Terahertz Kerr and Reflectivity Measurements on the Topological Insulator Bi$_2$Se$_3$

G. S. Jenkins,$^1$ A. B. Sushkov,$^1$ D. C. Schmadel,$^1$ N. P. Butch,$^1$ P. Syers,$^1$ J. Paglione,$^1$ and H. D. Drew$^1$

$^1$Center for Nanophysics and Advanced Materials
Department of Physics, University of Maryland, College Park, Maryland 20742, USA
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We report the first terahertz Kerr measurements on bulk crystals of the topological insulator Bi$_2$Se$_3$. At $T=10$ K and fields up to 8 T, the real and imaginary Kerr angle and reflectance measurements utilizing both linearly and circularly polarized incident radiation were measured at a frequency of 5.24 meV. A single free carrier bulk response can not describe the line-shape. Surface states with a small mass and surprisingly large associated spectral weight quantitatively fit all data.

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I. INTRODUCTION

The discovery$^{1–8}$ and realization of 3D strong topological insulators (TIs)$^{9,10}$ have generated much recent interest. A strong spin-orbit coupling can produce a topologically protected metallic surface state which has a Dirac-like dispersion with a non-degenerate chiral spin state that can not be destroyed or gapped by (non-magnetic) impurity scattering. Since the spin direction of the quasiparticle is locked to the momentum direction$^{10}$ suppression of Anderson localization results from the destructive interference associated with the quasiparticle’s Berry phase upon backscattering.$^{11}$ With the action of local ordered magnetic moments, the surface state becomes gapped$^{7}$ In zero magnetic field with the Fermi energy within the gap, a quantized Kerr angle associated with the half quantized Hall conductance is a predicted signature of the topological origin of the surface state$^{10}$.

These and other predicted properties have stimulated a recent flurry of activity to probe the exotic surface state. Although the surface states have been observed by angular resolved photo-emission spectroscopy (ARPES)$^{9,12}$ unambiguous separation of bulk from surface state conduction has plagued transport characterization of the surface state due to the rather large intrinsic charge density in the bulk conduction band in as-grown Bi$_2$Te$_3$ and Bi$_2$Se$_3$. Various schemes to circumvent this difficulty in bulk crystals have either implemented counter doping during growth$^{14}$ minimized Se vacancies$^{15}$ or fine tuned annealing processes$^{14}$ One intermediate goal has been to sufficiently deplete the bulk to allow gating or chemical doping of the surface in suitably thin crystals to move the chemical potential of the bulk into the gap$^{16}$.

There are several advantages of terahertz Kerr measurements over other transport measurements which allow separation of the surface from the bulk response. The surface states are expected to have a a different mass than the bulk, depending upon the location of the chemical potential, giving rise to two different resonant cyclotron frequencies. This effect can be further exaggerated by application of an electric field either from gating or chemically doping the surface causing ‘band bending’ effects, pushing the Dirac point closer to the chemical potential.

Secondly, the line-shape as a function of magnetic field associated with a surface state is qualitatively different than the bulk response. Thirdly, no electrical contacts are required. Lastly, Kerr measurements on thick crystals allow probing of a single surface state.

In this article, Kerr measurements are reported on two bulk crystals of Bi$_2$Se$_3$ at an optical frequency of $5.24$ meV in fields up to 8 T. Line-fits of the data show evidence of surface states. For a measured surface state cyclotron mass of $m_{cs} = 0.033 m_e$, the associated spectral weight is about a factor of 20 times larger than expected based upon the corresponding number density estimated from ARPES data. Measurements of other samples from the same as-grown crystal show a higher carrier concentration. Alternatively, a simple model of the optical response which assumes the presence of two crystallites with different carrier concentrations can qualitatively describe all of our data. Future work to optically characterize carrier density homogeneity as well as an analysis of surface state signals expected by gating are discussed.

II. EXPERIMENTAL DESCRIPTION

A. Kerr System Description and Calibration

For calibration purposes, the Faraday rotation and ellipticity induced by a sample in magnetic field was measured utilizing a polarization modulation technique with a heterodyne detection scheme, a technique detailed elsewhere.$^{12}$ The complex Faraday angle and relative transmission were concurrently measured.

The instrument was modified allowing simultaneous measurement of the complex Kerr angle and relative reflection by the insertion of a beam splitter, polarizer, and collection optics which include an off-axis parabolic aluminum mirror and a 2 K silicon composite bolometer as depicted in Fig. 1. The Kerr and Faraday angle are measured separately.

Assuming a rotationally invariant sample, the complex
Kerr and Faraday angle data shown in Fig. 2(a-d) are simultaneously fit where the mobility $\mu$, cyclotron mass $m$, 2D number density $n$, and sheet resistance of the AR coating $R_\square$ are free parameters. The B-field value of the resonance determines the mass $m$, the width of the resonance determines the number density $n$, and deviations of the AR coating sheet resistance $R_\square$ away from the ideal value tends to mix the real and imaginary parts of the angles giving rise to anisotropies in the resonant peaks.

