} - \frac{z_d^2 n_e^2}{n_o^4}
\end{pmatrix}.
\]

The degenerate surface \( \bar{S}_D \) is somewhat simpler than \( \bar{S}_{SO} \) and \( \bar{S}_{ED} \) because \( \bar{S}_D \) has no \( x^2 \) or \( y^2 \) dependence; hence, there is only curvature in \( z \) (like a bent piece of paper). The ellipsoid \( \bar{S}_{ED} \) is the most complicated surface because it is displaced from the origin and has less symmetry than the sphere. Therefore, I solve for the intersection between the sphere \( \bar{S}_{SO} \) and the degenerate surface \( \bar{S}_D \). As discussed earlier, solving for the intersection of the sphere \( \bar{S}_{SO} \) with the degenerate surface \( \bar{S}_D \) is identical to solving for the intersection of the sphere \( \bar{S}_{SO} \) with the ellipsoid \( \bar{S}_{ED} \).

Equations 5.18-5.19 when inserted into Eq. 5.7 provide two equations with three unknowns. I solve for \( x \) and \( y \) in terms of \( z \) because \( \bar{S}_D \) is linear with respect to \( x \) and \( y \), but quadratic with
respect to $z$:

\[
x = \frac{Lx_d \pm \sqrt{M}}{N}, \quad y = \frac{Ly_d \pm \sqrt{Mx_d/y_d}}{N},
\]

\[L = n_0^6 + n_0^4 (x_d^2 + y_d^2 - z^2 - n_0^2) + n_0^2 n_o^2 (z - z_d)^2,
\]

\[M = -n_0^4 y_d (n_0^4 (x_d^2 + y_d^2 + z^2 - n_0^2)^2 + n_0^4 (n_0^2 - (z - z_d)^2)^2
\]

\[\quad - 2 n_0^2 n_o^2 (n_0^2 + x_d^2 + y_d^2 - z^2)(n_0 + z - z_d)(n_o - z + z_d)),
\]

\[N = 2n_0^4 (x_d^2 + y_d^2).
\]

Figure 5.3 illustrates my code solving an intersection of an ellipsoid with a sphere.

Equation 5.20 will break down under certain conditions. For instance if both $x_d$ and $y_d$ equal 0, then $N = 0$ and the solutions for $x$ and $y$ will go to infinity. However when $x_d$ and $y_d$ equal 0, the solution simplifies to

\[z = \frac{n_o^2 z_d - \text{Sign}(z_d) \sqrt{n_0^2 (n_0^4 + n_0^4 + n_0^2 (z_d^2 - 2n_o^2))}}{n_0^2 - n_o^2}, \]

\[\frac{x^2}{n_o^2} + \frac{y^2}{n_o^2} = 1 - \frac{z^2}{n_o^2},
\]

which is a circle in the xy-plane for a constant value of $z$ as determined by Eq. 5.24. If $x_d \neq 0$ but $y_d = 0$, then the $y$-solutions for Eq. 5.20 will still go to infinity. This problem can be avoided by interchanging the $x$ and $y$ solutions in Eq. 5.20 while interchanging $x_d$ for $y_d$.

5.4 $M_2$ Search Algorithm

With Bragg matched solutions, I can search for the maximum $M_2$ value in a given material for a fixed $\lambda$ and $f_\alpha$. My graphical representation sweeps in spherical coordinates ($\phi, \theta$) through all possible propagation directions of the acoustic wave $\hat{m}$. However, when symmetry permits, I reduce the search to avoid repeating redundant calculations. For instance if a crystal possesses a mirror plane symmetry, then the calculations can be reduced by a factor of two because the results will repeat themselves on the other side of the mirror plane.

For every propagation direction, there are three different acoustic modes that I label $\alpha = 1, 2, \text{and} 3$ for the quasi-longitudinal (QL), fast quasi-shear (QFS), and slow quasi-shear (QSS) modes,
respectively. Each mode has a corresponding velocity $V_\alpha^a$, particle displacement vector $U_\alpha^i$, strain $S_{ij}^\alpha$, and local rotation $\omega_{ij}^\alpha$. From the velocity of the mode $V_\alpha^a$, I calculate the Bragg matched solutions for each of the three different types of optical diffraction that I label $\beta = 1, 2, \text{and } 3$ for $\hat{o} \rightarrow \hat{o}$, $\hat{e} \rightarrow \hat{e}$, and $\hat{e} \rightarrow \hat{o}$ diffraction, respectively. For a given direction $\hat{m}$, there are 9 different $M_{2}^{\alpha,\beta}$ calculations because there are three different acoustic modes that each can cause three different types of AO diffraction. Below is a summary of my algorithm:

- **For-loop**: Sweep in spherical coordinates through azimuthal angle $\phi$ and polar angle $\theta$
  - Calculate the acoustic velocity $V_\alpha^a(\phi, \theta)$ with Eq. 3.10 for the three modes
  - Calculate the acoustic wavevector $\bar{K}_\alpha^a$ for each mode indexed by $\alpha$:
    $$\bar{K}_\alpha^a(\phi, \theta) = \frac{2\pi}{V_\alpha^a(\phi, \theta)} f_a$$
  - Calculate Bragg matched solutions for each type of AO diffraction
- **For-loop**: Sweep parametrically through Bragg degenerate solutions indexed by $s$
  - Calculate $M_{2}^{\alpha,\beta}(\phi, \theta, s)$
- **End For-loop**
* Store \( \text{Max} \left( M_2^{\alpha,\beta}(\phi, \theta, s) \right) \) for each diffraction type

- **End For-loop** Plot \( M_2^{\alpha,\beta}(\phi, \theta) \) and print max \( M_2 \) values for each diffraction type

In the following subsections I present a few examples of my graphical representation for isotropic and uniaxial crystals. For each result, I use a one degree resolution when sweeping in spherical coordinates through all of the possible propagation directions of the acoustic wave.

There are several other search methods for maximizing \( M_2 \) [73, 77, 78, 79]. Of these methods, Buryy et al. [73] provide the most exhaustive \( M_2 \) graphical representation. The method from Buryy et al. [73] is similar to ours, but Buryy et al. solve for Bragg matching by first choosing the incident optical wavevector \( \vec{k}_i \), then centering the acoustic momentum surface at the tip of \( \vec{k}_i \), and then numerically solving with Dragilev’s method for the intersection of the displaced acoustic momentum surface with an optical momentum surface. The intersection describes the acoustic momentum wavevectors \( \vec{K}_a \) and diffracted optical wavevectors \( \vec{k}_d \) that satisfy Bragg matching for a given \( \vec{k}_i \), and is exactly the SBD pattern for that AO geometry. My method for solving for Bragg matching uses analytic solutions; thus, is computationally less expensive and easier to implement.

5.4.1 Isotropic Crystals

The optical momentum surface for isotropic media, as discussed in Chapter 2, is a degenerate sphere. With a single-sheeted-momentum surface, only one type of Bragg matching (a sphere with an identical sphere) needs to be solved. However, the polarizations for the optical vectors are not well defined (unlike the case for uniaxial media). The eigenpolarizations of the electric flux \( \vec{D} \) lie in a plane tangent to the momentum surface, which is orthogonal to the propagation direction of the optical wave \( \hat{k} \).

To find the maximum \( M_2 \) value for a given AO geometry, the effective photoelastic coefficient \( p \) must be maximized by trying all the possible combinations of the optical eigenpolarizations for both the incident and diffracted optical waves. The effective photoelastic coefficient is determined
by
\[ p = \hat{d}_d \cdot \vec{p} \cdot \vec{S} \cdot \hat{d}_i. \]  

(5.26)

I find the maximum photoelastic coefficient for a given AO geometry by sweeping in the plane
tangent to \( \hat{k}_i \) for all possible values of polarization of the incident wave \( \hat{d}_i \). I then determine how
\( \hat{d}_i \) transforms after multiplication with \( \vec{p} \) and \( \vec{S} \):
\[ \vec{t} = \vec{p} \cdot \vec{S} \cdot \hat{d}_i. \]

Next I determine the polarization of the diffracted wave \( \hat{d}_d \) that maximizes \( p \) by projecting \( \vec{t} \) onto
two orthogonal vectors \( \hat{d}_1 \) and \( \hat{d}_2 \) that span the plane orthogonal to \( \hat{k}_d \):
\[ \hat{d}_d = \hat{d}_1 (\hat{d}_1^* \cdot \vec{t}) + \hat{d}_2 (\hat{d}_2^* \cdot \vec{t}). \]  

(5.27)

Solving for \( \hat{d}_d \) with Eq. 5.27 avoids the need, when determining the maximum \( p \) for a given AO
geometry, to sweep through all possible \( \hat{d}_d \) for every \( \hat{d}_i \); thus, saving computational time.

I have applied the \( M_2 \) search algorithm for optically isotropic media to both Potassium
Bromide (KBr) and Gallium Arsenide (GaAs). Table 5.2 lists the pertinent physical properties and
Fig. 5.4 illustrates the acoustic-slowness surfaces for both crystals.

KBr is a salt with a large optical transmission window of \( 0.2 - 30 \) \( \mu \)m, which allows for AO
devices operating in a wide wavelength range. However, KBr is so soft, a Mohs hardness of 1.5,
that KBr devices are difficult to polish and are easily scratched. In addition, KBr is hygroscopic,
so a KBr device might deteriorate in quality in humid climates.

Table 5.1 lists the maximum \( M_2 \) values and Fig. 5.5 illustrates the \( M_2 \) surfaces for the three
different acoustic modes in KBr for \( \lambda = 589 \) nm and \( f_a = 80 \) MHz. The QL mode for KBr has the
maximum \( M_2 \) value despite being the fastest mode because the photoelastic coefficients associated
with longitudinal acoustic modes \( (p_{11} \text{ and } p_{12}) \) are much larger than the photoelastic coefficients
associated with the shear modes \( (p_{44}) \). The QL \( M_2 \) surface looks very similar in shape to the
QL slowness surface illustrated in Fig. 5.4, meaning that \( p(\phi, \theta) \) is very isotropic. The shear \( M_2 \)
surfaces, on the other hand, display a greater asymmetry than seen in the slowness surfaces. In
addition the maximum $M_2$ values do not occur in the direction of the slowest acoustic waves, and require a graphical $M_2$ representation to find the peak $M_2$ values. All three $M_2$ surfaces, along with the slowness surfaces, display the cubic symmetry of the crystal as they must. The many equivalent maximum $M_2$ geometries can be selected between by choosing the geometry that results in the simplest and easiest crystal cut and orientation.

GaAs is a versatile, piezoelectric crystal manufactured for integrated circuits, infrared light-emitting diodes, and solar cells. In addition, GaAs has a large $M_2$ value of $86 \times 10^{-15} s^3/kg$ that comes from the large index of refraction of GaAs ($M_2 \sim n^6$). However, a large index of refraction implies large optical reflection at the air-GaAs interface; thus, decreasing the throughput of the device. An anti-reflection coating can be applied to the crystal to mitigate optical loss to reflection.

Similar to KBr, the QL mode $M_2$ and slowness surfaces look similar in GaAs, but the shear mode surfaces differ considerably. In addition, the QL has the largest $M_2$ values because $p_{11}$ and $p_{12}$ are about three times larger than $p_{44}$.
Figure 5.5: $M_2$ surfaces in units of $10^{-15}s^3/kg$ for optically isotropic KBr (left column) and GaAs (right column). The red, blue, and green surfaces represent AO diffraction from the quasi-longitudinal, fast quasi-shear, and slow quasi-shear acoustic modes, respectively.
Table 5.1: Maximum $M_2$ values and corresponding acousto-optic geometries for KBr and GaAs.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$\bar{k}_i$</th>
<th>$\bar{k}_d$</th>
<th>$K_a$</th>
<th>$M_2^{\text{max}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\phi$</td>
<td>$\theta$</td>
<td>$\phi$</td>
<td>$\theta$</td>
</tr>
<tr>
<td>KBr ($\lambda = 532$ nm, $f_a = 80$ MHz)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>-135.00</td>
<td>29.70</td>
<td>-135.00</td>
<td>30.30</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>10.96</td>
<td>111.08</td>
<td>10.65</td>
<td>112.02</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>134.48</td>
<td>89.85</td>
<td>135.53</td>
<td>90.15</td>
</tr>
<tr>
<td>GaAs ($\lambda = 1150$ nm, $f_a = 80$ MHz)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>134.95</td>
<td>89.86</td>
<td>135.06</td>
<td>90.14</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>134.77</td>
<td>89.92</td>
<td>135.23</td>
<td>90.08</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>89.76</td>
<td>89.93</td>
<td>90.24</td>
<td>90.07</td>
</tr>
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</table>

Table 5.2: Physical properties of KBr [80] and GaAs [81].

