High mobility topological interface state probed by terahertz measurements

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We report gated terahertz cyclotron resonance measurements on epitaxial Bi2Se3 thin films capped with In2Se3. The properties of a single topological interface state are measured from the vicinity of the Dirac point to above the conduction band edge. A precipitous drop in the scattering rate with Fermi energy indicates decoupling of the surface states from bulk states and a shift of the Dirac cone towards mid-gap. Near the Dirac point, the mobility is 3500 cm²/V·s with measured potential fluctuations of 60 meV.

The topological insulator is a unique state of matter that is a bulk insulator possessing a topologically protected metallic surface state (TSS) of massless particles known as Dirac fermions. Key properties of this spin textured TSS are suppression of back scattering and an intrinsic magneto-electric effect.2,3 The experimental realization of this state of matter in Bi2Se3 and other compounds has sparked great interest owing not only to their potential use in spintronic devices but in the investigation of the fundamental nature of topologically nontrivial quantum matter.4,5 Bi2Se3 is one of the most important topological insulators due to a single Dirac point existing above the valence band edge and a large bulk bandgap of 300 meV offering the prospect of room temperature applications.4 Unfortunately, unintentional doping of these materials has led to large bulk densities at the surface that mask the electronic properties of the surface state.6,7 Capping materials provide a way to control this doping, both chemically and as a platform for gating, but the usual surface probes that have studied the vacuum/TSS interface states2,12,13 can not measure buried interfaces where critical Dirac cone properties are expected to change.14,15 In this letter we report gated terahertz (THz) cyclotron resonance measurements on passivated Bi2Se3 thin films that directly measure the scattering rate and cyclotron mass of a single topological surface state from above the conduction band edge to the vicinity of the Dirac point.

Transmission measurements through a thin film of topological insulator probe all carriers, which can include topological surface states, accumulation layers, and bulk carriers as depicted in Figure 1(a). Modulating the topologically protected interface state of a capped film with a semitransparent top gate while measuring the modulated terahertz transmission allows extraction of its properties, even in the presence of much larger background conductivity contributions. Such measurements performed in magnetic field give not only the carrier density and scattering rate but also the sign of the charge and cyclotron mass of the carriers. For negative gate biases that move the Fermi level of the surface state well below the conduction band edge, Dirac cone electrons are expected to exhibit a small cyclotron mass \( m_c = \hbar k_F / v_F \) since the Fermi wavevector \( k_F \) goes to zero while the Fermi velocity \( v_F \) remains finite at the Dirac point.16,17

An epitaxial Bi2Se3 60 quintuple layer film and a 10 nm In2Se3 capping layer were deposited without breaking vacuum onto a sapphire substrate.19 The dielectric parylene-C and a NiCr semitransparent gate were subsequently deposited (see the Supplemental Materials for details). The device is depicted in Figure 1(b).

Zero-gate normally incident transmission measurements are shown in Figure 1(c,d). Transmission \( T \) through a thin film is related to the two dimensional conductivity \( \sigma \) by \( T \sim |n_{sub} + 1 + Z_0\sigma|^2 \), where \( n_{sub} \) is the index of refraction of the substrate and \( Z_0 \) is the impedance of free space. \( \sigma \) is a sum of all contributing channels. The Drude conductivity in a magnetic field expressed in the circular polarization basis is \( \sigma^\gamma = (\omega_p^\gamma/Z_0)(\gamma - i(\omega \pm \omega_c)) \), where the two dimensional plasma frequency is \( \omega_p^\gamma = Z_0 ne^2/m \) and \( \omega \) is the radiation frequency. The cyclotron frequency \( \omega_c \) is related to the carrier charge \( e \) and cyclotron mass \( m_c \) by \( \omega_c = eB/(cm_c) \), where \( B \) is the applied magnetic field. In zero magnetic field, this conductivity expression depends only on the two dimensional carrier density \( n \), effective mass \( m \), and scattering rate \( \gamma \).

