Characterization of Silicon Carbide Crystal used for Electro-Optic Measurements

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Abstract

A new technique for non-destructive measurements of ultra-short relativistic electron bunches utilizes the electro-optic (EO) detection of the electromagnetic field from the bunch. This technique is unique in that it uses both the incoherent visible radiation and coherent THz radiation from the same bunch, on a bunch-by-bunch basis. 6H Silicon Carbide has favorable properties as an EO crystal due to the high resonance frequency of optically active phonons. To maximize the EO signal, the bunch radiation inside the crystal should be at a $45°$ angle with respect to the crystallographic axis. Reflection and transmission measurements from the THz to the visible/UV range show that the THz phase velocity and visible group velocity match at a wavelength of 562 nm, which is required for strong EO signal.

INTRODUCTION

Accurate measurements of the temporal distribution of charge in an electron bunch are required for high energy light sources used in modern acclerators. Using the radiation emitted from the bunch allows shape measurements to be performed without affecting the bunch itself. In most cases, this radiation comes in the form of synchrotron radiation or transition radiation. Synchrotron radiation is emitted when the electron bunch undergoes centripetal acceleration, and is common in synchrotrons or storage rings. Transition radiation occurs when the bunch travels into a dielectric media, such as a sheet of conducting foil.

Some previous methods involve the use of a thermal detector to analyze the bunch radiation, but these detectors can often be unreliable, especially in the THz region. This is why the electro-optic (EO) effect is used to measure the bunch electromagnetic radiation. When the radiation spectrum is obtained using EO readout, the spectrum contains all the information on the bunch shape, including assymetry.

In this method, radiation in two different spectral regions from the same bunch are used. The coherent THz radiation causes a birefringence (change in the refractive index) in the EO crystal, due to the THz electric field redistributing the bond charges in the crystal. An incoherent visible pulse from the same bunch then samples this birefringence, changing the polarization of the electric field. This signal is detected using either a pair of cross-polarizers or a balanced diode detector in combination with a quarter-wave plate and Wollaston prism.

Studies have been done on analyzing the electro-optic properties of crystals such as Zinc Telluride or Gallium Phosphide [1]. This project looks to do a similar analysis for hexagonal Silicon Carbide (6H-SiC). First, the optimal angle between the THz k-vector and the 6H-SiC c-axis which maximizes EO signal must be found. This is done by running a simluation of the EO effect over every possible crystal orientation. Second, the matching condition between THz phase velocity and visible group velocity required for maximum signal is calculated, using reflection and transmission measurements from the THz to the visible/UV range.

OPTIMIZING CRYSTAL ORIENTATION

In this section, we work to find the incident angle of the THz radiation onto the EO crystal which gives optimum signal. First, a brief description of the theory behind the electro-optic effect is given. This is followed by a step-by-step explanation of the simulation, which was written in Wolfram Mathematica. The results and implications are then considered at the end of the section.

Theory

For electromagnetic radiation travelling in an anisotropic crystal medium, the modes of propagation can be found from the index ellipsoid. In its simplest form, the index ellipsoid is given by

$$\frac{u_1^2}{n_1^2} + \frac{u_2^2}{n_2^2} + \frac{u_3^2}{n_3^2} = 1 \tag{1}$$

where n_i is the refractive index in the ith direction and $\vec{u}_1$, $\vec{u}_2$, and $\vec{u}_3$ are the principal axes. The principal axes point in the directions where $\vec{E}$ and the displacement vector $\vec{D}$ are parallel inside the crystal. In these conditions where $\vec{E}$ and $\vec{D}$ are parallel, the relative dielectric permittivity ϵ is a symmetric rank-2 tensor. For a general anistropic crystal the relative dielectric permittivity then becomes

$$\boldsymbol{\epsilon} = \begin{bmatrix} \epsilon_1 & 0 & 0 \\ 0 & \epsilon_2 & 0 \\ 0 & 0 & \epsilon_3 \end{bmatrix} \tag{2}$$