<table>
<thead>
<tr>
<th>Material</th>
<th>Point Group</th>
<th>$\rho$ (g/cm$^3$)</th>
<th>Mohs Hardness</th>
<th>$c_{11}$</th>
<th>$c_{12}$</th>
<th>$c_{44}$</th>
<th>$p_{11}$</th>
<th>$p_{12}$</th>
<th>$p_{44}$</th>
<th>$\epsilon_{11}$</th>
<th>$\epsilon_{14}$</th>
<th>$r_{14}$</th>
<th>Transparency</th>
</tr>
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<tbody>
<tr>
<td>KBr</td>
<td>$m3m$</td>
<td>3.46</td>
<td>2.750</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>5.15</td>
<td>5.6</td>
<td>-0.023</td>
<td>-0.16</td>
<td>-0.13</td>
<td>-0.05</td>
<td>0.2-30 µm</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\epsilon_{11} = 4.9$</td>
<td>$\epsilon_{14} = -0.16$</td>
<td></td>
<td>$n = 1.559$</td>
</tr>
<tr>
<td>GaAs</td>
<td>$\bar{4}3m$</td>
<td>118</td>
<td>5.317</td>
<td>4.5</td>
<td>4.5</td>
<td>4.5</td>
<td>59.4</td>
<td>53.5</td>
<td>-0.05</td>
<td>-0.16</td>
<td>1.43 pm/V</td>
<td>0.9-17.3 µm</td>
<td>@ 589 nm</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\epsilon_{14} = -13.0$</td>
<td>@ 1150 nm</td>
<td></td>
<td>$n = 3.442$</td>
</tr>
</tbody>
</table>

5.4.2 Uniaxial Crystals

This section presents the results of my $M_2$ graphical representation for the uniaxial crystal Lithium Niobate (LiNbO$_3$). A detailed discussion is provided of the $M_2$ surface dependence on AO frequency $f_a$ and optical wavelength $\lambda$. Appendix B provides further examples with tellurium dioxide (TeO$_2$, potassium didideuterium (KDP) and lead molybdate (PbMoO$_4$). Chapter 7 discusses the $M_2$ surfaces of $\alpha$-BBO.

LiNbO$_3$ is commonly fabricated for use in acousto-optics, electro-optics, and nonlinear optics. Similar to $\beta$-BBO, LiNbO$_3$ belongs to the $3m$ point group, making LiNbO$_3$ piezoelectric. LiNbO$_3$ has a small acoustic attenuation of $\sim 0.1$ (dB/µs · GHz$^2$), allowing for devices with large acoustic-frequencies. Figure 5.6 illustrates the acoustic-slowness surface for LiNbO$_3$. The slowness surface
Figure 5.6: LiNbO\textsubscript{3} acoustic-slowness surface. The red, blue, and green surfaces represent the quasi-longitudinal, fast quasi-shear, and slow quasi-shear acoustic modes, respectively.

displays little asymmetry, so variation of the $M_2$ surfaces for the different modes will depend more on the effective photoelastic coefficient $p(\phi, \theta)$ than the acoustic velocity $V_a(\phi, \theta)$. I used the values listed in Table 5.3 for all velocity and $M_2$ calculations in LiNbO\textsubscript{3}.

Figure 5.7 illustrates the $M_2$ surfaces for $\lambda = 632.8$ nm, $f_a = 500$ MHz, $n_o = 2.2865$, and $n_e = 2.2034$. Each of the 9 different surfaces represent a different type of AO diffraction. In addition, each surface must possess the $3m$ symmetry of LiNbO\textsubscript{3}, as most noticeably seen in the $\hat{\epsilon} \rightarrow \hat{o}$ diffraction for both the QFS and QSS modes. Table 5.4 lists the maximum $M_2$ values for each surface. The largest $M_2$ value of 15.98 (10$^{-15}$s$^3$/kg) comes from the QSS mode for $\hat{e} \rightarrow \hat{o}$ optical polarization diffraction. Notice how the maximum $M_2$ values occur at complicated, off-axes angles that require a graphical representation to find.

The results presented so far in this chapter have been for a fixed AO frequency $f_a$ and a fixed
Figure 5.7: $M_2$ surfaces for different types of acousto-optic diffraction in LiNbO$_3$ for $f_A = 500$MHz and $\lambda = 633$nm.
Table 5.3: Physical properties of LiNbO$_3$ [73, 82].

<table>
<thead>
<tr>
<th>Point Group</th>
<th>Elastic Stiffness Tensor (GPa)</th>
<th>Photoelastic Tensor</th>
<th>DE Const.</th>
<th>EO Const. ($\frac{C}{m^2}$)</th>
<th>Transparency Index of ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rho$ (g/cm$^3$)</td>
<td>$c_{11}$</td>
<td>$p_{11}$</td>
<td>$\epsilon_{11} = 44$</td>
<td>$r_{13} = 10.1$</td>
<td>0.33-5.5 $\mu$m</td>
</tr>
<tr>
<td>Mohs Hardness</td>
<td>$c_{12}$</td>
<td>$p_{12}$</td>
<td>$\epsilon_{33} = 27.9$</td>
<td>$r_{22} = 6.79$</td>
<td>$n_o = 2.2865$</td>
</tr>
<tr>
<td></td>
<td>$c_{13}$</td>
<td>$p_{13}$</td>
<td>$\epsilon_{15} = 3.67$</td>
<td>$r_{33} = 33.2$</td>
<td>$n_e = 2.2034$</td>
</tr>
<tr>
<td></td>
<td>$c_{14}$</td>
<td>$p_{14}$</td>
<td>$\epsilon_{22} = 2.38$</td>
<td>$r_{31} = 31.1$</td>
<td>(@ 632.8nm)</td>
</tr>
<tr>
<td></td>
<td>$c_{33}$</td>
<td>$p_{33}$</td>
<td>$\epsilon_{31} = 0.34$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$c_{44}$</td>
<td>$p_{44}$</td>
<td>$\epsilon_{33} = 1.6$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$3m$</td>
<td>199.2</td>
<td>-0.021</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4.628</td>
<td>54.7</td>
<td>0.172</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5-5.5</td>
<td>70.0</td>
<td>-0.052</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>7.9</td>
<td>0.141</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>240</td>
<td>0.118</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>59.9</td>
<td>-0.109</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

However, it is worth understanding how the $M_2$ surfaces depend on both $f_a$ and $\lambda$. Figure 5.8 illustrates the maximum $M_2$ values of the 9 different types of AO diffraction as a function of $f_a$ and $\lambda$ in LiNbO$_3$. As the figure shows, the $M_2$ values for $\hat{e} \rightarrow \hat{e}$ and $\hat{\sigma} \rightarrow \hat{\sigma}$ diffraction show little change when $f_a$ is changed from 10 MHz to 2.0 GHz. The $M_2$ values for the $\hat{e} \rightarrow \hat{\sigma}$ diffraction, on the other hand, see a change as large as 450%. With the optical wavelength set to 632.8 nm, the maximum $M_2$ value of $18.48 \times 10^{-15} s^3/kg$ occurs at $f_a = 0.94$ GHz.

The difference between the dependence of isotropic and anisotropic diffraction is due to the difference in how the Bragg-matched solutions evolve as a function of $f_a$. Figure 5.9 illustrates the Bragg-matched solutions for both isotropic and anisotropic diffraction at 0.15 GHz and 1.5 GHz. The solutions for the isotropic diffraction do not change significantly between the two frequencies. At both frequencies, the angular variation in the Bragg-matched solutions for the optical waves is very large, and the optical wavevectors of the solutions are nearly orthogonal the $\vec{K}_a$. The only difference between the two frequencies is that the solutions become farther apart, but this does significantly affect the effective photoelastic coefficient $p$ as the polarizations of the optical
Table 5.4: Maximum $M_2$ values and corresponding acousto-optic geometries for a given type of diffraction in LiNbO$_3$ with a $\lambda = 633$ nm and $f_a = 500$ MHz acoustic wave while including linear electro-optic effect.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$\vec{k}_i$</th>
<th>$\vec{k}_d$</th>
<th>$\vec{K}_a$</th>
<th>$M_{2\text{max}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\phi$</td>
<td>$\theta$</td>
<td>$\phi$</td>
<td>$\theta$</td>
</tr>
<tr>
<td>Isotropic diffraction $\hat{o} \rightarrow \hat{o}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>-150.00</td>
<td>8.59</td>
<td>-150.00</td>
<td>7.41</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>164.57</td>
<td>109.10</td>
<td>163.78</td>
<td>111.00</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>73.60</td>
<td>68.36</td>
<td>72.93</td>
<td>70.32</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>90.78</td>
<td>52.99</td>
<td>89.23</td>
<td>52.99</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>-58.55</td>
<td>32.94</td>
<td>-55.91</td>
<td>31.77</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>139.16</td>
<td>55.34</td>
<td>137.44</td>
<td>57.08</td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>-12.85</td>
<td>21.02</td>
<td>-10.44</td>
<td>21.74</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>90.76</td>
<td>52.98</td>
<td>90.76</td>
<td>51.22</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>90.00</td>
<td>105.07</td>
<td>90.00</td>
<td>106.18</td>
</tr>
</tbody>
</table>

Figure 5.8: Maximum $M_2$ value as a function of applied AO frequency with $\lambda = 632.8$ nm (left) and as a function of the incident optical wavelength with $f_a = 500$ MHz (right). The red, blue, and green lines refer to $\hat{o} \rightarrow \hat{o}$, $\hat{e} \rightarrow \hat{e}$, and $\hat{e} \rightarrow \hat{o}$ diffraction, respectively. The solid $\circ$, $\nabla$, and $\triangle$ refer to diffraction from the QL, QFS, and QSS acoustic modes respectively.

Wavevectors do not change dramatically. This analysis reveals that the $M_2$ surfaces and calculations will not significantly vary in optically isotropic crystals.

For the anisotropic diffraction, the Bragg-matched solutions change dramatically. At low AO frequencies, the solutions appear closely around the optic axis with little angular variation. When $f_a = 1.5$ GHz, the angular variation of the solutions increases significantly and the optical
Figure 5.9: Bragg matching solutions for \( \hat{e} \rightarrow \hat{e} \) (left column) and \( \hat{e} \rightarrow \hat{o} \) (right column) at 0.15 GHz and 1.5 GHz for \( \lambda = 633 \text{ nm} \), \( \phi = 30^\circ \), and \( \theta = 28^\circ \) for the QSS mode in LNiBO\(_3\).
Figure 5.10: $M_2$ surfaces for the QSS mode in LNBO$_3$ for different AO frequencies.
wavevectors of the solutions become more orthogonal to \( \hat{K}_a \). This dramatic change causes the effective photoselastic coefficient \( p \) (and therefore \( M_2 \)) to have a significant dependence on \( f_a \). Figure 5.9 illustrates the Bragg-matched solutions for the QSS, so the results will be less dramatic for the faster acoustic modes. In LiNbO\(_3\), the change of the Bragg-matched solutions causes the \( M_2 \) values to increase with an increasing \( f_a \). In crystals with a different photoelastic tensor, however, the \( M_2 \) values could decrease with increasing \( f_a \). The evolution of the \( M_2 \) surfaces is illustrated in Fig. 5.10, which shows how the shape and the location of the maximum \( M_2 \) value of the \( M_2 \) surfaces depends on \( f_a \).

The dependence of the \( M_2 \) surfaces on \( \lambda \) is similar to the dependence on \( f_a \). As Fig. 5.8 shows, the maximum \( M_2 \) values decrease as a function of increasing \( \lambda \) because the indices of refraction decrease with increasing \( \lambda \). Note that the overall diffraction efficiency scales as \( 1/\lambda^2 \), but this \( 1/\lambda^2 \) term is not included in the \( M_2 \) calculation. Thus, the diffraction efficiencies for smaller optical wavelengths are relatively larger than the \( M_2 \) values suggest. With \( M_2 \sim n_i^3n_d^3 \), the decreasing indices of refraction simply apply a scaling factor to the \( M_2 \) surfaces and maximum \( M_2 \) values. For anisotropic diffraction, the decreasing indices of refraction and increasing optical wavelength decrease the magnitude of the optical momentum surfaces (\(|k| = \frac{2\pi n}{\lambda}\)), which gives the illusion of the acoustic wavevector increasing in size. This provides the same effect as increasing \( f_a \), which as we have already seen causes the maximum \( M_2 \) value to increase for anisotropic diffraction. Therefore, the maximum \( M_2 \) values for the anisotropic diffraction in Fig. 5.8 are sometimes seen to increase with increasing \( \lambda \).

5.5 Conclusion

This chapter presented a graphical representation for the maximum \( M_2 \) value in a given material, AO frequency \( f_a \), and optical wavelength \( \lambda \). The graphical representation only considers Bragg-matched geometries, for which I provide simple solutions. I also discussed how the solutions evolve as a function of \( f_a \) and \( \lambda \). With this algorithm, analysis, and all of the appropriate physical parameters precisely measured, the AO geometry with the largest \( M_2 \) value can be found.
Chapter 6

Optimizing Acousto-Optic Geometry

The algorithm presented in Chapter 5 searched for the largest acousto-optic (AO) figure of merit for a given material at a fixed AO frequency $f_a$ and optical wavelength $\lambda$. However, a large figure of merit does not completely define the quality of a device. Other parameters such as the AO bandwidth, resolution, angular aperture, acoustic walk-off, acoustic absorption, and fabrication feasibility are all important qualities. Thus, this chapter presents three different graphical representations for AO geometries with specific, inherit strengths.

The first algorithm presented represents AO geometries in which the acoustic wavevector $\vec{K}_a$ is tangent to the optical momentum surface of the diffracted light. Such AO geometries are known as tangentially-degenerate and intrinsically have large AO bandwidths [71]. The second algorithm searches for sub-collinear AO tunable filter geometries in which the acoustic Poynting vector $\vec{S}_a$ is parallel with the incident optical Poynting vector $\vec{S}_i$. Sub-collinear AO geometries allow for long interaction lengths of the acoustic wave with the incident optical wave, resulting in AO devices with high spectral resolution appropriate for spectral filters and pulse shapers [83, 84]. The third and final method searches for AO geometries with parallel incident and diffracted optical Poynting vectors. These AO geometries, known as parallel-tangents, are appropriate for tunable optical filters and have a large angular aperture.

