Far infrared cyclotron resonance and Faraday effect in Bi$_2$Se$_3$

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The complex Faraday angle and transmission for the topological insulator Bi$_2$Se$_3$ were measured in the far infrared in magnetic fields up to 8 T and analyzed in terms of a simple Drude-Lorentz magneto-plasma dielectric model. Bulk carriers dominate the optical response. The bulk carriers are electrons as we determined from the sign of the Faraday angle. We obtain the bulk band edge cyclotron mass $m_{cb} = 0.16 \pm 0.01 m_e$ and free electron concentration in the $10^{17}$ range. Electron-phonon interaction effects were found to be weak.

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I. INTRODUCTION

It was shown recently theoretically and confirmed experimentally by ARPES that Bi$_2$Se$_3$ and Bi$_2$Te$_3$ have an odd number (one) of Dirac cone surface states in the unit cell and thus are nontrivial topological insulators. This makes them distinct from graphene which also has Dirac electrons but an even (two) number of cones per unit cell. Predictions of several novel properties have generated a great interest in these materials.

Although the optical properties of Bi$_2$Se$_3$ have been studied in the past, only recently it has been probed for unusual far infrared magneto-optical responses. We have grown Bi$_2$Se$_3$ crystals with low free electron concentration which enabled observation of the cyclotron resonance and Faraday rotation below the strong low frequency polar phonon. In this paper, we report results of these measurements together with the model calculations.

II. EXPERIMENTAL

Single crystals of Bi$_2$Se$_3$ were grown as described in Ref. 3 and we used a sample from batch iv. A 2×2×0.014 mm flake was exfoliated on a firm surface. The sample thickness was obtained from etalon oscillations and confirmed by electron microscope images of the sliced flake. Infrared data were obtained on a Bomem DA3 Fourier-transform (FTIR) spectrometer and an optically pumped far infrared laser at a frequency of 42.3 cm$^{-1}$. The samples were placed either in an optical cryostat for measurements in magnetic fields up to 8 T in the Faraday geometry: $k_{\text{light}} \parallel B$ and $B \parallel c$-axis. Our technique for the complex Faraday angle measurements in the far infrared is described in Refs. 3 and 8.

III. MODEL

The experiments reported here differ from conventional cyclotron resonance in semiconductors because the relatively high carrier density and low lying phonon leads to strong magneto-plasma effects. This results in a very low transmission near the plasma frequency and multiple reflections inside the flake in transparent regions (etalon effect) which produces a complicated optical response. Therefore, it is necessary to model the magneto-optical response in order to obtain the material parameters. We model our experiment as transmission and reflection of a slab with bulk optical constants and (possibly) two identical 2D layers with surface optical constants on both surfaces of the slab. For our free carrier concentration level, the Fermi energy is less than 20 meV above the bottom of the lowest conduction band. This is the only filled band because, according to Ref. 8, the second conduction band minimum is 40 meV higher. Since $\hbar \omega$ and $E_F$ are small compared with the band gap $\approx$300 meV, a parabolic dispersion of the bulk electrons is assumed. Therefore, experiment gives essentially the band edge mass.

In this case, we can use a Drude magneto-optical response of the free electron gas (1st term in Eq. 1). The corresponding complex dielectric function for the bulk is:

$$
\epsilon(\omega) = \frac{\omega_p^2}{\omega(\omega \pm i\gamma_b - i\gamma_b)} + \sum_j S_j \frac{\omega_j^2}{\omega^2 - \omega(\omega - i\gamma_j)} + \epsilon_\infty
$$

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, $\omega_{pb}^2$ 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; $\omega_{cb} = eB/(m_b c)$ is the cyclotron frequency, where $B$ is magnetic field, $m_{cb}$ is bulk cyclotron mass, and $c$ is speed of light. The ± sign refers to the right and left circular polarized electromagnetic modes. For a fixed (laser) frequency, we use a quarter-wave plate to obtain circular polarized light. In case of the far infrared spectra, we used linear polarized light thus measuring an average of two circular polarizations. For a general polarization state, measured transmission is $T = (0.5 + \beta)T^+ + (0.5 - \beta)T^-$, where $-0.5 \leq \beta \leq 0.5$.

The surface states of a topological insulator are 2D
Dirac quasiparticles that split into Landau levels in an applied $B||c$ magnetic field:

$$E_n = sgn(n) \sqrt{\hbar \omega_{cs}},$$
$$\omega_{cs} = v_F \sqrt{2eB/\hbar}$$

where integer $n$ enumerates energy levels, $e$ and $v_F$ are charge and Fermi velocity of the Dirac particles. The full band dispersion for Bi$_2$Se$_3$ has been recently reported in Ref. 10. The optical dipole selection rules allow transitions $|n| \rightarrow |n| + 1$ for the cyclotron resonance active mode of circularly polarized light. The linear dispersion leads to the square root dependence on $n$ and $B$. However, in our sample the chemical potential $\mu$ leads to the square root dependence on $s$.