The fit parameters are found to be the following: $n=2.0 \times 10^{11}$ cm$^{-2}$, $\mu = -1.19 \times 10^5$ cm$^2$/V sec where the negative sign denotes the sign of the carriers, $m=0.0695 m_e$, and $R=1.01 \times R_{AR}$ where $m_e$ is the mass of an electron and $R_{AR}$ is the sheet resistance for the perfect AR coating condition of GaAs given by $Z_0/(n_{GaAs} - 1) = 145\ \Omega/\square$ where $Z_0 = 377\ \Omega/\square$ is the impedance of free space and $n_{GaAs} = 3.6$ is the index of refraction for GaAs. The mass is consistent with the known mass of GaAs$^{18}$ and the antireflection coating is very close to ideal. The fits are overlayed on top of the reflection and transmission data shown in Fig. 2(e,f). Essentially, all 6 curves are very well reproduced with only two parameters, $n$ and $\mu$. Together with the polarizer calibration, the 2-DEG GaAs calibration indicates that both the Kerr and Faraday experiments perform exceptionally well.

This particular data set also illustrates the robustness of the Kerr system. Instability associated with the two-laser system occurred between +8 and $\sim+5.5$ T appearing as a loss of power and commensurate increase of relative detector noise in the Kerr and reflection data sets. Although the reflection data was visibly affected, the Kerr signal was essentially unaffected. The reflection measurement is derived by the ratio of the signals from two detectors (‘Kerr detector’ and ‘reference detector’ in Fig. 1) and therefore susceptible to large spacial mode fluctuations, but the Kerr signal is derived from the modulated signal measured by a single detector.

### B. Bi$_2$Se$_3$ sample description

Bulk crystals of Bi$_2$Se$_3$ were grown by varying the heating conditions of the melt and stoichiometric ratio of bismuth and selenium$^{18}$. Surfaces were cleaved perpendicular to the c-axis, and the back sides were cut and ground yielding a total bulk thickness of about 0.5 cm. Samples were mounted such that the applied magnetic field was parallel to the c-axis and the radiation propagation direction. The effective area measured was defined by the optical spot size $\sim g1.0$ mm. During a time span of about 10 seconds, the surfaces of the crystals were cleaved using scotch tape in air and immediately inserted into a vacuum of $\sim 10^{-7}$ Torr.
The Kerr angle and concurrent reflection measurements of linearly polarized light are shown in Fig. 3(a-c) and (e-g) for two samples both measured at 10 K in fields up to 8 T. A separate measurement was performed on Sample 1 where the waveplate in the rotator depicted in Fig. 1 was statically set producing elliptically (nearly circularly) polarized light. The reflection was then measured and normalized to the B=0 T value. The data is reported in Fig. 3(d). $\beta$ characterizes the degree of mixing between left and right circular polarization states via $(1-\beta)R_+ + \beta R_-$. where $R_+$ is the reflection produced by purely circularly polarized light. In this measurement, $\beta$ is found to be 0.13.

To analyze the results, a model is constructed where the bulk is considered to be a thick slab whose optical response is characterized by a dielectric function consisting of a Drude carrier response with an associated unscreened bulk plasma frequency $\omega_{pb}$, bulk cyclotron mass $m_{cb}$, and bulk scattering rate $\gamma_b$ along with the addition of two TO phonon modes whose parameters have been previously determined. The surface state resides at the interface whose admittance is modelled within a Drude model with an associated 2-D surface plasma frequency $\omega_{ps}$, surface cyclotron mass $m_{cs}$, and surface scattering rate $\gamma_s$. Details of the model are discussed in Sec. A.3.

FIG. 2: (color online) Calibration of the Faraday and Kerr angle measurement system. The Faraday angle, Kerr angle, and relative transmission and reflection from a GaAs 2-DEG heterostructure were measured at an optical frequency of 5.24 meV and temperature of 80 K as a function of magnetic field. The Faraday and Kerr angles are simultaneously fit with effectively two parameters, the number density and mobility, found to be $n=2.0 \times 10^{11}$ cm$^{-2}$ and $\mu = -1.19 \times 10^9$ cm$^2$/V sec. The fits (red line) are overlayed on top of the data (black dots) for comparison. The same two parameters are used to calculate reflection and transmission (normalized to the B=0 value) and overlayed on the data.

III. KERR AND REFLECTION MEASUREMENTS ON Bi$_2$Se$_3$ AND MODEL DESCRIPTION

The Kerr angle and concurrent reflection measurements of linearly polarized light are shown in Fig. 3(a-c) and (e-g) for two samples both measured at 10 K in fields up to 8 T. A separate measurement was performed on Sample 1 where the waveplate in the rotator depicted in Fig. 1 was statically set producing elliptically (nearly circularly) polarized light. The reflection was then measured and normalized to the B=0 T value. The data is reported in Fig. 3(d). $\beta$ characterizes the degree of mixing between left and right circular polarization states via $(1-\beta)R_+ + \beta R_-$. where $R_+$ is the reflection produced by purely circularly polarized light. In this measurement, $\beta$ is found to be 0.13.