For all three presented search algorithms, the AO frequency $f_a$ is a free variable, but $\lambda$ is held fixed. I appraise each AO geometry with the $M_2$ AO figure of merit, as was done in Chapter 5, but any figure of merit could be used. I apply each algorithm to LiNbO$_3$ in this chapter, and
KDP and PbMoO$_4$ in Appendix B. The AO geometries of the three different algorithms require birefringent crystals, so I do not apply our search algorithms to optically isotropic crystals.

6.1 Tangentially-Degenerate Acousto-Optic Geometries

Acousto-optic devices with large bandwidths are required when tuning a laser over a large frequency range. For the quantum computer experiments with $^{9}$Be$^+$ ions, the laser light needs to be tuned over a range of about 1.24 GHz to properly address the different atomic transitions used for cooling, imaging, and manipulating the ions. The transducer of an AO device can be decreased in size to increase the bandwidth of the device, but doing so decreases the diffraction efficiency. A popular alternative is to use a phased-array transducer that changes the direction of the acoustic wavevector as function of the applied AO frequency $f_a$. This beam steering technique, when properly designed, can increase the AO bandwidth without significantly decreasing the diffraction efficiency. Instead of engineering the transducer to achieve large AO bandwidths, a properly chosen AO geometry can be used. For instance, tangentially-degenerate AO geometries achieve large AO bandwidths without special transducer customizations. Below I discuss tangentially-degenerate geometries and how I search a given crystal for an efficient, tangentially-degenerate geometry.

In a tangentially-degenerate geometry, the outer-polarization mode, which is the ordinary-polarization mode for a negative uniaxial crystals like LiNbO$_3$ and BBO, is diffracted to the inner-polarization mode at a point of tangency. This point of tangency occurs where the Poynting vector of the diffracted light $\vec{S}_d$, which is normal the optical momentum surface of the diffracted light, is orthogonal to the acoustic wavevector $\vec{K}_a$:

$$\vec{K}_a \cdot \vec{S}_d = 0.$$  

Figure 6.1a illustrates the tangentially-degenerate diffraction geometry in momentum space. With $\vec{K}_a$ parallel with the tangent of the diffracted momentum surface, the Bragg-mismatch increases slowly with an increasing or decreasing magnitude of $\vec{K}_a$. The bandwidth of a tangentially-degenerate device can be further improved by rotating the AO device to slightly displace the
6.1.1 Tangentially-Degenerate Solutions for Negative-Uniaxial Crystals

I search for tangentially-degenerate solutions by first defining the propagation direction of the diffracted wave \( \vec{k}_d = (k_x, k_y, k_z) \). For tangentially-degenerate geometries in negative-uniaxial crystals, \( \vec{k}_d \) must lie on the extraordinary momentum surface. The tangent plane to the extraordinary momentum surface for a given \( \vec{k}_d \) is defined by

\[
\frac{k_x}{n_e^2} x + \frac{k_y}{n_e^2} y + \frac{k_z}{n_o^2} z = 1.
\]

The orientation of this tangent plane can be described by a single vector:

\[
\vec{p} = \left( \frac{k_x}{n_e^2}, \frac{k_y}{n_e^2}, \frac{k_z}{n_o^2} \right).
\]
The displacement of the plane from the origin along $\hat{p}$ ($\hat{p} = \hat{p}/|\hat{p}|$) is given by

$$d = \frac{1}{\sqrt{(k_x/n_o^2)^2 + (k_y/n_o^2)^2 + (k_z/n_o^2)^2}}.$$  

With the plane defined by $\hat{p}$ and $d$, the intersection of the plane with the ordinary momentum surface, which is a sphere, needs to be solved. The intersection of a sphere with a plane is a circle of radius $r = \sqrt{k_0^2n_o^2 - d^2}$, where $k_0 = 2\pi/\lambda$. The solutions of the intersection $\vec{c}(s)$ are thus given by

$$\vec{c}(s) = r(\hat{r}_1 \cos s + \hat{r}_2 \sin s) + d \cdot \hat{p}.$$  

(6.1)

Here $\hat{r}_1$ and $\hat{r}_2$ are any two orthogonal vectors that span the plane defined by $\hat{p}$. With Eq. 6.1, the solutions of the intersection of the tangent plane to $\vec{k}_d$ with the ordinary surface can easily be solved. Figure 6.2 illustrates the solutions of tangentially-degenerate geometries in a negative-uniaxial crystal for a given $\vec{k}_d$. 

Figure 6.2: (a) Illustration of a plane tangent to the extraordinary momentum surface at point defined by the diffracted optical vector $\vec{k}_d$, whose direction is represented by the red arrow. The black arrow presents $\vec{p}$. (b) Solutions of the intersection of the plane with the ordinary momentum surface. The birefringence $(n_o - n_e)$ is greatly exaggerated to better illustrate the solution.
6.1.2 Tangentially-Degenerate Solutions for Negative-Uniaxial Crystals

For positive-uniaxial crystals, the tangentially-degenerate solutions are found by solving for the intersection of a plane with an ellipsoid. With a defined diffracted wavevector \( k_d = k_o n_o (\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta) \), the tangent plane is then defined by

\[
\hat{p} = \frac{k_d}{(k_o n_o)},
\]

and is displaced from the origin by

\[
\vec{d} = k_o n_o \hat{p} = \vec{k}_d.
\]

The resulting intersection is an ellipse:

\[
\vec{c}(s) = (a \cos \theta - a_0) \hat{r}_1 + b \sin \theta \hat{r}_2 + k_d,
\]

where,

\[
a = k_o \frac{2 n_e n_o \sqrt{(n_e^2 - n_o^2) \sin^2 \theta}}{n_e^2 + n_o^2 + (n_e^2 - n_o^2) \cos^2 \theta},
\]

\[
b = k_o n_e \sqrt{\frac{(n_e^2 - n_o^2) \sin^2 \theta}{n_e^2 \cos^2 \theta + n_o^2 \sin^2 \theta}},
\]

\[
a_0 = k_o \frac{n_o (n_e^2 - n_o^2) \cos \theta \sin \theta}{n_e^2 \cos^2 \theta + n_o^2 \sin^2 \theta},
\]

\[
\hat{r}_1 = (-\cos \phi \cos \theta, -\sin \phi \cos \theta, \sin \theta),
\]

\[
\hat{r}_2 = (\sin \phi, -\cos \phi, 0).
\]

The equations above provide solutions for all Bragg-matched, tangentially-degenerate geometries in positive-uniaxial crystals for a given \( \vec{k}_d \).

6.1.3 Tangentially-Degenerate Search Algorithm

I developed an algorithm to calculate the \( M_2 \) values for tangentially-degenerate AO geometries in uniaxial crystals. The algorithm works by sweeping the diffracted optical vector \( \vec{k}_d \) in spherical coordinates as a function of its azimuth angle \( \phi \) and polar angle \( \theta \). For each diffracted optical wavevector \( \vec{k}_d \), I find the plane tangent to the inner optical momentum surface (the extraordinary
momentum surface for negative-uniaxial crystals) as illustrated in Fig. 6.2a. Next I solve for the intersection of the tangent plane with the outer momentum surface as illustrated in Fig. 6.2b. All of the intersection points represent incident optical wavevectors \( \bar{k}_i \) that form a tangentially-degenerate geometry with the defined diffracted optical wavevector \( \bar{k}_d \). The acoustic wavevector \( \bar{K}_a \) is then easily calculated as \( \bar{K}_a = \bar{k}_d - \bar{k}_i \). This acoustic wavevector \( \bar{K}_a \) can be any of the three modes with their corresponding acoustic velocity. The frequency of the acoustic wavevector is calculated by \( |\bar{K}_a| = \frac{2\pi}{V_a(\bar{K}_a, \alpha)} f^\alpha_a \), where \( f^\alpha_a \) is frequency of the acoustic wave and \( V_a(\bar{K}_a, \alpha) \) is the velocity of the acoustic wave, which is a function of the orientation \( \bar{K}_a \) and the acoustic mode indexed by \( \alpha = (QL,QFS,QSS) \). For each direction of the diffracted optical vector \( \bar{k}_d \), I find all of the tangentially-degenerate geometries and calculate the \( M_2 \) values. I select the maximum \( M_2 \) value calculated and the corresponding acoustic frequency for each direction. Notice how the acoustic frequency is not fixed for this search method and is allowed to vary. Below is a summary of the algorithm:

- **For-loop**: Sweep in spherical coordinates through all possible \( \bar{k}_d(\phi, \theta) \)
  
  * Determine the tangent plane to \( \bar{k}_d(\phi, \theta) \)
  
  * Solve for intersection of tangent plane with the outer optical momentum surface \( \bar{c}(s) \)
    
    \( \bar{k}_i(s) \equiv \bar{c}(s) \)

  * **For-loop**: Sweep parametrically through solutions indexed by \( s \)
    
    \( \bar{K}_a = \bar{k}_d - \bar{k}_i(s) \)

    * Determine the acoustic frequency for each mode: \( \bar{K}_a | = \frac{2\pi}{V_a(\bar{K}_a, \alpha)} f^\alpha_a \)

    * Calculate \( M_2^\alpha(\phi, \theta, s) \)

  * **End For-loop**

  * Store Max \( (M_2^\alpha(\phi, \theta, s)) \) and corresponding \( f^\alpha_a \) for each diffraction type

- **End For-loop** Plot \( M_2^\alpha(\phi, \theta) \) and print max \( M_2 \) values for each diffraction type
Table 6.1: Maximum $M_2$ values and corresponding tangentially-degenerate AO geometries in LiNbO$_3$ with $\lambda = 632.8$ nm.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$f_a$ (GHz)</th>
<th>$\vec{k}_{ord}$</th>
<th>$\vec{k}_{ext}$</th>
<th>$\vec{K}_a$</th>
<th>$M_2^{\text{max}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\phi$</td>
<td>$\theta$</td>
<td>$\phi$</td>
<td>$\theta$</td>
<td>$\phi$</td>
</tr>
<tr>
<td>No Restriction on $f_a$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>6.61</td>
<td>81.49</td>
<td>74.95</td>
<td>96</td>
<td>69</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>4.45</td>
<td>176.48</td>
<td>100.94</td>
<td>161</td>
<td>102</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>2.78</td>
<td>270.00</td>
<td>126.76</td>
<td>270</td>
<td>114</td>
</tr>
</tbody>
</table>

6.1.4 Results

I applied the tangentially-degenerate algorithm to LiNbO$_3$ with $\lambda = 632.8$ nm. Table 6.1 presents the results from our search and lists the largest $M_2$ of $16.98 \times 10^{-15}$ s$^3$/kg occurring for the QSS mode at $f_a = 2.78$ GHz. This $M_2$ value is close to the absolute maximum $M_2$ value in LiNbO$_3$ of $18.48 \times 10^{-15}$ s$^3$/kg that was discovered in Chapter 5. The QSS mode should have the largest $M_2$ values because shear modes diffract polarization-switching diffraction more efficiently than longitudinal modes, and QSS is the slowest mode ($M_2 \sim 1/V_a^3$). The QSS modes also require smaller AO frequencies, as Table 6.1 shows, because $K_a = \frac{2\pi f_a}{V_a}$, making the fabrication of the transducer more feasible. If I were to increase the wavelength $\lambda$ for the algorithm, then the AO frequencies would decrease because both the birefringence and the magnitude of the optical wavevectors decrease with increasing $\lambda$.

Figure 6.3 illustrates the resulting $M_2$ surfaces. Recall that the $M_2$ surfaces for this search represent the maximum $M_2$ values of a tangentially-degenerate geometry as a function of the propagation direction of $\vec{k}_d$ and not $\vec{K}_a$. Therefore, values near the optic axis where the birefringence is small require in general smaller acoustic frequencies. Values in the XY plane where the birefringence is largest in general require larger acoustic frequencies. However, the anisotropy of the acoustic slowness surface could compensate for the birefringence in some crystals.
6.2 Harris Filter

Before discussing both the sub-collinear and parallel-tangents geometry, it is worth discussing the Harris filter [85]. The Harris filter is an AO device in which the acoustic and optical waves propagate in parallel in the XY-plane of a crystal. Figure 6.4 illustrates the Harris Filter both in momentum space and real space. In the Harris filter, a launched acoustic wave reflects off a prism to propagate in parallel with the incident and optical waves. The prism is designed to completely reflect the acoustic waves but is indexed matched to transmit the optical wave. The acoustic wave diffracts the incident optical wave into the orthogonal mode, and is then reflected by a second prism to an acoustic absorber to avoid acoustic back reflections. Upon exiting the crystal, the incident and diffracted optical waves are separated by a polarizing beam splitter.