The Fourier transform transmission measurement of Figure 1(c) was performed in zero magnetic field with unpolarized incident light. The cyclotron resonance transmission measurement is shown in the inset of Figure 1(c). It was performed with circularly polarized incident light at a fixed laser frequency as a function of magnetic field applied normal to the film. The cyclotron resonance measurement exhibits a single Lorentzian absorption. The resonant position located in positive magnetic field cor-

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responds to n-type carriers of mass $m_e \approx 0.19 \, m_0$. The Faraday angle $\theta_F$ reported in Figure (d) was measured with polarization-modulated incident light at a fixed frequency, a method detailed elsewhere. Fits to the zero-gated transmission data in Figure (c,d) using a single-fluid Drude model for the conductivity, depicted by the blue curves, reveal a large carrier density $n \approx 10^{13}$ cm$^{-2}$ and a cyclotron mass $m_c \gtrsim 0.19 \, m_0$ that is much larger than the conduction band edge mass $m_\text{c} = 0.15 \, m_0$. Deviations between the $\theta_F$ data and fit are outside of measurement error, hinting at small contributions from other conducting channels.

Transport measurements in similarly grown epitaxial films, performed over a wide range of thicknesses, show the bulk contribution to the conductivity is a small fraction compared to the surface layer contributions. Therefore, the observed large carrier density is likely associated with a top and/or bottom surface accumulation layer due to defects.

Modulating the top gate voltage at $\sim 1$ Hz and measuring the difference in cyclotron resonance ($\Delta$-CR) reveals the top topological interface state properties. The largest negative average gate voltage represented by the red curve of Figure (a) shows a resonant component at $B \approx 2$ T. This corresponds to half the conduction band edge mass $m_e$. Such a small mass is evidence of the topological interface state Dirac cone and a Fermi level of approximately 50 meV above the Dirac point, far below the conduction band edge of 190 meV observed on freshly cleaved Bi$_2$Se$_3$ in vacuum.

The small mass associated with the top surface establishes that the large accumulation layer dominating the zero-gate response is on the bottom surface. Changing the gate from zero to the maximum of -170 V transfers $4.4 \times 10^{12}$ e/cm$^2$, a fraction of the $10^{13}$ e/cm$^2$ associated with the zero-gate measurement. Gating the top topological surface state to the vicinity of the Dirac point requires removing any accumulated charge in addition to depleting the surface state and a region of the bulk near the surface. Given the gate limitation, this is not possible if the top interface is accumulated. The large accumulation layer at the bottom surface includes a bottom surface topological surface state. These two large mass contributions are indistinguishable in our differential measurements since the bottom surface is not significantly modulated by a top gate.

The observed mass dependence on gate voltage is consistent with a Dirac cone surface state below the conduction band edge. In order to gain further insight into the gating behavior of the surface state, the optical response of the film is modeled in terms of the surface state dispersion as measured by angular-resolved photoemission spectroscopy (ARPES). The model incorporates three n-type conductivity contributions distributed in the film (details are provided in the Supplemental Material): one term for the top topological surface state (TSS), another for the top accumulation layer and/or interim bulk carriers (IB), and the third term for the bottom surface accumulation layer (BSAL). Each conductivity contribution is expressed within a Drude model with parameters for carrier density $n$, scattering rate $\gamma$, and cyclotron mass $m_e$. The BSAL is not modulated by the gate and its Drude parameters are chosen to be consistent with the zero-gate measurements. To reduce the number of parameters associated with the gate modulated carriers, the amount of charge a gate transfers on or off the topological surface state is determined by solving for the band bending in the film using Thomas-Fermi screening theory. The IB parameters are gate-dependent where $m_e$ and $\gamma$ are free parameters, but the change in carrier density is determined by the band bending model. The gate dependent TSS parameters $n$ and $m_e$ are set by the ARPES measured dispersion relation, while $\gamma$ is the only free Drude parameter.

The differential cyclotron resonance measurements are
Gated magneto-optical transmission measurements at 10K: Average gate values $V_g$ for each plot are coded according to the colored bar at the top of the figure. All optical signals are differences between two gate values, $V_g \pm \Delta V/2$, measured at every integer B-field value up to ±8 T. Shown on each data graph is the radiation frequency $\omega/(2\pi)$ and $\Delta V$ value. (a) Differential cyclotron resonance data normalized to the average with a standard deviation of the mean of 5 mrad. (b,c) Differential Faraday angle data measured with a standard deviation of the mean of 30 µrad. (d-f) The modeled optical responses are detailed in text. The colors of the modeled curves correspond to the same gate values appearing in the respective data sets (a-c).