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IV. DISCUSSION

A. Analysis of the optical response

Sample 1 data shown in Fig. 3(a-d) can not be quantitatively nor qualitatively reproduced by a homogenous bulk response with any arbitrary carrier concentration, cyclotron mass, or scattering rate. The blue dash-dot curves in Fig. 3(a-d) show a representative example of line-shapes expected from a simple bulk response, calculated with the specific parameters $\omega_{pb} = 420$ cm$^{-1}$ (corresponding to a number density of $n_b \sim 3.2 \times 10^{17}$ cm$^{-3}$), $\gamma_b = 12$ cm$^{-1}$, and $m_{cb} = 0.135 m_e$. Although the high field Kerr data in Fig. 3(a,b) is well described by this particular set of parameters, strong qualitative deviations exist at fields below $|B| \lesssim 4$ T suggesting that another carrier response with a much smaller cyclotron mass may be superimposed. The reflection data in Fig. 3(c,d) also show very different behavior at low fields $|B| \lesssim 4$ T compared to a bulk response. The small humps which appear at fields around $\sim \pm 1$ T in Fig. 3(e) and $\sim -1$ T in
Fig. 3: (color online) Kerr angle and reflection of linearly polarized light normalized to the B=0 T value were measured at 10 K and an optical frequency of 5.24 meV. Reflection of nearly circularly polarized light normalized to the B=0 T value was a separate measurement performed only on Sample 1. (a-d) Sample 1 data (black dots) is depicted with several different fits superimposed. A homogeneous bulk response cannot describe the data (blue dash-dot). A homogeneous bulk response with surface states (red solid) fit the data. The weighted optical response of two crystallites with different carrier density (green dash-dot) can reproduce the salient features of the data. Slightly elliptically polarized light, characterized by parameter $\beta=0.13$, is used in calculations for Fig. 3(d). (e-h) Sample 2 data (black dots) show a very different response but was only 2 mm away from Sample 1 in the as-grown crystal. A homogeneous bulk response (blue dash-dot) does not quite describe the data. Addition of a surface state response does not improve the fits. A weighted response of two crystallites with different carrier density (green dash-dot) more closely resembles the data. No reflectivity data using circular polarized light was taken for Sample 2. $\beta$ is assumed zero in calculations in Fig. 3(h).

Fig. 3(d) can not be qualitatively reproduced with any value of bulk carrier concentration, cyclotron mass, or scattering rate.

The addition of a surface state with parameters $\omega_{ps} = 41 \text{ cm}^{-1}$, $\gamma_s = 28 \text{ cm}^{-1}$, and $m_{cs} = 0.033 m_r$ results in the red solid curves in Fig. 3(a-d) which fit all the data remarkably well. The surface state cyclotron mass corresponds to a resonance at B=1.5 T (for a frequency of 5.24 meV). The scattering rate associated with the surface is substantially larger than that of the bulk, a result which might be expected based upon Shubnikov de Haas measurements on similarly grown samples and linewidths estimated from ARPES data. Reasonable linefits may be achieved with a surface state plasma frequency as low as $\omega_{ps} = 20 \text{ cm}^{-1}$ although deviations from the data begin to become more pronounced.

From the ARPES measured dispersion of the surface states (see Eq. A.1) the cyclotorn mass given by $h k_f / v_f$ and number density given by $\pi k_f^2 / (2\pi)^2$ may be parameterized in terms of the Fermi energy. A mass of 0.033$m_c$ corresponds to a Fermi energy of $\sim 20 \text{ meV}$ above the Dirac point, a number density of $10^{11} \text{ cm}^{-2}$, and corresponding surface plasma frequency of 1.3 cm$^{-1}$. The bulk plasma frequency corresponds to a Fermi energy of approximately 20 meV above the bulk conduction band edge which is itself located $\sim 200 \text{ meV}$ above the Dirac point.

ARPES observations correspond to a large occupation number of the surface states, $n_s \sim 10^{12} \text{ cm}^{-2}$. This amount of charge acting on the bulk carriers would deplete the free carriers to a depth $d = n_s / n_b \sim 100 \text{ nm}$ (where $n_b$ is the bulk number density). Since ARPES concurrently observed the bulk bands but the measurement technique is sensitive to depths of only $\sim 1 \text{ nm}$, it must be assumed that there is a fixed neutralizing positive surface charge coming from excess Se$^+$ or adsorbed ions, or possibly an intrinsic effect. Therefore, band bending effects are apparently small in vacuum cleaved crystals. However, for samples exposed to ambient conditions, band bending may be more severe possibly due to oxidation of the surface. This chemical doping moves the Dirac point toward the chemical potential and leads to a depletion region in the bulk, presumably with $d < n_s / n_b$.

Since the optical penetration depth is much larger than 100 nm at terahertz frequencies, an insulating depletion layer has no significant optical response and the exper-
ment sees the surface carriers only as a modification to the bulk.

More significantly, the surface plasma frequency obtained from the fits to the optical response are about a factor of 15 to 30 larger than predicted from conventional transport theory based on the ARPES measured surface state dispersion relation. A possible explanation is that the crystal has multiple surface layers within the optical penetration depth, perhaps separated by interstitial selenium, dividing the crystal into $\sim 10$ thin flakes which are optically very close but electronically decoupled. The cleave process may favor the region with layers of flakes where these interstitial selenium may be more prevalent offering a weaker link between planes.