The strengths of the Harris filter comes from two different physical properties: 1) the Poynting vectors of the acoustic and incident optical waves are parallel, and 2) the Poynting vectors of incident and diffracted optical waves are parallel. Strength 1 allows for long interaction lengths of the acoustic and optical waves, resulting in high spectral resolution. This high resolution restricts the bandwidth of the spectral frequencies that efficiently diffract the light into the orthogonal mode for a given AO frequency. As a result, the Harris filter can electronically filter the spectrum of the incident light by adjusting the AO frequency. Such devices are commonly known as acousto-optic tunable filters (AOTF or TAOF).
Strength 2 allows for a large angular aperture. The parallel Poynting vectors imply that the tangents of the optical wavevectors with respect to the corresponding momentum surfaces are parallel. With parallel tangents, the Bragg-matching condition will be satisfied over a large range of incident angles (see Fig. 6.7). With a large angular aperture, the incident light does not need to be well collimated.

The Harris filter is practical and useful design. However, the Harris filter diffracts xy-polarized light to z-polarized light, requiring large $p_{4n}$ or $p_{5n}$ coefficients. In some crystals, such as TeO$_2$, these coefficients are small making the corresponding diffraction efficiency small as well. In other crystals, such as BBO, the birefringence is so large that the Harris filter would only work at large, impractical AO frequencies. Therefore, I parse the strengths of the Harris filter into two different graphical representations. The first algorithm searches for sub-collinear AO geometries in which the acoustic and incident optical Poynting vectors are parallel to achieve high resolution. The second algorithm searches for parallel-tangents or noncollinear geometries in which the incident and diffracted optical waves have parallel Poynting vectors to achieve large angular bandwidths [83].
6.3 Sub-Collinear Acousto-Optic Tunable Filters

Increasing the interaction length of the acoustic and incident optical waves increases the diffraction efficiency and resolution of a device. For a deflector geometry, a larger interaction length can be achieved by increasing the length of the transducer, but this decreases the acoustic power density. Alternatively, the AO geometry can be chosen to maximize the interaction length. For instance, if the Poynting vectors of the acoustic and optical waves are made parallel ($\vec{S}_a||\vec{S}_i$), known as the sub-collinear geometry, then the interaction length will be as long as allowed by the finite size and acoustic absorption of the crystal. With the Poynting vectors of the acoustic and incident optical waves being parallel, some sort of acoustic or optical reflection in the crystal is required to achieve perfect overlap of the two waves. Figure 6.5 illustrates a sub-collinear device in both momentum and real space in which acoustic reflection in the device is used to achieve overlap of the acoustic and optical beams. The large interaction length achieved by sub-collinear geometries result in devices with high spectral resolution. This section presents the search algorithm for discovering and appraising sub-collinear AO geometries.

The Poynting vector $\vec{S}$ describes the direction of energy flow, and the direction is given by the normal to the momentum surface (see Chapter 2). For a uniaxial crystal, the optical momentum
surfaces are described by
\[ \frac{k_x^2 + k_y^2 + k_z^2}{n_o^2} = 1, \quad \text{(ordinary momentum surface)} \quad (6.3) \]
\[ \frac{k_x^2 + k_y^2 + k_z^2}{n_e^2} + \frac{k_x^2}{n_o^2} = 1. \quad \text{(extraordinary momentum surface)} \quad (6.4) \]

Here \( k_o = \frac{2\pi}{\lambda} \) has been divided out from both equations. The ordinary momentum surface is a sphere, so the normal to the surface is simply the radial line. For example, the normalized Poynting vector for a point on the ordinary momentum surface \( \{p_x, p_y, p_z\} \) is simply \( \hat{S}_o = \frac{\{k_x, k_y, k_z\}}{n_o} \). The extraordinary momentum surface is an ellipsoid, so in order to find the normalized Poynting vector \( \hat{S}_e \) I take the gradient of Eq. 6.4:
\[ \hat{S}_e = \left( \frac{k_x^2}{n_e^2} + \frac{k_y^2}{n_e^2} + \frac{k_z^2}{n_o^2} \right)^{-1/2} \left[ \frac{k_x}{n_e^2}, \frac{k_y}{n_e^2}, \frac{k_z}{n_o^2} \right]. \]

I solve for the acoustic Poynting vector \( \hat{S}_a \) by first determining the energy velocity vector \( \bar{V}_e \), which is the velocity of the Poynting vector:
\[ \bar{V}_e = \frac{\bar{c} \bar{U} \hat{K}_a}{\rho V_a}. \quad (6.5) \]

Here \( \bar{c} \) is the stiffness tensor, \( \bar{U} \) is the particle displacement of the acoustic wave, and \( \hat{K}_a \) is the direction of the acoustic wavevector. I normalize \( \bar{V}_e \) to solve for \( \hat{S}_a \) [24].

The TAOF search method begins by sweeping through the acoustic momentum surface in spherical coordinates. For each acoustic wave direction, I calculate the direction of the acoustic Poynting vector \( \hat{S}_a \), and then find an incident optical Poynting vector \( \hat{S}_i \), for either the ordinary or extraordinary optical polarization, that is parallel with \( \hat{S}_a \). I then calculate the diffracted vectors and acoustic frequencies required for a Bragg matched AO geometry for both the \( \hat{e} \to \hat{o} \) or \( \hat{o} \to \hat{e} \) diffraction. Finally I calculate the \( M_2 \) value. For each acoustic wavevector direction and polarization, I record the maximum \( M_2 \) value for both cases of diffraction.

After matching the Poynting vectors of the acoustic and incident optical vectors, I must determine the magnitude of \( M_2 \) that makes the AO geometry Bragg matched. For Bragg-matched geometries, the \( \hat{K}_a, \hat{k}_i, \) and \( \hat{k}_d \) wavevectors create a closed triangle in momentum space. For \( \hat{e} \to \hat{o} \) diffraction, \( \hat{k}_i \) lies on the extraordinary momentum surface and \( \hat{k}_i \) and \( \hat{k}_d \) have magnitudes of \( k_i = n_e(\theta_i)k_o \) and \( k_d = n_0k_o \). The law of cosines states that
\[ a^2 = b^2 + c^2 - 2bc \cos \alpha. \]
Here $a$, $b$, and $c$ are the lengths of the legs of a triangle and $A$ is the angle opposite of leg $a$. Applying the law of cosines to the Bragg-matched triangle, I get

$$n_o^2\omega^2 = K_a^2 + n_e^2(\theta_i)\frac{\omega^2}{c^2} - 2K_a(K_a \cdot \vec{k}_i). \quad (6.6)$$

Here I have applied the dot product rule $\vec{X} \cdot \vec{Y} = |a||b|\cos C$, where $C$ is the angle between $\vec{a}$ and $\vec{b}$. Applying the quadratic formula to Eq. 6.6 to solve for $K_a$, I get

$$K_a = \hat{K}_a \cdot \vec{k}_i \pm \sqrt{(\hat{K}_a \cdot \vec{k}_i)^2 + \omega^2(\theta_i)}.$$ \quad (6.7)

For the search algorithm, I only consider $K_a$ with the smallest absolute magnitude, because this results in the smaller corresponding $f_a$. With $K_a$, I solve for $\vec{k}_d$ by $\vec{k}_d = \vec{k}_i - \vec{K}_a$.

To solve for $\hat{\theta} \rightarrow \hat{e}$ diffraction, the magnitude of $\vec{k}_d$ is unknown. I circumnavigate this problem by applying to momentum space a scaling factor of

$$\vec{X} = \begin{pmatrix} \frac{1}{n_e} & 0 & 0 \\ 0 & \frac{1}{n_e} & 0 \\ 0 & 0 & \frac{1}{n_o} \end{pmatrix}.$$  

Next I solve for $K_a$ as I did for $\hat{e} \rightarrow \hat{\theta}$ diffraction, resulting in the following solution:

$$K_a = (\hat{K}_a \vec{X}) \cdot (\vec{k}_i \vec{X}) \pm \sqrt{(\hat{K}_a \vec{X}) \cdot (\vec{k}_i \vec{X}) - (\hat{K}_a \vec{X})^2 \left((\vec{k}_i \vec{X})^2 - 1\right)}.$$  \quad (6.8)

Once again I only consider the $K_a$ with the smallest absolute magnitude for our search algorithm. With Eqs. 6.6 and 6.8, I can solve for the magnitude of $K_a$ and for $\vec{k}_d$ for a given $\hat{K}_a$ and $\vec{k}_i$.

### 6.3.1 Results

Table 6.2 lists the results of the sub-collinear search. The QSS acoustic mode produces the largest $M_2$ value with the acoustics propagating in the YZ plane. The YZ-plane is a mirror plane in LiNbO$_3$, so the acoustic walk-off is constrained to the YZ-plane. In addition, the acoustic mode is purely z-polarized, making the fabrication of the device simpler. The largest $M_2$ value for the QL mode happens to be a Harris filter geometry. However, the required AO frequency is 862 MHz and the $M_2$ value is only $1.16 \times 10^{-15} s^3/kg$. 

Figure 6.6 illustrates the $M_2$ surfaces of the sub-collinear search for the QSS mode. The $\vec{S}_a || \vec{S}_{ext}$ and $\vec{S}_a || \vec{S}_{ord}$ searches produce similar results because of the small difference between the walk-off of the extraordinary and ordinary optical modes.

### Table 6.2: Maximum $M_2$ values and corresponding sub-collinear AO geometries in LiNbO$_3$.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$f_a$ (MHz)</th>
<th>$\vec{k}_{ord}$</th>
<th>$\vec{k}_{ext}$</th>
<th>$\vec{K}_a$</th>
<th>$M_2^{max}$ (10$^{-15}$s$^3$/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quasi-Long.</td>
<td>862</td>
<td>0 90</td>
<td>0 90</td>
<td>0 90</td>
<td>1.16</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>91.6</td>
<td>-30 24.72</td>
<td>-30 24.78</td>
<td>-30 16</td>
<td>5.15</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>412</td>
<td>270.00 66.32</td>
<td>270.00 66.58</td>
<td>270 58</td>
<td>12.98</td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>862</td>
<td>0 90</td>
<td>0 90</td>
<td>0 90</td>
<td>1.16</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>80.3</td>
<td>-30 23.10</td>
<td>-30 23.15</td>
<td>-30 16</td>
<td>4.97</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>407</td>
<td>270 65.86</td>
<td>270 65.86</td>
<td>270 59</td>
<td>12.71</td>
</tr>
</tbody>
</table>

When the incident and diffracted optical Poynting vectors are parallel, then the device will have a large angular bandwidth. Parallel Poynting vectors indicate that the tangents of the optical momentum surfaces at the points specified by $\vec{k}_i$ and $\vec{k}_d$ are parallel; hence, the name parallel-tangents. With parallel-tangents, the separation of the optical momentum surfaces to first order
Table 6.3: Maximum $M_2$ values and corresponding parallel-tangents AO geometries in LiNbO$_3$.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$f_a$ (MHz)</th>
<th>$\vec{k}_{ord}$</th>
<th>$\vec{k}_{ext}$</th>
<th>$\vec{K}_a$</th>
<th>$M_2^{max}$ $(10^{-15} s^3/kg)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quasi-Long.</td>
<td>862</td>
<td>0 90</td>
<td>0 90</td>
<td>0 90</td>
<td>1.16</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>638</td>
<td>134 47.12</td>
<td>134 45</td>
<td>134 109.05</td>
<td>1.53</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>544</td>
<td>150.00 108.68</td>
<td>150 110</td>
<td>150 74.17</td>
<td>7.65</td>
</tr>
</tbody>
</table>

does not change as $\vec{k}_i$ and $\vec{k}_d$ vary. Figure 6.7 illustrates how the momentum mismatch increases slowly as the incident angle varies, creating a large angular bandwidth.

I explore parallel-tangent AO geometries by sweeping in polar coordinates on the extraordinary momentum surface, and calculating $\hat{S}_{ext} = (k_x/n_e^2, k_y/n_e^2, k_z/n_o^2)$. The corresponding ordinary and acoustic momentum vectors are $\vec{k}_{ord} = n_o \hat{S}_{ext}$ and $\vec{K}_a = \pm (\vec{k}_{ext} - \vec{k}_{ord})$ ($\pm$ depending on positive or negative uniaxial crystals). I calculate the $M_2$ value and $f_a$ for each mode and plot the results.

Similar to the sub-collinear results, the QL produces the largest $M_2$ value for the Harris filter AO geometry. The QSS mode once again produces the largest $M_2$ value.
6.5 Conclusion

This chapter presents three different graphical representations for discovering AO geometries with large AO bandwidths, high resolution, or large angular bandwidths. I applied the algorithms to LiNbO$_3$ at 632.8 nm and illustrated the results with $M_2$ surfaces. For all three search algorithms, the AO frequency $f_a$ was allowed to vary to satisfy Bragg matching.

These three searches provide a practical means for searching for useful AO geometries in arbitrary directions that may have remained previously unexplored in a given medium. However, the results do not quantify the engineering feasibility of the AO geometries. For instance, the acoustic mode might have impractically large walk-off (tilt of the $K_a$ surface) or acoustic diffraction (curvature of the $K_a$ surface), or exciting the proper acoustic mode may be difficult. The sub-collinear geometries have the additional complication of overlapping the acoustic and incident optical waves. Despite not providing a complete AO device design, these searches do provide useful insights to the many unexplored possibilities for optimizing the AO interaction for a given crystal at a certain optical wavelength.
I have designed an $\alpha$ barium borate (BBO) acousto-optic (AO) device based on the research presented in the previous chapters. The AO geometry for the device was chosen using the optimization algorithm presented in Chapter 5. The calculations in the algorithm required the elastic-stiffness and photoelastic coefficients that were measured with the experiments described in Chapters 3 and 4 respectively. Lastly, I used the theory presented in Chapter 2 to ensure that the design was both practical and effective.