We define the impermeability tensor η_{ij} to be the inverse of the relative permittivity tensor,

$$\boldsymbol{\eta} = \begin{bmatrix} \frac{1}{\epsilon_1} & 0 & 0 \\ 0 & \frac{1}{\epsilon_2} & 0 \\ 0 & 0 & \frac{1}{\epsilon_3} \end{bmatrix} \tag{3}$$

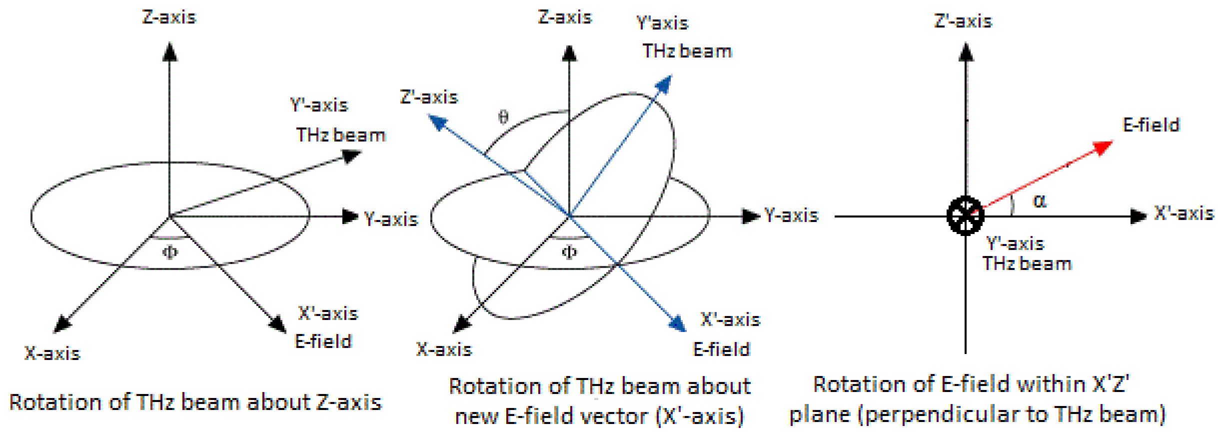

Figure 1: Diagram of the three rotation angles used in the simulation.

which is also a symmetric rank-2 tensor. Using the simple relation $n_i = \sqrt{\epsilon_i}$ for a nonmagnetic material, we rewrite the index ellipsoid in a condensed form:

$$\boldsymbol{u} \cdot \boldsymbol{\eta} \cdot \boldsymbol{u} = 1 \tag{4}$$

The eigenvectors of the impermeability tensor $\boldsymbol{\eta}$ are the same as the principal axes of the index ellipsoid, $\vec{u}_1$, $\vec{u}_2$, and $\vec{u}_3$. The corresponding eigenvalues λ_i are related to the principal indices of refraction by $\lambda_i = 1/\sqrt{n_i}$.

The electro-optic effect is traditionally represented as a change in the impermeability tensor due to the presence of an electric field $\vec{E}$. This change can be expressed in terms of the linear EO coefficients r_{ijk} and the quadratic EO coefficients s_{ijkl}. In scalar notation, the EO effect is given by

$$\eta_{ij}(\vec{E}) = \eta_{ij}(\mathbf{0}) + r_{ijk}E_k + s_{ijkl}E_kE_l \tag{5}$$

where a sum over indices k and l is implied. The linear coefficients r_{ijk} are also known as Pockels, and the quadratic coefficients s_{ijkl} are known as Kerrs. The quadratic EO effect is almost always negligible when the linear effect is nonzero. In our study of 6H-SiC, the linear effect is nonzero and the quadratic effect is neglected. For a more detailed analysis of the electro-optic effect, refer to [2].