Given the unconventional spin-chiral nature of the eigenstates, which imply spin currents perpendicular to the charge currents, an unconventional matrix element for Landau level transitions may also lead to enhancement of the spectral weight, a possibility that needs further investigation.

As a possible alternative explanation, the structure observed at low field in the optical response of Sample 1 may arise from spatial variation of the bulk charge carrier concentration. Consider the possibility that the optical spot covers a region where two crystallites of different carrier concentrations exist. Modelling this scenario, a simple weighted average of two individual bulk responses without surface states is performed with the following parameters: one bulk response weighted by 0.84 with parameters $\omega_{pb1} = 334 \text{ cm}^{-1}$, $\gamma_{pb1} = 6.5 \text{ cm}^{-1}$, and $m_{pb1} = 0.126$ with a second bulk response weighted by (1-0.84) with parameters $\omega_{pb2} = 548 \text{ cm}^{-1}$, $\gamma_{pb2} = 8.2 \text{ cm}^{-1}$, $m_{pb2} = 0.148 m_e$. The green dash-dot curves in Fig. 3(a-d) show all of the salient features of the data which was attributed to the surface states. The difference between the two bulk masses is unexpected for a simple parabolic band near a gapped band edge. The coarse qualitative features of the data may be reproduced by choosing the bulk masses to be the same with the other parameters chosen similarly although the fits are substantially worse. Therefore, even though the fit is not very good quantitatively, it is possible that a distribution of carrier concentrations may be the culprit giving rise to the observed line-shapes.

Perhaps lending more credence to the spatial variation of carrier concentration argument is that a completely different response was measured by a separate sample which was only 2 mm away from Sample 1 in the $d \sim 1.5 \text{ cm}$ as-grown crystal. The measurement results of Sample 2 are shown in Fig. 3(e-g). A single bulk response qualitatively describes all the features of the data, as depicted by the blue dash-dot curves, whose parameters are $\omega_{pb} = 583 \text{ cm}^{-1}$, $\gamma_b = 7 \text{ cm}^{-1}$, and $m_{pb} = 0.145 m_e$. $\beta$ is assumed zero in calculations in Fig. 3(b). This is a much higher bulk plasma frequency than found for Sample 1. Addition of a surface state response does not improve the fits. However, modelling the bulk carrier concentration variations as done for Sample 1 where one bulk response is weighted by 0.70 with parameters $\omega_{pb1} = 583 \text{ cm}^{-1}$, $\gamma_{b1} = 5.3 \text{ cm}^{-1}$, and $m_{b1} = 0.145 m_e$ and a second bulk response weighted by (1-0.70) with parameters $\omega_{pb2} = 250 \text{ cm}^{-1}$, $\gamma_{b2} = 15 \text{ cm}^{-1}$, $m_{b2} = 0.145 m_e$ yields a good quantitative fit to all three data sets as shown by the green dash-dot curves in Fig 3(e-g). A third sample measured 2 mm away from Sample 2 (and 4 mm away from Sample 1) yielded the same response as sample 2.

Controlling carrier concentrations to well within a factor of $\sim 10$ over cm-sized crystals is very challenging. Using the Kerr and reflectivity measurements, it is not possible to determine whether such a response originates from a distribution of carrier concentration in a single crystallite, or the presence of two (or more) crystallites with different concentrations.

B. Other methods to characterize surface states

Using the Kerr measurement technique as a routine diagnostic of sample carrier concentration variation is not feasible since it is resource intensive. Since inhomogeneity is an important general issue, a routine diagnostic tool is required. One method presently being explored utilizes FTIR spectroscopic reflection measurements. As representatively shown in the reflectivity spectral measurements in figure 4 of Ref. 19, a large dip occurs at the plasma edge. The frequency location of the dip gives information regarding the carrier concentration. Measurements on different parts of a sample utilizing a small spot size $\sim 0.5 \text{ mm}$ in the infrared spectral region is currently being investigated as a diagnostic tool to quantify the degree of carrier concentration variation.

Another method that may be favorable for observing surface states presently being developed utilizes a gate. If the surface states have a cyclotron mass and optical spectral weight commensurate with the ARPES measured dispersion, the line-fitting procedure as used for the above Kerr and reflection data for discerning the surface state will most likely be extremely difficult with the present signal-to-noise of the system, even for fairly small surface scattering rates (see Sec. [B1] for details).

By gating, the surface state optical response can be modulated with negligible effects produced by the bulk. Any modulated signal coming from the bulk would necessarily be due to the screening layer after the depletion layer which is of thickness $\sim 1/k_s$, the Thomas-Fermi screening length. $k_s \sim 15 \text{ nm}$ at $n_b = 10^{17} \text{ cm}^{-3}$, a typical carrier concentration. The charge in this layer $n_d \sim n_b/k_s \sim 10^{11} \text{ cm}^{-2}$ which is much less than the surface state carrier density. More important, however, is that $k_s$ does not depend on gate voltage and therefore is not modulated. Secondly, by modulating a gate, signal-to-noise is expected to increase by over an order of magnitude. Averaging between gate-on and gate-off configurations in fixed field on relatively short time scales eliminates magnetic field drift issues and minimizes time
drift issues. Thirdly, due to this large gain in sensitivity, even very high surface scattering rates should be detectable. Expected signals produced by gating, modelled in Sec. [2.2] are expected to be well within experimental capability. Fourthly, the expected Kerr response from the surface states for bulk plasma frequencies ranging from 200 to 600 cm⁻¹ have qualitatively the same line-shapes. Even with possible carrier inhomogeneities, the detection of surface states is possible by modulating a gate with modest shifts of the surface state Fermi energy. However, characterization of the surface states will require reasonably homogeneous carrier concentrations. Lastly, developing the technique is a necessary stepping stone for future experiments: measurement of the half-quantized Hall conductivity in topological insulators inside the gap created by a magnetic layer.