Ref. 10. The optical dipole selection rules allow transitions $|n| \rightarrow |n| + 1$ for the cyclotron resonance active mode of circularly polarized light. The linear dispersion leads to the square root dependence on $n$ and $B$. However, in our sample the chemical potential $\mu$ leads to the square root dependence on $s$. Therefore, we find $n \approx 25$ for 8 T magnetic field. At these values of $n$, we can use an approximate formula

$$E_{n+1} - E_n \approx \hbar eBv_F^2/E_n = \hbar \omega_{cs}$$

and the cyclotron resonance becomes semiclassical. The combination $E_n/v_F^2$ plays the role of the cyclotron mass $m_{cs}$ for surface states. Therefore, we model the optical response of the surface states as a 2D free electron gas.

We use conventional expressions for transmission and reflection of a slab (medium 2, bulk) between media 1 and 3 (both vacuum here) for light propagating from 1 to 3 with the following modifications. The presence of surface states at the interfaces 1-2 and 2-3 changes the complex Fresnel transformation between circular and linear polarization bases $t^\pm = t_{xx} \pm it_{xy}$ is assumed.

We estimate $\omega_{ps}$ using the ARPES data and expressions for 2D free Fermi gas (one spin per state): $k_F^2 = 4\pi N_s$. For $E_F \approx 10$ meV, the surface states are below $k_F = 0.08 \, \text{Å}^{-1}$ which gives $N_s \approx 5 \times 10^{12} \, \text{cm}^{-2}$ and $\omega_{ps} \approx 20 \, \text{cm}^{-1}$ assuming $m_s^* = 0.15m_e$.

### IV. EXPERIMENTAL RESULTS AND DISCUSSION

Zero field far infrared results for this sample were reported earlier. Set #1 shown in Table 1 is the result of a simultaneous fit of zero field reflectivity and transmission. Other sets of fit parameters shown in Table 1 model results from different magnetic field experiments discussed below. Data of experiment #2 were fitted by the model of two crystals with parameters #2-1 and #2-2 and effective areas 82 % and 18 %, correspondingly. We understand variation of the bulk concentrations from one experiment to another and within experiment #2 as a consequence of nonuniform carrier concentration of the sample. Characteristic size of the regions with different concentrations should be on the order of the infrared laser focal spot ($\approx 1$ mm). In FTIR measurements, the light spot was larger than the sample aperture. We also cannot rule out a possible increase of the effective bulk concentration with time due, for example, to exposure of the flake to air during the few months separating measurements #1 and #4.

Figure 1 shows the measured and calculated transmission spectra of the Bi$_2$Se$_3$ flake. Oscillations of the transmission in zero field appearing below 60 cm$^{-1}$ are due to the etalon effect within the transparency band between the plasma frequency and the TO phonon frequency. In non zero fields, the magneto-plasma dielectric function (1) changes due to the cyclotron resonance, and the etalon features for left and right circular polarization split in frequency and change their amplitudes. The material becomes more transparent in magnetic field because the $T^+$(cyclotron resonance inactive) polariz-

| Set | $\omega_{ph}$ | $\gamma_s$ | $m_{ch}$ | $\omega_{ph} (8\,\text{T})$ | $N_s$ | $B$ | $E_F$ |
|-----|---------------|------------|----------|-----------------|------|-----|------|
| #1  | (cm$^{-1}$)   | (cm$^{-1}$) | (meV)    | (10$^{17}$ cm$^{-3}$) | (meV) |
| FTIR | 382 | 8 | 0.15 | 50 | 2.3 | 8 |
| laser | 385 | 7.5 | 0.16 | 48 | 2.6 | 8 |
| laser | 750 | 7.5 | 0.16 | 48 | 9.9 | 20 |
| FTIR | 465 | 8 | 0.16 | 48 | 3.8 | 11 |
| laser | 520 | 8 | 0.17 | 44 | 5.1 | 12 |

*a* In chronological order. Fixed phonon parameters are $\omega_1 = 67 \, \text{cm}^{-1}$, $S_1 = 3.94 \times 10^6 \, \text{cm}^{-2}$, $\gamma_1 = 5 \, \text{cm}^{-1}$, $\omega_2 = 134 \, \text{cm}^{-1}$, $S_2 = 7056 \, \text{cm}^{-2}$, $\gamma_2 = 2 \, \text{cm}^{-1}$, and $\epsilon_{\infty} = 25.6$.

$b\, m^*_s = m_{ch}.$
which will affect the optical response in a nontrivial way. A distribution of concentrations will cause a distribution in line shape expected from a single uniform bulk response. Inhomogeneity may cause deviations from the simple calculations. Therefore, the free carriers in our Bi$_2$Se$_3$ flake have the same sign as for the Si:B wafer. However, the sign of Bi$_2$Se$_3$ carriers is opposite to holes in our magneto-optical experiments is determined by the sign of the carrier charge, the direction of magnetic field, the chirality of incident light, and other experimental conditions. To avoid any possible ambiguity, a boron doped Si wafer with an anti-reflection coating in which the free carriers are holes was measured as a reference sample. The measured Faraday rotation for the Bi$_2$Se$_3$ flake has the same sign as for the Si:B wafer. It should be noted that all measurements are in the free carrier concentration range set by parameters #2-1 and 2-2. The 50 K data is very well fit with the same parameters as fit the 10 K data except $\gamma$ requires an increase from 7.5 to 13.5 cm$^{-1}$. This suggests that only the free carrier damping rate changes over this temperature range. At higher temperatures, phonon parameters and effective free carrier concentration may also change.