It is the electric field of the coherent THz radiation that creates the birefringence in the EO crystal. If the electric field magnitude, electric field polarization, and linear EO coefficients are known, then Eq. 5 can be easily solved to obtain the impermeability tensor. The impermeability tensor then gives the principal refractive indices and the principal axes by solving the eigenvalue problem. The visible pulse with its own electric field then strikes the crystal at the same angle at the same time as the THz radiation. The electric field has been linearly polarized to a known direction before it hits the crystal. The plane containing the visible pulse electric field and the index ellipsoid intersect to make an ellipse with its own semi-major and semi-minor axes, which may or may not be equal to any of the principal axes. In either case, these semi-major and semi-minor axes are easily found, since the index ellipsoid and the angle of the plane is known. The two components of the visible pulse electric field along the semi-major and semi-minor axes undergo a relative phase shift Γ. This is directly proportional to the difference in the indices of refraction along the directions of the semi-major and semi-minor axes, which will hereon be referred to as n_2 and n_1.

$$\Gamma \propto (n_2 - n_1) \tag{6}$$

Note that n_1 and n_2 will likely *not* be the same as the princpal indices of refraction of the ellipsoid; it is important to remember that the axes of the ellipsoid and index ellipse will not always be the same. In the next subsection we consider how the simulation works to find the quantity $n_2 - n_1$.

The Simulation

The goal of this simulation is to find the angle between the THz k-vector and the 6H-SiC crystallographic axis which gives the greatest signal (particularly, the highest value of Γ). For 6H-SiC, this is not so straighforward. As we saw in the previous subsection, Γ is proportional to the quantity $n_2 - n_1$. However, 6H-SiC is anisotropic even without the presence of an electric field, so Γ is already nonzero. Instead, we must find the difference in Γ after applying the THz electric field.

The simulation uses something similar to Euler's Rotation Theorem to loop over every possible orientation of the THz electric field. Euler's Rotation Theorem says that any arbitrary rotation about a point in 3-space can be expressed in terms of three angles. These angles are illustrated in Figure 1. The crystallographic axis is aligned with the z-axis. Originally, the THz k-vector points in the +y-direction and the correspond electric field points in the +x-direction. There is a rotation about the +z-axis by an angle ϕ. Then the THz k-vector is rotated about the E-field by

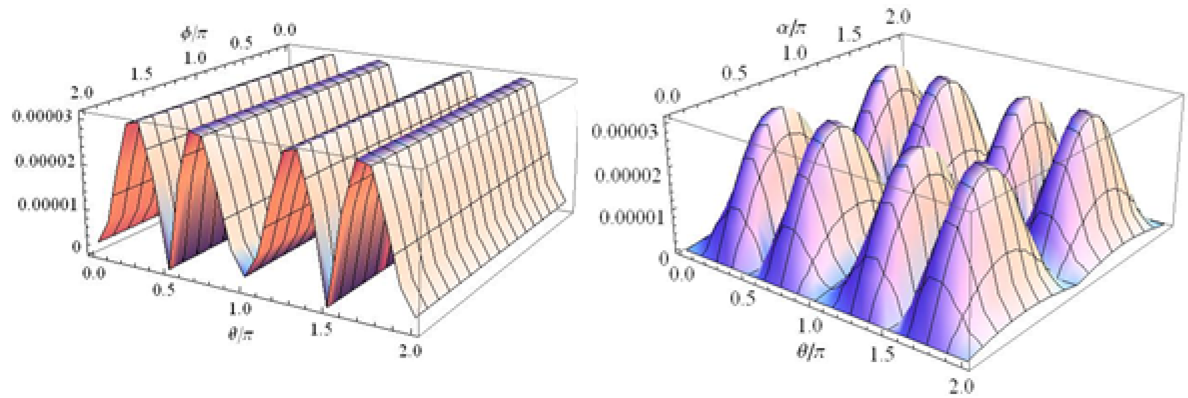

Figure 2: Three-dimensional plots of signal vs (ϕ, θ)(left) and signal vs (θ, α)(right).

an angle θ. Finally, the E-field is rotated about the THz k-vector by an angle α. The azimuthal rotation ϕ, polar rotation θ, and polarization angle α are the adjustable parameters of the loop.