V. CONCLUSION

Kerr measurements using heterodyne detection in the terahertz regime are reported. Measurements of a 2-DEG GaAs heterostructure prove the system works exceptionally well. Results on Bi₂Se₃ crystals show that the technique is sensitive to bulk carrier concentration. For one particular sample, the Kerr and reflectance data are very well described by including surface states. The measured surface spectral weight ωₚₜ (with an associated surface cyclotron mass mₚₜ = 0.025 mₜ) is ~20 times larger than expected for a single surface state based upon ARPES measurements. Spatial variation of carrier concentration can also qualitatively account for the data. Separation of bulk and surface states may be accomplished in the Kerr geometry with greater sensitivity by incorporating a gate, a proposed next step to clearly separate the bulk and surface state effects.

VI. ACKNOWLEDGEMENT

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Appendix A: Multilayer response analysis

1. General Notation

The formalism for deriving the Fresnel reflection and transmission amplitudes for a system consisting of multiple layers of ‘dielectric’ slabs with thin films at the interfaces are derived for a general number of layers. The model will be applied to two specific cases: a GaAs 2-DEG heterostructure with a metallic antireflection coating and a bulk Bi₂Se₃ crystal with a surface state. Using Maxwell’s equations for radiation at normal incidence, boundary conditions on the electric and magnetic field may be applied across any boundary between two dielectric slabs i and i + 1 of thicknesses dbᵢ and dbᵢ₊₁ with bulk complex indices nᵢ and nᵢ₊₁ and a metallic thin film at the interface with a sheet conductivity of σᵢ dfᵢ, where dfᵢ is the thickness of the film. The vector notation (Eᵢ⁺,Eᵢ⁻) represents right and left travelling waves. The electric field may be expressed in either a linear polarization basis notated as (Eᵢ₊ₓ,Eᵢ₊ₓ) and (Eᵢ₋ₓ,Eᵢ₋ₓ) or in a circular polarization basis as (Eᵢ₊ₓ,Eᵢ₋ₓ) and (Eᵢ₋ₓ,Eᵢ₊ₓ). To derive the equations generally, we use the notation (Eᵢ⁺,Eᵢ⁻) to represent one of the polarization basis states y, x, +, or -.

The matrix Mᵢ,α which transfers an arbitrary E-field (Eᵢ⁺,Eᵢ⁻) across an interface consistent with Maxwell’s equations is given by the following:

\[
Mᵢ,α = \frac{1}{2} \sum_{i+1,α} \left( 
\frac{nᵢ,α + nᵢ₊₁,α - yᵢ,α}{2} \right) 
\frac{nᵢ,α + nᵢ₊₁,α + yᵢ,α}{2} 
\left( 
\begin{array}{cc}
-1/nᵢ,α & dbᵢ/c \\
nᵢ,α & dbᵢ/c 
\end{array}
\right)
\]

where the admittance of the thin film is yᵢ,α = Z₀σᵢ,α dfᵢ and Z₀ is the impedance of free space.

The matrix Kᵢ,α transfers an arbitrary oscillating E-field (Eᵢ⁺,Eᵢ⁻) at frequency ω across a dielectric of thickness dbᵢ and complex index nᵢ,α is given by the following:

\[
Kᵢ,α = \left( 
\begin{array}{cc}
e^{i nᵢ,α dbᵢ/c} & 0 \\
0 & e^{-i nᵢ,α dbᵢ/c}
\end{array}
\right)
\]

In general, an incident right travelling wave will be assumed incident on the first interface with a resulting net reflected travelling wave to the left from the first interface and a net right travelling transmitted wave from the last interface. The Fresnel transmission and reflection amplitudes for a stack of dielectrics of m interfaces with thin metallic films are then found by the relation:

\[
\left( 
\begin{array}{c}
t₀ \\
0
\end{array}
\right) = M_{m,α} \prod_{i=1}^{m-1} K_{i+1,α} M_{i,α} \left( 
\begin{array}{c}
1 \\
r₀
\end{array}
\right)
\]