Figure 3 shows the transmission of circular polarized light and the complex Faraday angle (Faraday rotation and circular dichroism) together with a model calculation with one set of parameters (#4). The green dash-dot curves were calculated using fit #1 parameters and clearly deviate strongly from the experimental results. The qualitative difference in shapes of the transmission curves for figures 2 and 3(a) is also understood in terms of carrier concentration inhomogeneities.

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The sign of $\omega_c$ in our magneto-optical experiments is determined by the sign of the carrier charge, the direction of magnetic field, the chirality of incident light, and other experimental conditions. To avoid any possible ambiguity, a boron doped Si wafer with an anti-reflection coating in which the free carriers are holes was measured as a reference sample. The measured Faraday rotation for the Bi$_2$Se$_3$ flake has the same sign as for the Si:B wafer. However, the sign of Bi$_2$Se$_3$ carriers is opposite to holes because of a sign change of the Faraday response due to an etalon effect in the flake as we see from the model calculations.
sample are electrons.

These results also bear on electron interaction effects. While the Coulomb interaction is weak because of the large static screening ($\epsilon_0 \approx 100$), a significant electron-phonon interaction may be expected in this strongly polar material. In either case, the carriers scattering rates and effective masses reflect the significance of the many body interaction effects. Electron-electron scattering is expected to have $T^2 + \omega^2$ dependence from Coulomb scattering and an onset of scattering for $\omega > \omega_{LO}$ due to LO phonon emission. In addition, the optically measured cyclotron mass differs, in principle, from the value measured from quantum oscillations measurements which includes the self energy corrections to the band mass due to interactions. For the optical measurement of cyclotron resonance, the Kohn theorem insures that the resonance is controlled by the band mass provided that the electro-dynamics is sufficiently local. In the Faraday geometry of our experiments, the non locality leads to a Doppler shift of the cyclotron resonance at $\omega = \omega_c - qv_F$, where $q$ is a characteristic electrodynamic wave vector. For this system $q \approx \omega_p/c \approx 10^{10} << \omega_c/v_F$, so that the expected shifts are very small. Therefore, the observed cyclotron mass should correspond to the band mass. The optically measured $m_{cb} = 0.15 \pm 0.01$. The cyclotron mass deduced from SdH measurements on samples from the same batch was $m_{SdH} = 0.15 \pm 0.01$. The near equality of these two values implies that mass enhancement from many body interactions is very small in this material.

In figure 4 we show the room temperature reflectivity spectrum of a crystal with a higher free carrier concentration ($N_b = 8.5 \times 10^{18}$) than the flake. The spectrum shows a plasma edge at 500 cm$^{-1}$. The plasma edge feature is very sensitive to the electron relaxation rate at this frequency. The electronic relaxation rate implied by this spectrum is $\gamma_b = 58$ cm$^{-1}$. It is noteworthy that this relaxation rate is relatively small considering that the plasma edge is significantly above the LO phonon frequency which is $\omega_{LO} \approx 150$ cm$^{-1}$. This relaxation rate is comparable to the relaxation rate measured at lower frequencies at room temperature so that LO phonon scattering does not add significantly to the relaxation rate.

Therefore, from these observations, we see that the electron-phonon interaction effects are too small to be observed on the scale of the uncertainty of the effective mass and the impurity scattering rate. This surprising conclusion may result from the free carrier screening of the electron-phonon interaction.

It was argued in Ref. [5] that the electron-phonon interaction is strong based in part on the anomalous line shape of the lowest infrared phonon. In fig. 4 (inset), we show results of our model calculation for this phonon in magnetic field at low temperatures. The line shape of the phonon in magnetic field is an average of two very different reflectivities $R^+$ and $R^-$ which results in apparent softening and broadening of the phonon line. Our trans-
mission data presented in fig. 1 also show insensitivity of
the lowest phonon to applied magnetic field. Thus, the
observations of Ref. 5 on this phonon at least partially
originate in cyclotron resonance physics rather than in
electron-phonon and magneto-electric couplings.

In conclusion, we have measured the bulk cyclotron
mass for a low doped Bi$_2$Se$_3$ single crystal which is close
to the mass observed in SdH measurements. Free carri-
ers are electrons as we determined from the sign of the
Faraday angle. We have shown that an effective Drude-
Lorentz model well describes the magneto-optical data
which is dominated by etalon effects in the slab sample.

No signature of strong electron-phonon interaction is ob-

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