Knowing the orientation of the electric field is not enough, however. From Eq. 5 we see that the linear electro-optic coefficiencts r_{ijk} must be known. The form of the r_{ijk} is known just from the symmetry of the crystal: $r_{113} = r_{223}$, $r_{232} = r_{131}$, and r_{333} are nonzero. Since indices i and j commute due to the symmetry of the r_{ijk} tensor, there are seven total nonzero values of this tensor, and only three independent values. Here we take the rough approximation that all seven nonzero values are equivalent. In order to lift this approximation, an experiment which finds the magnitude of the linear coefficients would need to be done. Since the crystal structure of 6H-SiC is not very different from that of 3C-SiC, we take these linear coefficients to be equal to 2.7 pm/V (see [3]).

For every combination of ϕ, θ, and α, the simulation goes through all the steps described in Theory. The three components of the THz electric field are expressed in terms of the three rotation angles, and with the r_{ijk} values, this gives the value of the impermeability tensor η_{ij}. The three eigenvalues λ_i of the tensor are used to find the principal refractive indices n_i from the relationship $\lambda_i = 1/\sqrt{n_i}$. These, and the eigenvectors $\vec{u}_1$, $\vec{u}_2$, and $\vec{u}_3$ are used to calculate the index ellipsoid (Eq. 1). The plane perpendicular to the incident THz radiation is easily calculated, and the plane/ellipsoid intersection gives the ellipse "seen" by the visible pulse electric field.

The difference in the lengths of the semi-major and semi-minor axes of this ellipse is equal to the quantity $n_2 - n_1$, which is the desired quantity in the simulation. Since the exact equation of the ellipse is known, this is easily calculated. First, this quantity is found for the case in which $\vec{E} = 0$. This is then subtracted from the value in the case where the THz field $\vec{E}$ is nonzero. This gives the final quantity $n_2 - n_1$, taking into account the anisotropy of the crystal.

Simulation Results

There are three independent parameters of the simulation: ϕ, θ, and α. The plots in Figure 2 show three-dimensional graphs of signal vs (ϕ, θ) and signal vs (θ, α). From these plots, there is a maximum value when $\alpha = \pi/2$ rad and $\theta = \pi/4$ rad. Note that there is no dependence on the azimuthal angle ϕ; only the polarization and the incident angle with the crystallographic axis affect the signal. This is not surprising, since 6H-SiC is uniaxial, and because of the rough assumptions made about the r_{ijk} tensor. With accurate measurements of r_{ijk} for this crystal, some azimuthal dependence may arise.

From these plots we see that when the electric field polarization angle α is zero, there is no signal. If EO experiments are run with 6H-SiC, the polarization of the THz electric field should be as close to $\alpha = \pi/2$ as possible.

The results of this simulation are true only for the THz radiation *inside* the EO crystal. In order to find what angles the THz radiation/electric field should satisfy before it strikes the crystal, we must consider refraction. To obtain the angle of maximum signal at $45°$ with the c-axis, the 6H-SiC sample should be cut at that angle. That way, the radiation would be at normal incidence and refraction would have no effect. However, if the sample is cut perpendicular to the c-axis, it is impossible to achieve $45°$ inside the crystal, since the effective refractive index of the crystal is 2.93 (see the results of the next section). In this case, the best choice for incident angle would be grazing incidence ($90°$), which gives a refractive angle inside the crystal of $19.9°$. Using $\alpha = \pi/2$ rad and $\theta = 19.9°$, the signal achieved would be 34% of the maximum signal at $\theta = 45°$. Another option in this case is the Brewster angle: for a crystal refractive index of 2.93, this angle would be near $71°$. This gives a refractive angle inside the crystal of $18.8°$, which results in a signal 33% of the maximum signal. This is illustrated in Figure 3 (next page).