FIG. 2: Gated magneto-optical transmission measurements at 10K: Average gate values $V_g$ for each plot are coded according to the colored bar at the top of the figure. All optical signals are differences between two gate values, $V_g \pm \Delta V/2$, measured at every integer B-field value up to ±8 T. Shown on each data graph is the radiation frequency $\omega/(2\pi)$ and $\Delta V$ value. (a) Differential cyclotron resonance data normalized to the average with a standard deviation of the mean of 5 mrad. (b,c) Differential Faraday angle data measured with a standard deviation of the mean of 30 µrad. (d-f) The modeled optical responses are detailed in text. The colors of the modeled curves correspond to the same gate values appearing in the respective data sets (a-c).

mainly sensitive to the absorptive (real) part of the conductivity. Additional information is gained from the differential complex Faraday angle data reported in Figures 2(b,c). The real (imaginary) part of $\theta_F$ is related to the reactive (absorptive) part of the conductivity.

Figures 2(d-f) show the modeled differential optical signals. The red curves in Figures 2(c,d) show the zero-gate Fourier transform spectroscopy and Faraday angle model results. A comparison to other ∆-CR measurements at higher frequencies as well as gated spectroscopy data are shown in the Supplemental Materials. Figure 3 summarizes the Drude parameters which reproduce the data sets. All the features are reproduced over a wide range of applied voltages, magnetic fields, and frequencies.

In the ∆-CR data of Figure 2(a), the observed peak-dip-hump structure evolution at negative gate voltage is reproduced by the model, showing that the cyclotron mass $m_c = \hbar k_F/v_F$ is consistent with the Dirac dispersion measured by ARPES (a more thorough intuitive discussion of the model is presented in the Supplemental Materials). However, the modeled differential response at the highest negative gate voltages is larger across all data sets. The differential optical signals are proportional to changes in the optical spectral weight, $n^2/m_c$. The Dirac cone dispersion predicts $n/m_c \propto k_F v_F$ which goes to zero at the Dirac point as do both $n$ and $m_c$. In the presence of potential fluctuations, these quantities saturate to some non-zero rms values as the Dirac point is approached. This saturation of the spectral weight in the vicinity of the Dirac point reduces the differential spectral weight below that predicted by a Dirac cone dispersion. The observed smaller differential optical data near the Dirac point demonstrate deviations from Dirac cone expectations, consistent with potential fluctuations.

These deviations provide a means to characterize the potential fluctuation energy. The first significant deviation between model and data begins with average voltage $V_g = -105$ V with its associated lowest voltage of −130 V. From Figure 3(b), the TSS Fermi level corresponds to ~60 meV above the Dirac point. This estimate of the potential fluctuations agrees well with other estimates.

The TSS scattering rate, shown in Figure 3(c), monotonically decreases with Fermi level transitioning to a much flatter behavior at ~70 meV above the Dirac point, providing another estimate of the puddling energy scale. The independently measured mass and scattering rate of the topological surface state translate into a mobility, reported in Figures 3(f). It is largest in the vicinity of the Dirac point, 3500 cm$^2$/V·s.

The scattering rate suffers a precipitous drop at ~70 meV below the inferred conduction band edge that is based upon the ARPES dispersion. This behavior is very different from that observed in graphene. The scat-
FIG. 3: Model parameters: Three n-type Drude terms model the reported differential optical measurements. The BSAL parameters are $n = 1.0 \times 10^{13}$ cm$^{-2}$, $m_e = 0.25 \ m_0$, and $\gamma = 9.4$ THz. The red through purple dots on all graphs depict the same average voltages applied as in Figure 2(a). The solid orange lines depict the depletion onset and the red dashed lines depict the conduction band edge, inferred from the ARPES measured dispersion.(a) The fraction of charge moved by the gate in the IB (blue) carrier contribution and the TSS (black), where $E_{F_0} = 15$ meV (screening length $\lambda_s = 13.8$ nm), shown as a function of $V_g$. (b) The TSS Fermi level with respect to the Dirac point inferred from ARPES measurements. (c) The cyclotron mass in units of the bare electron mass $m_0$ and (d) the scattering rate are shown for the IB (blue) and TSS (black) carrier contributions. (e) Scattering rates shown as a function of the TSS Fermi level. (f) The mobility, calculated from the measured mass and scattering rate, shown versus the TSS number density.

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See Supplemental information at [URL] for details regarding methods, optical and Thomas-Fermi screening models, gated optical data addenda, and the expected Dirac point shift of topological interface states.