This chapter reviews the design requirements for David Winleand’s group at the National Institute of Standards and Technology (NIST). Next the $M_2$ graphical representations are presented that aide in locating an efficient AO geometry in $\alpha$-BBO. From the $M_2$ results, I identify a practical AO design that meets the requirements for NIST. The chapter concludes by discussing the theoretical performances of the device, and compares them to the measured experimental results. Lastly, the $\alpha$-BBO design is compared to current AO devices.

### 7.1 Device Requirements

In Chapter 1 I discussed the requirements for a UV transparent AO device that Wineland’s group needs for a scalable quantum computer. However, most of these design requirements are not urgent but are required for future experiments. Currently, the group needs an efficient, UV transparent acousto-optic frequency shifter (AOFS) that operates at 313 nm and 280 nm for their $^9\text{Be}^+$ and $^{24}\text{Mg}^+$ experiments, respectively. For the 313 nm light, the light needs about 1.24 GHz
of total frequency shift. Brimrose, my collaborators, face manufacturing difficulties fabricating a
device with such a large AO frequency. The highest, achievable AO frequency that they are able
to fabricate is \( \sim 400 \text{ MHz} \). Therefore, I achieve the 1.24 GHz frequency shift by passing the light
back and forth through the AOFS four times with a frequency shift of 310 MHz for each pass.
For the 280nm light, the light needs a total frequency shift of 1.686 GHz, or 422 MHz for each
of the four passes. Since the required frequency shift can depend on the applied magnetic field, a
bandwidth of \( \pm 80 \text{ MHz} \) is desired, requiring \( \pm 20 \text{ MHz} \) bandwidth for each pass because the total
frequency shift is the sum of the frequency shifts from each pass (\( f_t = 4(f_o + \Delta f) \)). To achieve a
3dB ripple across the total bandwidth, I choose a 0.75dB ripple (or 84\%) for each AO diffraction
since the total efficiency is the product of the efficiencies for each pass. Ideally each diffraction
should approach 80-90\% at low enough RF drive power to avoid deleterious heating effects.

### 7.2 \( M_2 \) Optimizations

I have applied my acousto-optic figure of merit \( M_2 \) graphical representation from Chapter 5
using the measured elastic-stiffness and photoelastic coefficients from Chapters 3 and 4 to deter-
mine an efficient AOFS device design using \( \alpha\)-BBO. Figure 7.1 illustrates the resulting \( M_2 \) surfaces
and Table 7.2 lists the AO geometry of the maximum \( M_2 \) values. The largest calculated \( M_2 \) value
of \( 94.90 \times 10^{-15}\text{s}^3/\text{kg} \) occurs for the slow quasi-shear (QSS) acoustic mode for the optical polarization
switching diffraction \( \hat{e} \rightarrow \hat{o} \). Unfortunately the polarization switching diffraction calculations de-
pend greatly on the \( p_{44} \) coefficient that I measured with an error bar of 100\%, so these calculations
will have large errors as well. The \( p_{44} \) coefficient is associated with polarization switching diffraction for \( \hat{x} \rightarrow \hat{z} \) or \( \hat{y} \rightarrow \hat{z} \) polarized light. For instance, the effective photoelastic coefficient for the
maximum \( M_2 \) geometry equals \( p = -0.08p_{11} + 0.08p_{12} - 0.79p_{14} - 0.11p_{41} - 0.59p_{44} \). With \( p_{14} \) and
\( p_{41} \) both being small compared to \( p_{44} \), the magnitude of \( p \) relies predominately on \( p_{44} \). Therefore,
I am unable to trust this or most other polarization switching diffraction calculations.

In addition to the \( M_2 \) value being unreliable, the AO geometry of the maximum \( M_2 \) calculation
presents fabrication difficulties because the acoustic and optical beams are separated by 30\°.
Table 7.1: Photoelastic coefficients used for $M_2$ search in $\alpha$-BBO.

<table>
<thead>
<tr>
<th>$p_{11}$</th>
<th>$p_{33}$</th>
<th>$p_{44}$</th>
<th>$p_{12}$</th>
<th>$p_{13}$</th>
<th>$p_{14}$</th>
<th>$p_{31}$</th>
<th>$p_{41}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.12</td>
<td>0.24</td>
<td>0.2</td>
<td>0.21</td>
<td>0.22</td>
<td>-0.007</td>
<td>0.21</td>
<td>-0.02</td>
</tr>
</tbody>
</table>

Figure 7.1: $M_2$ surfaces for $\hat{o} \rightarrow \hat{o}$ diffraction in $\alpha$-BBO for (left) the quasi-longitudinal mode, (middle) quasi-fast shear mode, and (left) quasi-slow shear mode. Freq=300 MHz, $\lambda = 300$ nm.

Ideally the acoustic and optical beams are nearly orthogonal or parallel so that the optical beams pass through a crystal face that either is parallel or orthogonal to the acoustic wave, respectively. With the acoustic and optical waves separated by an angle of $30^\circ$, the crystal would require a complicated, skewed cut and a large external angle for the incident beam.

I searched through the $M_2$ calculations for a AO geometry that would have a large $M_2$ value with a small error bar, a large AO bandwidth, and an AO geometry that would be easy to fabricate. I chose to use the $\hat{z}$-propagating quasi-longitudinal mode for our AOFS, which has an $M_2$ value of
Table 7.2: Maximum $M_2$ values and corresponding acousto-optic geometries for a given type of diffraction in $\alpha$-BBO with a $\lambda = 313$ nm and $f_a = 300$ MHz acoustic wave.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$\vec{k}_i$ $\phi$</th>
<th>$\vec{k}_d$ $\phi$</th>
<th>$\vec{K}_a$ $\phi$</th>
<th>$M_2^{\text{max}}$ $(10^{-15} s^3/kg)$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\vec{\hat{o}}$ $\rightarrow$ $\vec{\hat{o}}$</td>
<td>$\vec{\hat{o}}$ $\rightarrow$ $\vec{\hat{o}}$</td>
<td>$\vec{\hat{K}}_a$ $\phi$</td>
<td></td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>-90.00</td>
<td>-90.00</td>
<td>90</td>
<td>10.28</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>-0.57</td>
<td>90.52</td>
<td>90</td>
<td>11.67</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>-52.10</td>
<td>49.84</td>
<td>9</td>
<td>12.93</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>-150.00</td>
<td>-150.00</td>
<td>30</td>
<td>18.88</td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>90.00</td>
<td>90.00</td>
<td>90</td>
<td>22.41</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>90.00</td>
<td>90.00</td>
<td>90</td>
<td>45.97</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>79.73</td>
<td>22.94</td>
<td>14</td>
<td>1.44</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>91.52</td>
<td>38.16</td>
<td>0</td>
<td>63.80</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>90.00</td>
<td>42.29</td>
<td>90</td>
<td>94.90</td>
</tr>
</tbody>
</table>

9.96 and $7.16 \times 10^{-15} s^3/kg$ for $\vec{\hat{o}}$ $\rightarrow$ $\vec{\hat{o}}$ and $\vec{\hat{e}}$ $\rightarrow$ $\vec{\hat{e}}$ diffraction, respectively. In the following section I explain the strengths and expected performance of this AO geometry.

### 7.3 Device Design

From the $M_2$ search in $\alpha$-BBO, I have identified the $Z$-propagating quasi-longitudinal mode as an easy to fabricate AO geometry with good acoustic properties, $M_2$ value, and AO bandwidth. Figure 7.2 illustrates the advantageous acoustic properties of the AOFS design by displaying the XZ and YZ cross-sections of the acoustic slowness surfaces of $\alpha$-BBO with the particle displacement polarizations depicted. To begin with, the acoustic mode is a pure longitudinal mode, as required by the crystal symmetry. A pure mode makes coupling between the transducer and the crystal simpler, so the coupling should be efficient. Also with the acoustic mode being purely $\hat{z}$-polarized, the $M_2$ value is insensitive to the choice of Bragg matched optical waves for both $\vec{\hat{o}}$ $\rightarrow$ $\vec{\hat{o}}$ and $\vec{\hat{e}}$ $\rightarrow$ $\vec{\hat{e}}$ diffraction. With the $M_2$ value insensitive to the choice of the Bragg-matched optical waves, a crystallographically cut crystal can be used to simplify the fabrication process. As discussed in Chapter 3, the crystal growers confused the $X$ and $Y$ axis for our $\alpha$-BBO sample. In addition, I received a customized $\alpha$-BBO AO device with the $Y$ and $Z$ axes of the crystal confused. Hence, a customized crystal cut reduces the probability of the designed AOFS from being correctly fabricated.
By choosing a longitudinal acoustic mode, I have decided to use a relatively fast acoustic mode when compared to the shear acoustic modes. The Z-propagating acoustic mode has velocity of $V_a = 3258 \text{ m/s}$. For an efficient AOFS, you want the acoustic mode to be slow because $M_2 \sim V_a^{-3}$; however, fast acoustic modes result in shorter rise times and smaller time delays in the device. Also, a faster acoustic mode has a smaller momentum vector, so a larger frequency change $\Delta f_o$ is required to Bragg mismatch the AO geometry. In addition, there is no walk-off for this acoustic mode, so I do not need to cut the crystal or strategically place the transducer to compensate for the acoustic walk-off. Lastly, the acoustic slowness surface of the longitudinal mode in the Z-direction has a relative curvature of $b = 0.93$ (see Chapter 2 for definition of $b$). A smaller relative curvature allows for a smaller transducer height without suffering the consequences of acoustic diffraction. Decreasing the height of the transducer increases the acoustic power density, and thus improves the diffraction efficiency (in %/W) of the device.

Figure 7.3 illustrates the AO interaction of the AOFS in momentum space for an optical wavelength of $\lambda = 313 \text{ nm}$ and an RF frequency $f_a = 310 \text{ MHz}$. Figure 7.3b is a magnified version of Fig. 7.3a to better illustrate the AO interaction. As the figure shows, the optics and acoustics propagate along the Y and Z axes respectively. The internal and external diffraction full angles for $\lambda = 313 \text{ nm}$ and $f_a = 310 \text{ MHz}$ are $0.97^\circ$ and $1.67^\circ$ respectively. For $\lambda = 280 \text{ nm}$ and $f_a = 422 \text{ MHz}$, the internal and external diffraction angles are $1.16^\circ$ and $2.04^\circ$.

Figure 7.3 also illustrates that the shear acoustic wave is in a nearly Bragg-matched AO geometry. Even more bizarre is that this shear wave is at the point of acoustic conical refraction. Fortunately the effective photoelastic coefficient for these shear acoustic waves is nearly equal to zero for both $\hat{x}$ and $\hat{z}$-polarized light, so barely any optical light will be lost to diffraction if shear acoustic waves are accidentally launched. The diffraction efficiency of the device will still decrease if acoustic power is lost to the shear modes, but by launching a pure longitudinal wave the device should lose very little power to the shear waves.

Figure 7.4 illustrates the designed AOFS. The crystal is cut along the crystalline axes with the face opposite the transducers cut with a tilt of a few degrees in both directions to Bragg
Figure 7.2: XZ and YZ cross-section of the acoustic slowness surfaces in α-BBO. In our longitudinal AOFS design, the acoustic wave propagates along the Z axis (represented by the green vector in these plots). The purple vectors depict the acoustic wave particle displacement polarization. The normalized acoustic eigen-polarizations are projected into the plane of the cross-section, so shear polarizations pointing out of the page are represented by a dot and are hard to see while longitudinal polarizations, in contrast, are easier to see and have longer looking vectors.

mismatch the reflected acoustic wave and prevent the acoustic wave from propagating straight back to the transducer. For both devices, the transducers are centrally located. The transducer for the first AOFS is a simple 0.5 × 6 mm transducer. The second transducer is a 15.45 mm long phased-array with 30 fingers. Each finger is separated by \( d = 515 \pm 10 \mu m \) and have a width of \( w = 383 \pm 10 \mu m \), leaving a 133±14 \( \mu m \) gap between fingers (see Appendix C for why I chose said dimensions). An acoustic phased-array transducer is a beam steering technique used to increase the efficiency-bandwidth product of an AO device. A long phased-array transducer produces a narrow angular radiation lobe that is ideal for maximizing the diffraction efficiency. Varying the applied RF frequency to the transducer steers the lobe of radiation so that the AO device remains well phased matched over a large bandwidth with only quadratic, rather than linear, deviation from perfect Bragg matching with detuning from the central frequency \( f_0 \). Two types of phased-arrays
Figure 7.3: AO interaction of our AOFS in momentum space for $\lambda = 313$ nm and $f_a = 310$ MHz. The green arrow represents the acoustic vector $\vec{K}_A$ and the red arrows represent the incident and diffracted optical vectors $\vec{k}_i$ and $\vec{k}_d$.

are commonly used, the constant time delay array and the constant phase delay array [86]. The constant time delay array uses a stepped-staircase transducer to create fixed time delay between adjacent transducers in the array. However, this stepped-staircase is difficult to manufacture and therefore will not be discussed any further. In the constant phased delay array, adjacent transducers have a constant $180^\circ$ phase delay. To create this $180^\circ$ phase delay, the RF is applied to alternate transducers that lie on top of a floating ground plane [31].