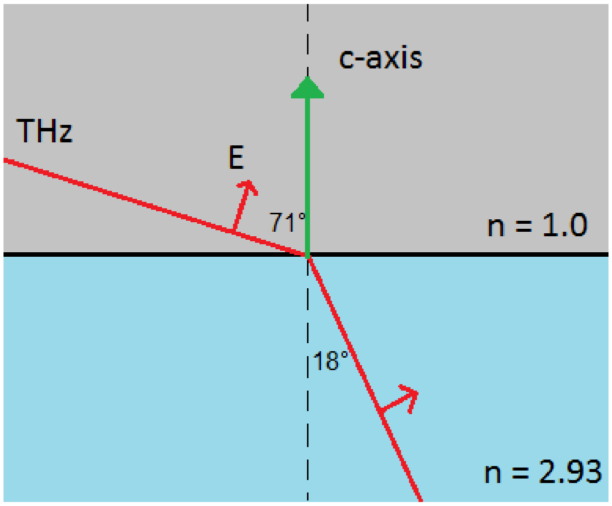

Figure 3: THz radiation incident at Brewster's angle. Refracted angle leads to a signal of 33% of the maximum possible signal.

PHASE VELOCITY / GROUP VELOCITY STUDY

In order to get maximum response with the EO effect, the co-propagating visible pulse and THz radiation must travel very near the same speed through the crystal. If the visible pulse propagates well with one frequency of the THz radiation, there will be a high response at that THz frequency, which is desirable in EO experiments. The goal is to have a high response over a large range of frequency, so that most of the THz spectrum from the electron bunch is obtained. This requires a match of the visible group velocity and the THz phase velocity in the crystal. To find the matching condition, the refractive indices in both the THz and the visible/UV region are needed. The following two subsections describe how the refractive indices in both regions were obtained, the third subsection discusses the results.

Reflectivity measurements in THz region

In the THz region, we performed reflectivity measurements of a 6H-SiC sample using a Bomem spectrometer. Various light sources and detectors were needed to get a large spectral range in the THz region: a thermal pyroelectric array detector was used with a globar thermal light source to cover the 550 - 5000 cm^{-1} range, and a helium-cooled bolometer with both a mercury thermal light source and the globar was used to cover the 10 - 550 cm^{-1} range.

The reflectivity of 6H-SiC is plotted in Figure 4. The large jump at 24.2 THz is due to transverse optical lattice oscillations in the crystal. All EO experiments done with these crystals must be done in the range below this TO oscillation. This is why 6H-SiC is a favorable candidate: previously studied EO crystals have these TO oscillations at much lower frequencies (5 THz for ZnTe and 11 THz for GaP [1]). The Drude-Lorentz model for dispersion in dielectrics is used to convert reflectivity measurements into a

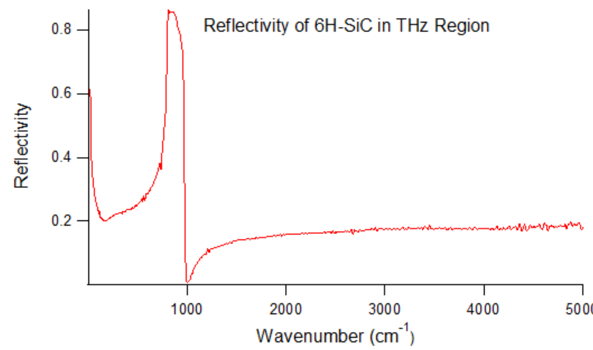

Figure 4: Reflectivity spectrum of hexagonal Silicon Carbide in Terahertz region. The large peak corresponds to a TO lattice oscillation at 805 cm^{-1}, or 24.2 THz.

model of the refractive index in the THz region. A model is fit to the reflectivity measurements, and from that model the dielectric function over that same frequency range is obtained. From the dielectric function model it is trivial to find the index of refraction.

The rapid increase in the reflectivity near zero is due to free charge carriers in the sample. This conductivity term is easily corrected for in the Drude-Lorentz model. In the DL model, the dielectric function is a sum of terms where each term represents a damped harmonic oscillator. One term comes from the oscillator at 24.2 THz (the peak in the reflectivity), and when the sample has conductivity, a term is added for an oscillator at 0 THz. To ignore the conductivity in the sample, this term is simply removed from the model. Another option is to cool the sample to a temperature where the free charge carriers are no longer active, and redo the reflectivity measurements.