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SUPPLEMENTAL MATERIAL

I. METHODS

A Bi$_2$Se$_3$ 60 quintuple layer film was grown epitaxially onto a 0.5 mm thick sapphire substrate of area 1x1 cm square. A 10 nm thick In$_2$Se$_3$ capping layer was immediately deposited without breaking vacuum. Two contacts were made to the film using Eccobond Solder 59C. Parylene-C was deposited to a thickness of 590 nm, conformally encasing the sample on both the top and bottom surfaces with a measured thickness uniformity better than 10 nm. NiCr films were evaporated onto the bottom surfaces with a measured thickness uniformity of 0.15. Two contacts to the gate were made using the same silver epoxy.

FTIR measurements were performed with a Bomem DA8. A far-infrared laser cavity pumped by a CO$_2$ laser provided the fixed frequency source for the Faraday and cyclotron resonance measurements. The polarization modulation technique used to measure the Faraday angle is detailed elsewhere. Circular polarized light for the cyclotron resonance measurement was generated using quartz quarter-waveplates with NiCr antireflection coatings. A GaAs 2-DEG heterostructure was used to set the waveplate angle, verify the retardance of the waveplates, and served to calibrate the Faraday angle measurements.

II. THOMAS-FERMI SCREENING AND OPTICAL MODELS

Topological surface state dispersion

Peaks associated with the topological surface state in the momentum distribution curves as measured by ARPES were fit along the Γ-K and Γ-M directions at many binding energies ranging from the valence band to well above the conduction band edge. Fits to the dispersion where energy is measured from the Dirac point give $E_{\Gamma-K} = 1.7k + 13.5$k$^2 - 4.600k^6$ and $E_{\Gamma-M} = 2.2k + 8.2k^2 + 4.200k^6$ where $E_F$ is in eV and $k$ is in Å$^{-1}$. There is little difference between the two dispersions from hexagonal warping since our range of Fermi level is from the Dirac point to just above the conduction band edge. The average of the two dispersions is used in the analysis and is given by $E_{av} = 1.9k + 12.6k^2 + 2,300k^6$. The TSS Fermi velocity $v_F = dE/d(k)$, cyclotron mass $m_r = \hbar k_F/v_F$, and carrier density $n = k_F^2/(4\pi)$ are used in analyses.

Back surface accumulation layer

The measured bulk mass near the conduction band edge is 0.15 $m_0$. The ARPES dispersion gives a mass, shown in Figure 3c, of 0.14 $m_0$. Both masses are expected to increase with Fermi level and remain similar in the vicinity of the conduction band edge. High in the conduction band, the two dispersions become very similar so the cyclotron masses are expected to be nearly the same.

The large carrier concentration measured by the zero-gate optical measurements in Figure 3n $n \approx 10^{13}$ cm$^{-2}$, indicates that the Fermi level is high above the conduction band edge. dc transport measurements on similar films show nearly the same 2-D carrier density over a wide range of film thicknesses.

If these carriers were uniformly distributed across the 60 nm film, the resulting bulk carrier density would be $n_b \sim 2 \times 10^{18}$ cm$^{-3}$. This value should be compared with single crystals that are routinely an order of magnitude lower in bulk carrier density. Assuming $E_F = (\hbar k_F)^2/2m^*$ and $n_b = k_F^2/(3\pi^2)$ where the Fermi edge effective mass is take to be 0.15 $m_0$, the resulting Fermi energy above the conduction band edge is 34 meV.

As mentioned in the main text, dc transport argues against this interpretation since similarly grown epitaxial films show the bulk has a very small contribution to the conductivity compared to the accumulation layers for such thin films. The observed large carrier density is shown to be associated with a bottom surface accumulation layer likely due to defects which can result in a very high Fermi level.

Thomas-Fermi screening model in topological insulators

Knowing the capacitance, a gate transfers a determined amount of net charge to or from a topological insulator. The net charge is distributed between the topological surface state and the bulk. Deriving this proportion requires solving the general band bending problem in a topological insulator due to screening of potentials.

In the Thomas-Fermi screening model, the potential satisfies Poisson’s equation which we solve for a semi-infinite slab of topological insulator subject to an externally applied electric field. Figure 5 shows band bending of the conduction band edge as a function of depth with a Fermi level $E_{Fb}$ defined away from the screened region.
the topological insulator, the solution is simply given by:

\[ n_{\text{net}} = \frac{1}{e \phi}, \]

screening length and \( \varepsilon \), the layer.