I have optimized the transducers length to achieve the required bandwidths for the the $^9\text{Be}^+$ and $^{24}\text{Mg}^+$ experiments while maximizing the diffraction efficiency. For the 6 mm long transducer, we expect the AOFS to have a bandwidth of 293-324 MHz and 409-434 MHz for the 313 nm and 280 nm light respectively. Figure 7.5a illustrates the calculated acousto-optic bandwidth for the 6 mm long transducer for 313 nm light. Figure 7.5b illustrates the performance I expect for the phased-array over various wavelengths. As Fig. 7.5b illustrates, I expect the acousto-optic bandwidth to increase to about 110MHz. AOFS with an acousto-electric bandwidth tuned to operate over the 250-450 MHz would be able to work for both experiments.
Figure 7.4: AOFS design in $\alpha$-BBO for (a) a single transducer and (b) a phased-array transducer. The $\alpha$-BBO crystal is cut along the crystalline axes with transducers launching longitudinal acoustic waves along the Z-axis. Appropriate cross-sections of the acoustic slowness surfaces are rendered on the crystal faces to provide information about the launched acoustic wave.

For a phased-array transducer with a finger width of $w = 0.742d$, the peak power of the side lobe is 0.461 of the peak power of a regular transducer with the same total length $L$ (see Appendix C). As mentioned earlier, the phased-array transducer increases the AO bandwidth of the device, allowing me to increase the length of the transducer. If I increase the transducer length $L$ by $2.17 \left( = \left( \frac{1}{0.461} \right)^{-1} \right)$, the diffraction efficiency will be equivalent to a normal transducer with the same total length $L$ and will hopefully still have a large AO bandwidth, as increasing the length $L$ decreases the bandwidth. For the AOFS design, the phased array transducer is 2.67 times longer than the normal transducer and has a 3.67 times larger bandwidth, so the phased-array for this design is very advantageous in terms of the efficiency-bandwidth product.

For $\lambda = 313$ nm and $f_a = 310$ MHz, I expect the $0.5 \times 6$ mm transducer and the phased-array to have a 75% diffraction at 0.19 W and 0.38 W, respectively. For $\lambda = 280$ nm and $f_a = 422$ MHz, I expect the $0.5 \times 6$ mm transducer and the phased-array to have a 75% diffraction at 0.14 W and
Figure 7.5: (a) Calculated acousto-optic bandwidth for the 0.5 × 6 mm transducer for 280 nm light. The red line indicates 84% AO diffraction. (b) Acousto-optic bandwidth for the phased array at various wavelengths. The vertical red and black lines mark 310MHz and 422MHz, which are the center RF frequencies for the 313nm and 280nm light for the NIST ion experiments.

0.29 W, respectively. I expect the smaller wavelength to have a larger diffraction efficiency because the diffraction efficiency scales inversely proportionally to the inverse square of the wavelength ($DE \sim \lambda^{-2}$) and a smaller wavelength has a larger index of refraction ($DE \sim n^6$).

7.4 Measured Performance

My collaborators at Brimrose made three different AOFSs based on the designs discussed in the previous section. Two of the AOFSs are the 0.5 × 6 mm transducer design with center frequencies at $f_c = 350$ and 450 MHz. The third device is the phased-array transducer design. Unfortunately, the phased-array transducer never diffracted a sufficient amount of light. Figure 7.6 explains why the phased-array device does not diffract light because the measured reflection coefficient of the input voltage $S_{11}$ of the device is near zero, which means that most of the electrical power is being reflected with little power being converted into acoustic waves. Thus, the diffracted light was barely measurable. I will no longer discuss the phased-array results because I was unable to make any significant AO measurements with this device. The measured $S_{11}$ coefficients for the other devices, however, show little electrical power being reflected around the center frequencies of
I will now discuss the AO performance of the two 0.5 × 6 mm transducer devices. For all of my measurements, I used $\lambda = 532$ nm light. Figure 7.7 illustrates the AO bandwidth of the two devices at their center frequencies. I measured an AO 3 dB bandwidth of 31 and 24 MHz, which agrees with the calculated AO bandwidths of 30 and 23 MHz. These bandwidths will increase at smaller wavelengths because the optical momentum surface scales as $k_o = \frac{2\pi}{\lambda}n(\lambda)$. With a larger optical momentum surface, the curvature will be smaller, so the rate of Bragg mismatch will be slower as a function of AO frequency $f_a$.

I measured the diffraction efficiency of the two devices at different acoustic powers and determined the measured $M_2$ value by fitting to these points. Figure 7.8 illustrates our measurements. For the $f_c = 350$ and $f_c = 450$ MHz devices, I measured $M_2$ values of 3.17 and $2.00 \times 10^{-15} s^3/kg$, respectively. I expect $M_2 = 7.89 \times 10^{-15} s^3/kg$ for $\lambda = 532$ nm light, so I am losing about 60-75% of the acoustic power. According to my $S_{11}$ measurements, the devices are losing less than 1% of
Figure 7.7: Measured AO bandwidth of the transducer with a center frequency of (left) 350 and (right) 450 MHz and $\lambda = 532$ nm. The diffraction efficiency has been normalized to the peak value.

the electrical power to reflection at the center frequencies. Some of the acoustic power could be lost by launching shear modes, but this is unlikely because launching a pure longitudinal mode is not difficult. Unfortunately I was unable to acousto-optically measure the power in the shear mode because the effective photoelastic coefficient is zero for this device. Even with misalignment of the crystal, the $M_2$ value and polarization of the acoustic mode should not significantly change, as Figs. 7.1 and 7.2 illustrate. Lastly, acoustic power could be lost to heating, but 60-75% is an excessive amount of power to be lost to heat. If the efficiency of the transducer could be improved, then the diffraction efficiency could be improved by over a factor of two.

With the measured $M_2$ values, I expect for $\lambda = 313$ nm and $\lambda = 280$ nm the devices to have a 75% diffraction efficiency at 0.48 W and 0.57 W. From my calculations in the previous section, I was hoping that the required acoustic powers would be 0.19 and 0.14 W instead.

### 7.5 Conclusion

I have measured the elastic and photoelastic properties of $\alpha$-BBO with both standard and my own modern versions of traditional experiments. I have also developed graphical representations for discovering efficient AO geometries. From my body of work, I was able to identify and design an AOFS that should be at least five times more efficient than current fused silica devices. Such
improvements should eliminate the heating issues that the quantum computing community are currently facing with fused silica devices.
Chapter 8

Conclusion

Acousto-optic devices are a versatile technology with a broad range of applications including acousto-optic scanners, filters, mode lockers, frequency shifters, and modulators. Currently there is no UV transparent acousto-optic device that can satisfy the experimental requirements of the atomic clock and quantum computing communities. This thesis presented my experimental research efforts for discovering a UV transparent media that could be used to make an efficient acousto-optic device.

This thesis presented techniques for measuring both the elastic-stiffness $C_{ijkl}$ and photoelastic $p_{ijkl}$ coefficients, which both need to be known for designing acousto-optic devices. For measuring both $C_{ijkl}$ and $p_{ijkl}$, I reviewed well established techniques (eg resonant ultrasound spectroscopy and the Dixon method) as well as novel techniques I pioneered (eg modified Schaefer–Bergmann measurements). I applied the various measuremental techniques to both $\alpha$ and $\beta$ barium borate and was able to measure their elastic-stiffness and photoelastic coefficients with a precision not previously published.

I also presented in this thesis various graphical representations that facilitate in discovering an acousto-optic geometry with desirable qualities. These graphical representations allow for the acousto-optic potential of a given medium to be maximized. I applied the graphical representations to $\alpha$-BBO to aide in designing an acousto-optic device that is a factor of three times more efficient that the current state-of-the-art, UV transparent acousto-optic devices.

The research presented in this thesis provides a manual for measuring the required physical
parameters and for creating algorithms to design an effective acousto-optic device for any new material. As more potential acousto-optic materials are discovered, the research in this thesis will be essential in optimizing any corresponding device.
Bibliography


[34] L. Bergmann and H. Hatfield. Ultrasonics and Their Scientific and Technical Applications. G. Bell and Sons Limited, 1938.


Appendix A

Analytic Cross-sections for Trigonal Slowness Surfaces

Below I provide the analytic solutions to the YZ, XY, and XZ cross-sections of the acoustic velocity and slowness surface for crystals with trigonal \( \overline{3}m \) symmetry. There are three solutions for each cross-section. The slowness is given by \( 1/V_m \).

A.1 YZ Cross-section Solutions

The YZ-cross-section has the simplest solutions. The solutions \( V_2 \) and \( V_3 \) represent the quasi-longitudinal mode and the slow-shear mode, respectively. The first solution \( V_1 \) is a pure-shear mode that is an ellipse and is not visible by SBD. Both \( C_{11} \) and \( C_{12} \) explicitly appear in this solution, but only as a representation of \( C_{66} \). This point is clearly illustrated by looking at the similarities of the derivatives of \( C_{11} \) and \( C_{12} \) in Fig. 3.7. With the pure shear mode being unable to diffract light, this cross-section can measure all of the elastic-stiffness coefficients except for \( C_{12} \).

\[
\rho V_1^2 = C_{44} \cos^2[\theta] + \frac{1}{2}(C_{11} - C_{12})\sin^2[\theta] + C_{14}\sin[2\theta]
\]

\[
2\rho V_2^2 = C_{44} + C_{11}\sin^2[\theta] + C_{33}\cos^2[\theta] - C_{14}\sin[2\theta]
\]

\[
\pm \sqrt{(C_{11}\sin^2[\theta] - C_{33}\cos^2[\theta] + C_{44}\cos[2\theta] - C_{14}\sin[2\theta])^2 + ((C_{13} + C_{44})\sin[2\theta] - 2C_{14}\sin^2[\theta])^2}
\]
A.2 XY Cross-section Solutions

I solved for the XY and XZ cross-sections using Vieta’s substitution. The XY-cross-section allows for the optimization of $C_{11}$, $C_{12}$, $C_{14}$, and $C_{44}$, but not $C_{13}$ or $C_{33}$.

\[
R = \frac{1}{216} \left( (3C_{11} - C_{12} - 4C_{44}) \left( (C_{12} + C_{44})(3C_{11} + C_{12} - 2C_{44}) - 9C_{14}^2 \right) - 27C_{14}^2(C_{11} + C_{12}) \cos[6\phi] \right)
\]

\[
Q = \frac{1}{36} \left( 6C_{11}C_{44} - 3C_{11}^2 - C_{12}^2 - 2C_{44}(C_{12} + 2C_{44}) - 12C_{14}^2 \right)
\]

\[
D = Q^3 + R^2
\]

\[
S = (R + \sqrt{D})^{1/3}
\]

\[
T = (R - \sqrt{D})^{1/3}
\]

\[
A = \frac{1}{6}(3C_{11} - C_{12} + 2C_{44})
\]

\[
\rho V_1^2 = A + (S + T)
\]

\[
\rho V_2^2 = A - \frac{1}{2}(S + T) + \frac{i}{2}\sqrt{3}(S - T)
\]

\[
\rho V_3^2 = A - \frac{1}{2}(S + T) - \frac{i}{2}\sqrt{3}(S - T)
\]

The only angle dependence of the solution is a $6\phi$ term, which means that the acoustic velocity pattern repeats itself every $60^\circ$. This reflects the three-fold symmetry of the Z-axis. If $D > 0$, then one of the solutions will be real and the other two will be complex solutions, which is not physically possible. If $D = 0$, then all of the solutions are real and two of the solutions are degenerate. This will happen when $C_{44} = \frac{C_{11} - C_{12}}{2}$ and $C_{14} = 0$, which is the condition for an isotropic crystal. If $D < 0$, then all three roots are real and unequal.