Transmission measurements in visible/UV region

In the visible/UV region, transmission measurements were taken of the same 6H-SiC sample with the same spectrometer. Light from a tungsten lamp transmitted through the sample onto a silicon photodiode. A color filter increased the signal-to-noise ratio; this resulted in a range of usable data from 15000 - 24000 cm^{-1}. Figure 5 (next page) shows the entire range of transmission data as well as a plot in a small subrange from 19000 - 19500 cm^{-1}.

The oscillations in the transmission spectrum demonstrate thin film interference. This intereference depends on the refractive index of the crystal. This means the interference spectrum gives the refractive index dispersion in the same range. This is done by fitting a sine wave at every small range of wavenumbers and finding the frequency of oscillations (a Fourier transform could work just as well). The frequency of oscillations is related to the sample thickness and refractive index by simple thin film interference relations.

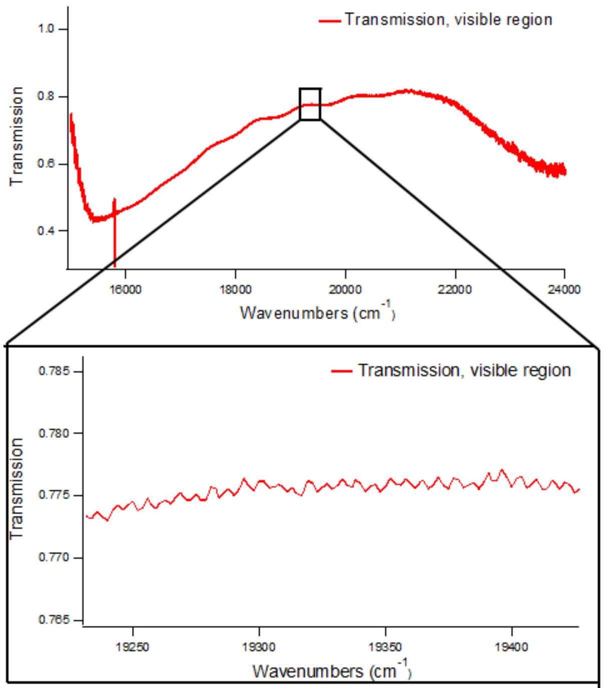

Figure 5: Transmission spectrum of hexagonal Silicon Carbide in visible/UV region.

Finding the matching condition

The previous two subsections describe how the refractive index in both the THz region and the visible/UV region is obtained. Figure 6 shows the results plotted on the same graph. The asymptotic value of the refractive index in the THz region (below the TO lattice oscillation) occurs at a value of 2.93, and is extended to the visible/UV range, and the intersection is found. This intersection occurs at 17773 cm^{-1}, or about 562 nm wavelength.

When the visible pulse of radiation is at this wavelength, the pulse will travel at nearly the same (group) velocity as the THz radiation phase velocity. It cannot travel at exactly the same velocity: when the matching condition was found, the asymptotic value of the refractive index in the THz region was used. From Figure 6, we see that even below the TO lattice oscillation, the refractive index slowly rises. Choosing the asymptotic value as the refractive index matching condition is still a fair approximation for the usable THz range, though it will not exactly represent the true refractive index for the entire range.

EO Response Function

The electro-optic response function *G(f)* is a way of measuring the EO signal, based on how well the THz phase velocity and visible group velocity match. Here we use the same definition used in [1]: $G(f)$ is the integral of the co-propagating visible pulse and THz wave at one frequency.