2. GaAs 2-DEG

The above formalism can be applied to calculate the Kerr and Faraday angle as well as the reflection and transmission for the calibration sample. A GaAs 2-DEG is modelled as a thin metallic sheet whose sheet conductance is described within a simple Drude model located at the interface between vacuum and a GaAs dielectric slab. The backside of the sample consists of an interface between a GaAs dielectric slab and vacuum with an antireflection coating, a thin metallic NiCr film, modelled within a Drude model. The antireflection coating is fully characterized by the dc sheet resistance, \( R_⟂ = 1/(σ df) \), since the scattering rate of NiCr is very large compared.
The reflectance (normalized to zero field) in a pure circular or linear state are calculated. Each row of graphs represents a different set of Drude parameters describing the bulk and surface state response. The second and third row graphs, \((e-h)\) and \((i-l)\), are offset for clarity such that the Kerr angle is zero and \(R/R[0T]\) is unity in zero magnetic field. The graphs with the Fermi energy \(E_F = 0\) correspond to the Dirac point where there is no contribution from the surface state, only bulk response. All graphs are calculated with a bulk mass \(m_c = 0.15 m_e\) and a bulk scattering rate \(\gamma_b = 10 \text{ cm}^{-1}\) and using the bulk phonon parameters as measured in Ref.13. (a-d) Very low bulk density to demonstrate the response expected from the surface states: \(\omega_{pb} = 10 \text{ cm}^{-1}, \gamma_b = 10 \text{ cm}^{-1}\). (e-h) \(\omega_{ph} = 400 \text{ cm}^{-1}, \gamma_s = 3 \text{ cm}^{-1}\) (i-l) \(\omega_{pb} = 400 \text{ cm}^{-1}, \gamma_s = 10 \text{ cm}^{-1}\). (m-p) \(\omega_{ph} = 400 \text{ cm}^{-1}, \gamma_s = 25 \text{ cm}^{-1}\).

to the optical frequency. The circular polarization basis is easiest to work with in applied magnetic field.

The parameters are as follows: \(n_{1, \pm} = 1, n_{2, \pm} = 3.6, n_{3, \pm} = 1, \omega = 5.24 \text{ meV}, (Z_0 \sigma d f)_{1, \pm} = \omega_{ps1}/(\gamma_1 s (\omega + \omega_{ps1})), \omega_{db1} = 0.5 \text{ mm}, \) and \((Z_0 \sigma d f)_{2, \pm} = Z_0/R[\square] \) where \(\omega_{ps1} = eB/(mc)\) is the cyclotron frequency of the 2-DEG, \(\gamma_1 = e/(m \mu)\) is the scattering rate of the 2-DEG, \(\omega_{ps1} = 4\pi ne^2/(mc)\) is the plasma frequency of the 2-DEG, \(m\) is the quasiparticle mass, \(B\) is the applied magnetic field, \(e\) is the electronic charge, \(c\) is the speed of light, \(\mu\) is the mobility, and \(n\) is the 2-D number density.

The Fresnel amplitudes \(r_\perp\) and \(t_\parallel\) as a function of \(n, \mu, R[\square], m,\) and \(B\) are then found by substituting the above parameters into the following equations:

\[
\begin{align*}
(t_+ & ) = M_{2, +} K_{2, +} M_{1, +} \begin{pmatrix} 1 \\ \tau_+ \end{pmatrix} \\
(t_- & ) = M_{2, -} K_{2, -} M_{1, -} \begin{pmatrix} 1 \\ \tau_- \end{pmatrix}
\end{align*}
\]  

(A1)

The reflection \(R\) and transmission \(T\) for unpolarized
light, reflection $R_\pm$ and transmission $T_\pm$ for circularly polarized light, and the Faraday $\theta_F$ and Kerr $\theta_K$ angles are calculated from the Fresnel amplitudes as follows:

$$R_\pm = r_\pm r_\mp^*$$
$$T_\pm = t_\pm t_\mp^*$$
$$R = (1/2) (R_+ + R_-)$$
$$T = (1/2) (T_+ + T_-)$$
$$\tan(\theta_F) = i(t_+ - t_-)/(t_+ + t_-)$$
$$\tan(\theta_K) = i(r_+ - r_-)/(r_+ + r_-)$$

where $^*$ notates the complex conjugate.

3. $\text{Bi}_2\text{Se}_3$

$\text{Bi}_2\text{Se}_3$ is modelled as a surface state Drude response on top of a very thick bulk slab whose response consists of a free carrier Drude term with two phonon resonances. The bulk phonon parameters as well as the approximate cyclotron mass and scattering rate have previously been measured.\(^{22}\)

The bulk dielectric function is given by the following:

$$\epsilon_\pm = \frac{\omega_{pb}^2}{\omega(\omega \pm \omega_{cb} - i\gamma_b)} + \sum_{j=1}^{2} \frac{S_j}{\omega_j^2 - \omega(\omega - i\gamma_j)} + \epsilon_\infty$$

where $j$ enumerates the phonon oscillators, $\omega_j$ is the TO phonon frequency, $S_j$ is the phonon spectral weight, a subscript $b$ refers to bulk electrons, $\epsilon_\infty$ is the dielectric constant at higher infrared frequencies, $\omega_{pb}$ is the unscreened plasma frequency for free electrons: $\omega_{pb}^2 = 4\pi N_b e^2 / m_b^*$, where $N_b$, $e$ and $m_b^*$ are concentration, charge, and an effective mass of electrons; $\gamma_b$ and $\gamma_j$ are the damping rates; $\omega_{cb} = eB/(m_e c)$ is the bulk cyclotron frequency, where $B$ is magnetic field, $m_e$ is bulk cyclotron mass, and $c$ is the speed of the light.