When $D < 0$, then $S$ and $T$ will be imaginary and can thus be rewritten as

\[
S = r^{1/3}e^{i\theta/3}
\]

\[
T = r^{1/3}e^{-i\theta/3}
\]

\[
\theta = \cos[R/\sqrt{-Q^3}]
\]

\[
r = -Q^3
\]
With these new definitions for $S$ and $T$, the solutions become

$$\rho V_1^2 = A + 2r \cos[\theta/3]$$
$$\rho V_2^2 = A + 2r \cos[(\theta + 2\pi)/3]$$
$$\rho V_3^2 = A + 2r \cos[(\theta + 4\pi)/3]$$

$V_1$, $V_2$, and $V_3$ correspond to the longitudinal, slow-shear, and fast-shear modes respectively

### A.3 XZ Cross-Section Solution

The XZ cross-section allows for optimization of all elastic-stiffness coefficients. Below $R$, $Q$, and $A$ are defined. The solutions to the velocities of the three different modes are found by following the same procedure as for the XY cross-section.

$$R = \frac{1}{6912} \left[ 2 \left( 9C^2_{11}(5C_{12} + C_{33} + 4C_{44}) - 5C^2_{12} - 3C^2_{12}(C_{33} + 4C_{44}) - 18C_{11}(3C^2_{14} + C_{33}(C_{12} + C_{33}) + 4C_{12}C_{44} + 4C^2_{44}) + 6C_{12}(6C^2_{13} - 27C^2_{14} + C^2_{33} + 12C_{13}C_{44} + 10C^2_{44}) + 4 \left( 9C^2_{14} + 9C^2_{14}(-7C_{33} + 16C_{44}) + 18C_{13}(6C^2_{14} + C_{33}C_{44}) + C_{33}(10C^2_{33} - 24C_{33}C_{44} + 33C^2_{44}) \right) \right) - 3 \left( 3C^2_{11}(15C_{12} + C_{33} + 14C_{44}) - 5C^2_{12} - C^2_{12}(C_{33} + 14C_{44}) + 2C_{12}(6C^2_{13} - 57C^2_{14} - C^2_{33} + 4(3C_{13} + C_{33})C_{44} + 18C^2_{44}) - 6C_{11}(33C^2_{14} - (C_{33} - 6C_{44})(C_{33} + 2C_{44}) + C_{12}(C_{33} + 14C_{44})) \right) + 4 \left( 3C^2_{14}(9C_{33} + 10C_{44}) - 3C^2_{13}(C_{33} - 2C_{44}) - (C_{33} - 2C_{44})(10C^2_{33} - 8C_{33}C_{44} + 11C^2_{44}) + 6C_{13}(6C^2_{14} + C_{44}(-C_{33} + 2C_{44})) \right) \right) \cos[2\theta]$$

$$+ 6 \left( 6C_{12}C^2_{14} - C^2_{12} - 12C_{12}C^2_{33} + 144C_{13}C^2_{44} + C^2_{12}C_{33} - 12C^2_{12}C_{33} + 84C^2_{14}C_{33} - 2C_{12}C^2_{33} + 8C^2_{33} \right) + 4 \left( \left( C^2_{12} + 6C_{12}C_{13} + 4C^2_{14} + 6C_{13}C_{33} + 8C^2_{33} \right)C_{44} - 4(C_{12} - 5C_{33})C_{44} + 3C^2_{13}(3C_{12} - C_{33} + C_{44}) \right) 6C_{11} \left( C_{33}(C_{12} - 21C^2_{14} + C_{33}) - C_{12}C_{44} - 4C^2_{44} \right) \cos[4\theta]$$

$$+ \left( C^2_{12} + 36C_{12}C^2_{14} - 54C_{12}C^2_{14} + 432C_{13}C^2_{44} - 3C^2_{12}C_{33} - 36C^2_{12}C_{33} + 324C^2_{14}C_{33} - 6C_{12}C^2_{33} + 8C^2_{33} + 18C_{11}(15C^2_{14} + (C_{33} - 2C_{44})(-C_{12} + C_{33} - 2C_{44})) \right) + 6 \left( C^2_{12} + 4C_{12}(3C_{13} + C_{33}) + 4(3C^2_{14} - 9C^2_{14} - 3C_{13}C_{33} - 2C^2_{33}) \right)C_{44} + 12(C_{12} + 12C_{13} + 5C_{33})C^2_{44} + 8C^2_{44} - 9C^2_{12}(C_{12} - C_{33} + 2C_{44}) \right) \cos[6\theta]$$

$$Q = \frac{1}{288} \left[ 6C_{11}(C_{33} + 2C_{44}) - 9C^2_{12} - 3(C^2_{12} + 4(C^2_{13} + 7C^2_{14})) - 2C_{33}(C_{12} + 6C_{33}) - 4(C_{12} + 6C_{13} - 4C_{33})C_{44} + \frac{1}{28} \left( 3C^2_{11} + C^2_{12} + 12C^2_{14} - 4C^2_{33} - 6C_{11}C_{44} + 2C_{12}C_{44} + 8C_{33}C_{44} \right) \cos[2\theta] \right.$$

$$\left. + 4 \left( 3C^2_{11} + 2C^2_{12} + 12C^2_{14} - 4C^2_{33} - 6C_{11}C_{44} + 2C_{12}C_{44} + 8C_{33}C_{44} \right) \cos[2\theta] \right]$$

$$\left. + \left( 3C^2_{11} + C^2_{12} - 12(C^2_{13} + 3C^2_{14}) - 2C_{12}C_{33} + 4C^2_{33} + 6C_{11}(C_{33} - 2C_{44}) + 4(C_{12} - 6C_{13} - 4C_{33})C_{44} + 4C^2_{44} \right) \cos[4\theta] \right)$$

$$A = \frac{1}{12} \left( 3C_{11} - C_{12} + 2C_{33} + 6C_{44} + ( - 3C_{11} + C_{12} + 2(C_{33} + C_{44}) \right) \cos[2\theta] \right)$$
Appendix B

Maximum Acousto-Optic Figure of Merit Search

This appendix provides the results of our $M_2$ graphical representations for Lead Molybdate ($\text{MoO}_4\text{Pb}$), Potassium Dideuterium (KDP), and Tellurium Dioxide ($\text{TeO}_2$). For each material, a table is provided that lists the relevant physical parameters used for the calculations.

B.1 Lead Molybdate
Table B.1: Physical properties of PbMoO$_4$ [81].

<table>
<thead>
<tr>
<th>General</th>
<th>Elastic Stiffness Tensor (GPa)</th>
<th>Photoelastic Tensor</th>
<th>Optical Properties</th>
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<td>$p_{11}$</td>
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<td>$n_e$</td>
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<td>$p_{16}$</td>
<td>$\lambda$ = 632.8 nm</td>
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<tr>
<td></td>
<td></td>
<td>$p_{31}$</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>$p_{33}$</td>
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</tr>
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<td>$\phi$</td>
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<td>$\theta$</td>
<td>$\phi$</td>
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<td>Quasi-Shear Slow</td>
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<td>-50.26</td>
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<td>172.12</td>
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<td>Quasi-Shear Fast</td>
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<td>128.17</td>
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<td>Quasi-Shear Slow</td>
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<td>Quasi-Shear Fast</td>
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<td>Quasi-Shear Slow</td>
<td>-139.69</td>
<td>179.33</td>
<td>-67.71</td>
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Figure B.1: $M_2$ surfaces for PbMoO$_4$ with $\lambda = 633$ nm and $f_a = 80$ MHz acoustic wave.

Table B.3: Maximum $M_2$ values and corresponding tangentially-degenerate AO geometries in PbMoO$_4$ with $\lambda = 632.8$ nm.

<table>
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<tr>
<th>Acous. Mode</th>
<th>$f_a$ (GHz)</th>
<th>$\vec{k}_{ord}$</th>
<th>$\vec{k}_{ext}$</th>
<th>$\vec{K}_a$</th>
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<td>-69.44 59.83</td>
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<td>Quasi-Shear Fast</td>
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<td>2.47 179.00 -12 179</td>
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<td>Quasi-Shear Slow</td>
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<td>-87.61 178.92 -102 179</td>
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<td>27.63</td>
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</table>
Figure B.2: $M_2$ surfaces for tangentially-degenerate AO geometries in PbMoO$_4$.

Table B.4: Maximum $M_2$ values and corresponding sub-collinear AO geometries in PbMoO$_4$.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$f_a$ (MHz)</th>
<th>$\vec{k}_{ord}$</th>
<th>$\vec{k}_{ext}$</th>
<th>$\vec{K}_a$</th>
<th>$M_2^{max}$ $(10^{-15} s^3/kg)$</th>
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<tr>
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<td>$\phi$</td>
<td>$\theta$</td>
<td>$\phi$</td>
<td>$\theta$</td>
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<tr>
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</tr>
<tr>
<td>Quasi-Shear Fast</td>
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Figure B.3: $M_2$ surfaces for sub-collinear AO geometries in PbMoO$_4$. 
Table B.5: Maximum $M_2$ values and corresponding parallel-tangents AO geometries in PbMoO$_4$.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$f_a$ (MHz)</th>
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<th>$\bar{k}_{ext}$</th>
<th>$\bar{K}_a$</th>
<th>$M_{max}^{max}$ $(10^{-15}s^3/kg)$</th>
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<td>1.06</td>
<td>71</td>
<td>71.00</td>
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Figure B.4: $M_2$ surfaces for parallel-tangents AO geometries in PbMoO$_4$.
### B.2 Potassium Dideuterium (KDP)

Table B.6: Maximum $M_2$ values and corresponding acousto-optic geometries for a given type of diffraction in KDP with $\lambda = 300$ nm and $f_a = 80$ MHz acoustic wave.

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<td>$\theta$</td>
<td>$\phi$</td>
<td>$\theta$</td>
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<tr>
<td>Isotropic diffraction $\hat{o} \to \hat{o}$</td>
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<td></td>
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<td></td>
</tr>
<tr>
<td>Quasi-Long.</td>
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<td>179.89</td>
<td>135.00</td>
<td>179.89</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
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<td>174.02</td>
<td>53.91</td>
<td>174.32</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>133.24</td>
<td>5.63</td>
<td>130.88</td>
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<tr>
<td>Isotropic diffraction $\hat{e} \to \hat{e}$</td>
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<td>Quasi-Shear Slow</td>
<td>-42.67</td>
<td>39.44</td>
<td>-42.10</td>
<td>39.13</td>
</tr>
<tr>
<td>Anisotropic diffraction $\hat{e} \to \hat{o}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>146.70</td>
<td>179.58</td>
<td>116.66</td>
<td>179.58</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>64.62</td>
<td>154.12</td>
<td>64.61</td>
<td>154.29</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>21.57</td>
<td>0.91</td>
<td>-8.26</td>
<td>0.94</td>
</tr>
</tbody>
</table>
Figure B.5: $M_2$ surfaces for KDP with $\lambda = 300$ nm and $f_a = 80$ MHz acoustic wave.

Figure B.6: $M_2$ surfaces for tangentially-degenerate AO geometries in KDP.
Table B.7: Maximum $M_2$ values and corresponding tangentially-degenerate AO geometries in KDP with $\lambda = 300$ nm.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$f_a$ (GHz)</th>
<th>$\vec{k}_{ord}$</th>
<th>$\vec{k}_{ext}$</th>
<th>$\vec{K}_a$</th>
<th>$M_{max}^{2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$\phi$</td>
<td>$\theta$</td>
<td>$\phi$</td>
<td>$\theta$</td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>1.99</td>
<td>34.13</td>
<td>122.09</td>
<td>24</td>
<td>115</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>1.27</td>
<td>30.32</td>
<td>90.00</td>
<td>17</td>
<td>90</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>1.27</td>
<td>32.32</td>
<td>90.00</td>
<td>19</td>
<td>90</td>
</tr>
</tbody>
</table>

Table B.8: Maximum $M_2$ values and corresponding sub-collinear AO geometries in KDP.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$f_a$ (MHz)</th>
<th>$\vec{k}_{ord}$</th>
<th>$\vec{k}_{ext}$</th>
<th>$\vec{K}_a$</th>
<th>$M_{max}^{2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$\phi$</td>
<td>$\theta$</td>
<td>$\phi$</td>
<td>$\theta$</td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>262.0</td>
<td>101.00</td>
<td>67.32</td>
<td>100.30</td>
<td>67.50</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>112.6</td>
<td>135.00</td>
<td>49.25</td>
<td>135.00</td>
<td>49.25</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>189.8</td>
<td>-150.16</td>
<td>96.51</td>
<td>-151.51</td>
<td>96.18</td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>255.0</td>
<td>101.69</td>
<td>66.04</td>
<td>101.01</td>
<td>66.19</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>110.1</td>
<td>135.00</td>
<td>48.70</td>
<td>135.00</td>
<td>49.16</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>185.2</td>
<td>31.13</td>
<td>82.83</td>
<td>29.84</td>
<td>83.13</td>
</tr>
</tbody>
</table>

Figure B.7: $M_2$ surfaces for sub-collinear AO geometries in KDP.
Table B.9: Maximum $M_2$ values and corresponding parallel-tangents AO geometries in KDP.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$f_a$ (MHz)</th>
<th>$\bar{k}_{ord}$</th>
<th>$\bar{k}_{ext}$</th>
<th>$\bar{K}_a$</th>
<th>$M_2^{\text{max}}$ (10$^{-15}$s$^3$/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quasi-Long.</td>
<td>306</td>
<td>-130.00</td>
<td>-130.00</td>
<td>107.61</td>
<td>0.06</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>214.</td>
<td>-65.00</td>
<td>65</td>
<td>70.82</td>
<td>0.83</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>127.59</td>
<td>90.00</td>
<td>90</td>
<td>109.71</td>
<td>9.94</td>
</tr>
</tbody>
</table>

Figure B.8: $M_2$ surfaces for parallel-tangents AO geometries in KDP.
B.3 Tellurium Dioxide (TeO\textsubscript{2})

I chose to apply my graphical representations to TeO\textsubscript{2} because its large elastic anisotropy combined with its large photoelastic coefficients make it an exceptional AO material. Alternating weak-strong Te-O bonds and a star-shape truss structure on the basal plane create the large elastic anisotropy \cite{88}. The elastic anisotropy of TeO\textsubscript{2} creates a very slow shear-mode ($V_a = 623 \text{cm/s}$) along the [110] direction. This unusually slow acoustic mode of TeO\textsubscript{2} allows for exceptionally large $M_2$ values.