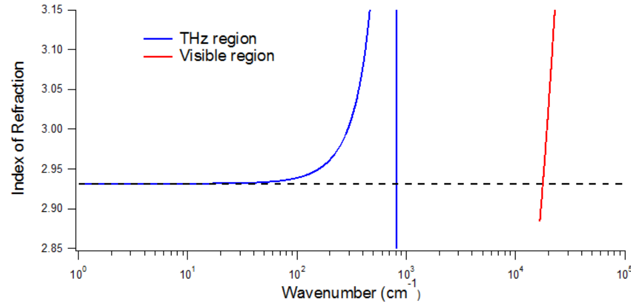

Figure 6: Results of transmission/reflectivity measurements. Index of refraction in both regions. Matching condition occurs where the asymptotic value of the THz region (n = 2.93) intersects the visible region.

$$G(f) = T * \frac{1}{d}\int_0^d \int_{-\infty}^{\infty} \exp\left[i(kz - 2\pi f t)\right]\delta(z - v_g t)\, dt\, dz \tag{7}$$

$$= T * \frac{1}{d}\int_0^d \exp\left[i2\pi f z\left(\frac{1}{v_{ph}(f)} - \frac{1}{v_g}\right)\right] dz \tag{8}$$

Here *d* is the thickness of the sample, $v_{ph}(f)$ is the phase velocity at THz frequency *f*, v_g is the group velocity at the chosen visible wavelength, and *T* is the transmission coefficient:

$$T = \frac{2}{1 + n(f) + i\kappa(f)} \tag{9}$$

where $n(f)$ and $\kappa(f)$ are the real and imaginary parts of the refractive index, respectively. If the visible pulse and THz wave at frequency f co-propagate at the same speed, $G(f)$ will have a high value (as high as the transmission coefficient T) for all frequencies lower than the TO lattice oscillation. At the TO lattice oscillation, $G(f)$ will always drop to near zero, since the bond charges in the crystal are rearranging much too quickly for any effective propagation.

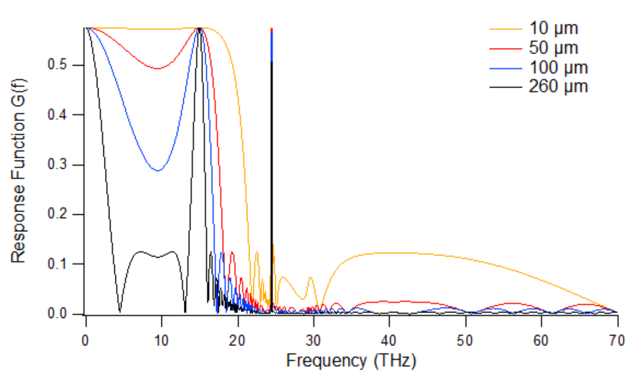

Figure 7: EO response function for 6H-SiC for various sample thicknesses.

The response function for 6H-SiC is shown in Figure 7. The group velocity at a visible wavelength of 562 nm is used in the calculation, with varying sample thickness. Our sample (260 microns) is too thick for effective co-propagation at all usable THz frequencies. However, Figure 7 shows that as the sample thickness decreases, the response function $G(f)$ approaches maximum value for usable THz frequencies.

CONCLUSIONS

The change in the refractive index caused by the EO effect depends on the crystal properties (the r_{ijk} tensor) and the orientation of the electric field. A computer simulation looped through every possible orientation of the THz k-vector and electric field, and with some rough assumptions on the r_{ijk} tensor, found that the optimal orientation was with the THz k-vector at a $45°$ angle with the c-axis and the electric field oriented at $\alpha = \pi/2$ rad.

The EO response is measured by the response function $G(f)$. When $G(f)$ is large, the visible pulse effectively co-propagates with the THz wave at frequency f. Reflection measurements in the THz region and transmission measurements in the visible/UV region give us the refractive indices in both ranges. These refractive indices match at a visible pulse wavelength of 562 nm.

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REFERENCES

[1] S. Casalbuoni et al. Numerical Studies on the Electro-Optic Sampling of Relativistic Electron Bunches. *Particle Accelerator Conference*, May 2005.

[2] A. Yariv and P. Yeh. Optical Waves in Crystals: Propagation and Control of Laser Radiation. (John Wiley & Sons, 1984) p. 220.

[3] X. Tang et al. Linear electro-optic effect in cubic silicon carbide *Applied Physics Letters 59*, 0003-6951/91/411938-02, 1991.