The reported phonon parameters are $\omega_1 = 67\text{ cm}^{-1}$, $S_1 = 3.94 \times 10^5\text{ cm}^{-2}$, $\gamma_1 = 5\text{ cm}^{-1}$, $\omega_2 = 134\text{ cm}^{-1}$, $S_2 = 7056\text{ cm}^{-2}$, $\gamma_2 = 2\text{ cm}^{-1}$, and $\epsilon_\infty = 25.6.\(^{22}\)$ The reported bulk cyclotron mass is approximately $m_{cb} \approx 0.16 m_e$, where $m_e$ is the bare electron mass and the approximate bulk scattering rate $\gamma_b \approx 8\text{ cm}^{-1}$, although both are left as parameters in the model. The relation $m_b^* = m_{cb}$ is assumed true, so that $N_b$ (or equivalently the plasma frequency, $\omega_{pb}$) is the third adjustable parameter associated with the bulk response. With these values, $\epsilon_\pm$ is a function of radiation frequency $\omega$ and applied magnetic field $B$, and the bulk plasma frequency $\omega_{pb}$, mass $m_{cb}$, and scattering rate $\gamma_b$.

Since the surface state penetration depth is $\sim \text{nm}$ which is much less than the optical wavelength for terahertz radiation, the surface state may be modelled as a two dimensional sheet of charge. The model of a 1 cm thick $\text{Bi}_2\text{Se}_3$ slab in vacuum with a Drude surface state response at the first interface are as follows:

$$n_{1,\pm} = 1, n_{2,\pm} = \sqrt{c_\pm}, n_{3,\pm} = 1, (Z_0\sigma df)_{1,\pm} = \omega_{ps}^2 / (\gamma_s - i(\omega \pm \omega_{cs})), dB_2 = 1\text{ cm}, (Z_0\sigma df)_{2,\pm} = 0$$

where $\omega_{cs} = eB/(m_e c)$ and $\omega_{ps} = (4\pi/e)n_s e^2/m_s$. Even for very low densities of bulk carriers, the properties of the last interface do not effect the reflected optical response due to the large attenuation within the thick bulk crystal.

Inserting the above values into Eqs.\(^{[A1]}\) give the reflection and transmission Fresnel amplitudes as a function of $\omega$, $B$, $\omega_{pb}$, $\gamma_b$, $m_{cb}$, $\omega_{ps}$, $\gamma_s$, and $m_{cs}$. The Fresnel amplitudes are also given in Ref.\(^{[19]}\) Eqs. 4 to 6.

Digitizing and fitting the reported ARPES measured dispersion relation for the surface state from Ref.\(^{[1]}\) gives the following functional form:

$$E_F = 2.02 k_f + 10.44 k_f^2$$

where the Fermi energy $E_F$ is measured in $\text{eV}$ above the Dirac point and $k_f$ is measured in $A^{-1}$. $n_s$ (or $\omega_{ps}$), $m_{cs}$, $\gamma_f$ and $k_f$ are fully determined by the dispersion relation for any given Fermi energy.

Using the dispersion relation, $R$, $T$, $R_\pm$, $T_\pm$, $\theta_F$, and $\theta_K$ are calculated from Eq.\(^{[A2]}\) as a function of $\omega$, $B$, $\omega_{pb}$, $\gamma_b$, $m_{cb}$, $E_F$, and $\gamma_s$.

Appendix B: Expected optical signatures of surface states based upon ARPES measurements

1. Expected Kerr and Reflectance signals

Figure\(^{[4]}\) shows calculated results for the expected Kerr and reflection signals, both from linearly and circularly polarized light, as a function of magnetic field for various Fermi energies ranging from 0 to 400 meV above the Dirac point. All graphs in Fig.\(^{[4]}\) are calculated assuming a bulk cyclotron mass value of $m_{cb} = 0.15 m_e$ and bulk scattering rate $\gamma_b = 10\text{ cm}^{-1}$. The surface state plasma frequency $\omega_{ps}$ and cyclotron mass $m_{cs}$ are determined by the ARPES measured dispersion (Eq.\(^{[A4]}\) for a specific surface Fermi energy. Each row of graphs are calculated with a different bulk plasma frequency $\omega_{pb}$ and surface scattering rate $\gamma_s$.

The optical response for a pure surface state with $\gamma_s = 10\text{ cm}^{-1}$ is shown in Fig.\(^{[4]}\)(a-d) (the response of the bulk is negligible for $\omega_{pb} = 10\text{ cm}^{-1}$). All signals are well within the capability of the Kerr measurements. Note that Re($\theta_K$) and Im($\theta_K$) show antiresonant- and resonant-like behavior. The cyclotron mass and strength of the response increase with Fermi energy. The reflection in purely circularly polarized light depicted in Fig.\(^{[4]}\)(d) exhibits a hump in the cyclotron resonant active mode. The reflection in linear polarization depicted in Fig.\(^{[4]}\)(c) (composed of equal amounts of left and right circularly polarized light) shows two humps since there is an active cyclotron mode in positive and negative fields. The qualitative structure of the Kerr and reflection signals is instructive since they appear superimposed on larger bulk responses at larger values of $\omega_{pb}$.