The magnitude and relative signs of the photoelastic coefficients of TeO\textsubscript{2} were measured by Uchida and Ohmachi using the Dixon-Cohen method \cite{89}. However, they used linearly polarized light for their measurements along the z-axis when elliptically polarized light is required for optically active crystals. Yano and Watanabe showed that not using elliptically polarized light significantly reduces the light into the Bragg-matched eigenmode in the Dixon-Cohen experiment, resulting in an underestimate of the measured effective photoelastic coefficient $p$ \cite{90}. For the $p = \frac{1}{2}|p_{11} - p_{22}|$ measurement, Yano and Watanabe measured $M_2 = 1200 \times 10^{-15} \text{s}^3/\text{kg}$ while Uchida and Ohmachi measured $M_2 = 793 \times 10^{-15} \text{s}^3/\text{kg}$. Unfortunately, Yano and Watanabe did not measure $p_{66}$, another measurement that requires light to propagate along the optic axes. In addition, Uchida and Ohmachi do not give explicit details such as applied acoustic frequency or optical polarizations for their $p = p_{66}$ measurement, so we do not know how much optical power went into the correct optical eigenmode. I have approximated that they underestimated the $p = p_{66}$ measurement by a factor of 1.4 based on the error for the $p = \frac{1}{2}|p_{11} - p_{22}|$ measurement and the relative acoustic velocities of the two measurements. Admittedly, this is a very imprecise guess.

I have taken the measurements from Uchida and Ohmachi along with the correction for $\frac{1}{2}|p_{11} - p_{22}|$ from Yano and Watanabe and the $p_{66}$ correction from my estimate, and did a least squares fit to determine the photoelastic coefficients listed in Table B.10. For the least squares fit, I used the elastic and piezoelectric coefficients and material density measured from \cite{88} to determine the acoustic velocities instead of the velocities measured by Uchida and Ohmachi. The $p_{13}$, $p_{31}$,
and $p_{33}$ coefficients were only slightly affected because their measurements do not require light to propagate along the optic axis. Uchida and Ohmachi do not explain how they measured $p_{44}$, so its value did not change. The largest changes are to the $p_{11}$, $p_{12}$, and $p_{66}$, as expected, even a sign change for $p_{11}$ from the Yano and Watanabe correction.

**Table B.10: Photoelastic coefficients of TeO$_2$**

<table>
<thead>
<tr>
<th>Constant</th>
<th>[44] Corrected Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>$p_{11}$</td>
<td>0.0074 -0.0088</td>
</tr>
<tr>
<td>$p_{12}$</td>
<td>0.187 0.218</td>
</tr>
<tr>
<td>$p_{13}$</td>
<td>0.340 0.341</td>
</tr>
<tr>
<td>$p_{31}$</td>
<td>0.0905 0.0957</td>
</tr>
<tr>
<td>$p_{33}$</td>
<td>0.240 0.242</td>
</tr>
<tr>
<td>$p_{44}$</td>
<td>$\sim -0.17$ $\sim -0.17$</td>
</tr>
<tr>
<td>$p_{66}$</td>
<td>-0.0463 -0.0558</td>
</tr>
</tbody>
</table>

**Table B.11: Physical properties of TeO$_2$** [88, 91, 81, 92].

<table>
<thead>
<tr>
<th>General</th>
<th>Elastic Stiffness Tensor (GPa)</th>
<th>Piezoelectric Props.</th>
<th>Optical Properties</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$c_{11}$</td>
<td>$c_{33}$</td>
<td>DE Const.</td>
</tr>
<tr>
<td></td>
<td>$c_{33}$</td>
<td>$c_{14}$</td>
<td>PE Const.</td>
</tr>
<tr>
<td></td>
<td>$c_{66}$</td>
<td>$c_{12}$</td>
<td>EO Const.</td>
</tr>
<tr>
<td></td>
<td>$c_{13}$</td>
<td></td>
<td>Transparency</td>
</tr>
<tr>
<td>Point Group</td>
<td></td>
<td></td>
<td>$n_o$</td>
</tr>
<tr>
<td>$\rho$ (g/cm$^3$)</td>
<td></td>
<td></td>
<td>$n_e$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$c_{33} = 24.9$</td>
<td>@632.8 nm</td>
</tr>
<tr>
<td>TeO$_2$</td>
<td>422</td>
<td>106.6</td>
<td>0.35-6.0 $\mu$m</td>
</tr>
<tr>
<td></td>
<td>5.984</td>
<td>26.6</td>
<td>2.2597</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>65.8</td>
<td>$e_{14} = 0.330$ (C/m$^2$)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>51.1</td>
<td>2.4125</td>
</tr>
<tr>
<td></td>
<td></td>
<td>22.9</td>
<td>$r_{41} = 0.62$ pm/V</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>86.9 (deg/m)</td>
</tr>
</tbody>
</table>
Table B.12: Maximum $M_2$ values and corresponding acousto-optic geometries for a given type of diffraction in TeO$_2$ with a $\lambda = 632.8$ nm and $f_a = 80$ MHz acoustic wave.

<table>
<thead>
<tr>
<th>Acous. Mode</th>
<th>$\vec{k}_i$</th>
<th>$\vec{k}_d$</th>
<th>$\vec{K}_a$</th>
<th>$M_2^{\text{max}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\phi$</td>
<td>$\theta$</td>
<td>$\phi$</td>
<td>$\theta$</td>
</tr>
<tr>
<td>Isotropic diffraction $\hat{o} \rightarrow \hat{o}$</td>
<td>90.00</td>
<td>160.20</td>
<td>-46.99</td>
<td>170.40</td>
</tr>
<tr>
<td>Isotropic diffraction $\hat{e} \rightarrow \hat{e}$</td>
<td>93.78</td>
<td>90.14</td>
<td>-6.90</td>
<td>156.49</td>
</tr>
<tr>
<td>Isotropic diffraction $\hat{e} \rightarrow \hat{o}$</td>
<td>-35.58</td>
<td>156.74</td>
<td>29</td>
<td>1</td>
</tr>
<tr>
<td>Anisotropic diffraction $\hat{e} \rightarrow \hat{e}$</td>
<td>-3.58</td>
<td>-6.70</td>
<td>29</td>
<td>1</td>
</tr>
<tr>
<td>Anisotropic diffraction $\hat{e} \rightarrow \hat{o}$</td>
<td>-130.00</td>
<td>179.02</td>
<td>45</td>
<td>90</td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>90.00</td>
<td>160.20</td>
<td>0</td>
<td>70</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>169.38</td>
<td>173.05</td>
<td>15</td>
<td>96</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>96.18</td>
<td>9.19</td>
<td>46</td>
<td>96</td>
</tr>
<tr>
<td>Quasi-Long.</td>
<td>93.78</td>
<td>90.14</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Quasi-Shear Fast</td>
<td>88.80</td>
<td>6.70</td>
<td>16</td>
<td>92</td>
</tr>
<tr>
<td>Quasi-Shear Slow</td>
<td>-174.91</td>
<td>8.81</td>
<td>44</td>
<td>85</td>
</tr>
</tbody>
</table>

Figure B.9: $M_2$ surfaces in TeO$_2$ for $\lambda = 632.8$ nm and $f_a = 80$ MHz acoustic wave.
Appendix C

Phased-Array Transducers

In this appendix we analyze the performance of the constant-phased-delay-array transducer. Figure C.1a illustrates an example of the output of a phased-array with a constant 180° phase delay between adjacent transducers. The launched acoustic waves can be decomposed into a spectrum of acoustic plane-wave eigenmodes that interact with the optical input. Figure C.2a shows the acoustic wave in the device and the resulting acoustic wavevector $\vec{K}_a$ for three different applied RF frequencies. The resulting acoustic wavevector is given by

$$\vec{K}_a = 2\pi f_a/V_a(\theta) \hat{z} + \pi/d\hat{y}. \quad (C.1)$$

Here $\hat{y}$ is the direction of the transducer elements, $f_a$ is the applied RF frequency, $d$ is the separation between adjacent phased-array elements, and $V_a(\theta)$ is the acoustic wave velocity in the direction of the wavevector.

Equation C.1 shows that the acoustic wavevector $\vec{K}_a$ traces out a straight line in the $\hat{z}$ direction as the RF frequency is varied. To achieve a large AO bandwidth, the AO device is rotated so that the wavevector $\vec{K}_a$ traces out a line that is tangent to the optical momentum surface, as illustrated in Fig. C.2c. Such an AO geometry is to first order well phased matched over a wide bandwidth.

To better understand the performance of the phased-array transducer, we must take the Fourier transform of the amplitude output. The amplitude output of the transducer as depicted in Fig. C.1a can be described as

$$t[y] = \left[ \text{rect} \left( \frac{y}{w} \right) \otimes \frac{1}{2d} \left( \text{comb} \left( \frac{y}{2d} + \frac{1}{4} \right) - \text{comb} \left( \frac{y}{2d} - \frac{1}{4} \right) \right) \right] \text{rect} \left( \frac{y}{N} \right). \quad (C.2)$$
Figure C.1: (a) Illustration of the output from the phased array at a certain snapshot in time when half the transducers are outputting the peak longitudinal strain, making the other transducers that are 180° out of phase output the negative peak strain. $L$ is the total length of the transducer, $w$ is the length of an individual transducer element, and $d$ is the separation between two adjacent transducer elements. (b) Power output as a function of transverse spatial frequency. The power is normalized by the peak power of a non-phased-array transducer with a length $L$.

where $w$ is the width of a transducer element and $N$ is the total number of transducer elements.

Taking the Fourier transform we get

$$\mathcal{F}\{t[y]\} = \left[w \text{sinc}(w f_y) \text{comb}(2df_y) \left(e^{i\pi f_y d} - e^{-i\pi f_y d}\right)\right] \otimes N d \text{sinc}(Ndf_y),$$

which can be simplified using Euler’s identity $\sin(x) = \frac{e^{ix} - e^{-ix}}{2i}$:

$$\mathcal{F}\{t[y]\} = \left[2wi \text{sinc}(w f_y) \text{comb}(2df_y) \sin(\pi f_y d)\right] \otimes N d \text{sinc}(Ndf_y).$$

We can further simplify by using the identity $\frac{1}{\Delta} \text{comb}(\frac{y}{\Delta}) \equiv \sum_m \delta(y - m\Delta)$, we have

$$\mathcal{F}\{t[y]\} = \left[\sum_{m=-\infty}^{\infty} 2wi \text{sinc}(w f_y) \left(\frac{1}{2d} \delta(f_y - \frac{m}{2d})\right) \sin(\pi f_y d)\right] \otimes N d \text{sinc}(Ndf_y)$$

$$= Nwi \sum_{m=-\infty}^{\infty} \text{sinc}\left(\frac{wm}{2d}\right) \sin\left(\frac{\pi m}{2}\right) \sin\left(N d \left[f_y - \frac{m}{2d}\right]\right).$$

The intensity distribution becomes

$$I[f_y] = (Nw)^2 \left[\sum_{m=-\infty}^{\infty} \text{sinc}\left(\frac{wm}{2d}\right) \sin\left(\frac{\pi m}{2}\right) \sin\left(N d \left[f_y - \frac{m}{2d}\right]\right)\right]^2.$$
Figure C.2:

Acoustic beam steering with a 180° phase delay transducer. (a) Illustration of the resulting acoustic wave and wavevector $K_a$ in the AOFS for three different applied RF frequencies. (b) The transducer radiation power. (c) Illustration of the AO interaction in momentum space. Notice how the acoustic wavevectors trace out a line that is perpendicular to the transducer and tangent to the optical momentum surface.
Figure C.1b plots Eq. C.6 and therefore illustrates the radiation power as a function of spatial frequency of the acoustic wave. There are two main power lobes with several smaller lobes, so immediately we can tell that with the 180° phase delay array we lose at least half its power to unwanted wavevectors. To maximize the total power of the phased-array, we can optimize the width of the transducer elements \( w \), but the element separation \( d \) and the transducer total length \( L \) are held fixed for phase-matching and acousto-optic bandwidth considerations. We only want to optimize the power of one of the side lobes. For this analysis we will optimize the \( m = 1 \) side lobe, making \( \sin \left( \frac{\pi m}{2} \right) = 1 \) in Eq. C.6. We will also set \( f_y = \frac{1}{2d} \) so that the last term in Eq. C.6 equals one as well. With these simplifications, Eq. C.6 reduces to

\[
I \left[ \frac{1}{2d} \right] = (Nw)^2 \text{sinc}^2 \left( \frac{w}{2d} \right). \tag{C.7}
\]

Next we need to consider that the power to drive a transducer equals summation of the intensity over the area of the device \( (P = \int I \cdot dA) \). Assuming the intensity is constant across the transducer that has an area of \( Nw \), the power in the \( m = 1 \) side lobe is

\[
P \left[ \frac{1}{2d} \right] = Nw \text{sinc}^2 \left( \frac{w}{2d} \right). \tag{C.8}
\]

Equation C.8 is maximized when \( w = 0.742d \). For our AOFS, we designed our phased-array transducer to have \( w = 0.742d \), or approximately \( \frac{3}{4}d \), in order to maximize the efficiency of the device.

For a phased-array transducer with \( w = 0.742d \), the peak power of the side lobe is 0.461 of the peak power of a regular transducer with the same total length \( L \). As we mentioned earlier, the phased-array transducer increases the AO bandwidth of the device, allowing us to increase the length of the transducer. If we increase the transducer length \( L \) by \( 2.17 \left( = \left( \frac{1}{0.461} \right)^{-1} \right) \), the diffraction efficiency will be equivalent to a normal transducer with the same total length \( L \) and will hopefully still have a large AO bandwidth, as increasing the length \( L \) decreases the bandwidth.