Row 2 of figures, namely Fig.\(^{[4]}\)(e-h), show the optical response expected with a bulk plasma frequency of $\omega_{pb} = 400\text{ cm}^{-1}$ and surface scattering rate $\gamma_s = 3\text{ cm}^{-1}$. The
As the Drude bulk parameters and mass are calculated based upon the Fermi level and the measured ARPES dispersion relation.

The last row of figures, Fig. 4(m-p), show the effect of increasing the surface scattering rate to \( \gamma_s = 25 \text{ cm}^{-1} \). Fitting lineshapes on this level to discern the effects produced by surface states is not conduciive to the current measurement scheme.

2. Expected Kerr signals with application of a gate

A gate which concurrently allows ample throughput optical power while inducing negligible Kerr or Faraday rotation has been accomplished. High resistivity silicon substrates with a 300 nm film of SiO\(_2\) were ion implanted with peak concentrations \( > 10^{19} / \text{cm}^3 \) resulting in a density profile thickness of \( \sim 0.3 \mu\text{m} \). Characterization of these devices show very small Faraday rotation up to 8 T, excellent optical throughput power, no carrier freeze out, and large break down voltages. The devices are currently being used to gate large CVD grown graphene in similar magneto-optical experiments.

Estimates of the shift in Fermi energy of the surface state associated with the application of electric fields commensurate with the typical breakdown voltages of SiO\(_2\) depends upon the Fermi energy. For currently available samples the surface state charge density is sufficiently large that it dominates the charge transfer to the sample due to the gate. As a result the band offset is reasonably given by the change in the Fermi energy due to the charge transfer to the surface states. The depletion depth in the bulk is then given to a good approximation by \( d^2 = 2U_0\epsilon_0 / e^2 n_b \) (where \( U_0 \) is the energy above the Dirac point and \( \epsilon_0 \) is the bulk static dielectric constant). Therefore, the net charge which one can add or subtract from the surface state without breakdown of the insulator is very nearly equal to the maximum allowable charge transfer with the application of a gate. Using a maximum applicable electric field value of \( 3.3 \times 10^8 \text{ V/m} \) for SiO\(_2\) is equivalent to a net change in surface charge density of \( 7 \times 10^{12} \text{ cm}^{-2} \). Based upon the ARPES measured dispersion, this gives a shift of Fermi energy \( \Delta E_f \approx 90 \text{ meV at } E_f =0 \), \( \Delta E_f \approx 30 \text{ meV at } E_f =200 \text{ meV} \), and \( \Delta E_f \approx 15 \text{ meV at } E_f =350 \text{ meV} \). Since the gate voltage can be reverse biased as well, roughly twice these changes can be realized. The resulting depletion depth

FIG. 5: (color online) Calculated optical response produced by a hypothetical gate voltage which shifts the Fermi energy of the surface state by 30 meV. The change in Kerr angle produced by this shift at various Fermi energies for a specific surface scattering rate is shown for each column. All graphs are calculated using the bulk phonon parameters measured in Ref. 19 as well as the Drude bulk parameters \( \omega_{ph} = 400 \text{ cm}^{-1} \), \( \gamma_b = 10 \text{ cm}^{-1} \), and \( m_b = 0.15 m_e \). The surface state 2-D plasma frequency and mass are calculated based upon the Fermi level and the measured ARPES dispersion relation. The surface scattering rates are assumed to be (a) \( \gamma_s = 10 \text{ cm}^{-1} \), (b) \( \gamma_s = 25 \text{ cm}^{-1} \), (c) \( \gamma_s = 50 \text{ cm}^{-1} \), and (d) \( \gamma_s = 100 \text{ cm}^{-1} \).
decouples the surface states, but is small enough that the insulating depletion layer is optically invisible at THz frequencies.

Based upon these estimates, the application of a gate is chosen to produce a change in Fermi energy of $\Delta E_f = 30$ meV. Fig. 5 shows the expected Kerr signals predicted by the ARPES measured surface state properties with bulk parameters of $\omega_{pb} = 400$ cm$^{-1}$, $\gamma_b = 10$ cm$^{-1}$, and $m_{cb} = 0.15 m_e$. Each graph shows Fermi level shifts of 0-30 meV, 30-60 meV, 70-100 meV, 120-150 meV, 170-200 meV, 270-300 meV, and 370-400 meV. Graphs (a-d) show the effects of increasing the surface scattering rate from $10 \text{ cm}^{-1}$ to $100 \text{ cm}^{-1}$. Reflectance measurements are not very sensitive, showing that $R/R[0 \text{T}] \lesssim 0.5\%$ for $\gamma_s = 25 \text{ cm}^{-1}$ and $\lesssim 0.1\%$ for $\gamma_s = 50 \text{ cm}^{-1}$.

Changes in Kerr angle at the level of $0.1 \text{ mrad}$ is very reasonably obtainable in this configuration. It should be noted that the signals are maintained at these general levels even for higher bulk plasma frequencies up to $600 \text{ cm}^{-1}$.

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