Conduction band edge bends downward creating an accumulation general form:

\[ \frac{\partial^2 \phi(x)}{\partial x^2} = \frac{\rho(x)}{\epsilon}, \]

where \( S \) is the screening length, and \( x \) is the distance away from the gated surface. Depletion onset occurs when \( e \phi = E_{FB} \). For larger \( e \phi \) (red), a depletion layer forms separating the topological surface state from the bulk carriers. Zero \( \phi \) (blue) defines flat band where the background ion density equals the electron density. For negative \( \phi \) (purple), the conduction band edge bends downward creating an accumulation layer.

\[ \frac{\partial^2 S}{\partial y^2} = (1 - (1 - S)^{3/2}) \]

The differential equation can be written in the following general form:

where \( S = \frac{e \phi}{E_{FB}}, y = \frac{y}{\lambda_S}, \) and \( \lambda_S^2 = \frac{e E_{FB}}{\epsilon^2 \sigma_0} \). \( \lambda_S \) defines the screening length and \( \epsilon \approx 100 \) for Bi2Se3.2 Defining \( E_S \equiv \frac{dS}{dy} \) and integrating, the second order differential equation is converted into a first order one. For the specific case of a semi-infinite slab where \( E_S \) and \( S \) are zero deep inside the topological insulator, the solution is simply given by:

\[ E_S = \pm (2S + \frac{4}{5}(1 - S)^{5/2} - \frac{4}{5})^{1/2} \]

Where the + (-) sign is for the case of band bending down (up), or equivalently \( S(0) < 0 \) (\( S(0) > 0 \)). Depletion onset occurs when \( S(0) = 1 \). Either \( E_S \) or \( S \) on the inside surface at \( y = 0 \) is determined given an externally applied gate voltage and the uncompensated charge associated with the topological surface state whose dispersion is characterized by ARPES.

Optical Models

The general optical models which were used in simulating the various data sets are described. Approximate formulas relating to various optical measurements are presented first which emphasize the physical concepts with minimal interference from details.

The normal-incidence transmission \( T \) for a thin film with multiple contributions to the conductivity \( \sigma \) relative to the transmission of the substrate \( T_0 \) is given by

\[ T = \frac{1}{1 \pm \sum_{i=1}^n y_i^2} \]

where \( y_i = \frac{Z_0 \sigma_i}{\omega}, \) is the surface admittance, \( n_{\text{sub}} \) is the index of refraction of the substrate, and \( Z_0 \) is the impedance of free space.

Fourier transform spectroscopy (FTIR) measurements performed in zero magnetic field are described within a Drude model defined by

\[ \sigma_{xx} = \frac{\omega_p}{\gamma} \]

where \( \omega_p = \frac{Z_0 e^2}{m} \) is the cyclotron frequency, \( m \) is the two dimensional plasma frequency, \( \gamma \) is the scattering rate, \( \omega \) is the radiation frequency, \( n \) is the two dimensional carrier density, \( e \) is the electronic charge, and \( m \) is the effective mass.

Cyclotron resonance (CR) transmission measurements are performed at fixed frequency with normally incident circularly polarized light as a function of applied magnetic field. The surface admittance terms are most simply given in the circular polarization basis where

\[ \sigma^\pm = \frac{\omega_p}{\gamma - i(\omega \pm \omega_\text{ps})} \]

\[ \omega_\text{ps} = \frac{eB}{mc} \]

The cyclotron resonances can be thought of as a sum of absorptive Lorentzian resonances centered at \( B_{\text{res,i}} = (\omega/e)m_i \). Differential cyclotron resonance measurements (\( \Delta \)-CR) can be thought of as a sum of differences of Lorentzians. The polarity and value of the B-field resonance determines the sign and mass of the carriers.

The complex Faraday angle is defined as

\[ \theta_F = i \frac{1}{\lambda_S} \sum_{i=1}^n \sum_{j=1}^n \frac{\eta_{i,j} \eta_{j,i}}{Z_{\sigma_j}^i} \]

where \( \tan \theta_F \approx \theta_F \). The transmission Fresnel coefficients in the circular polarization basis, and 2\( \sigma_{xy} = \sigma^+ - \sigma^- \) are the transmission Fresnel coefficients in the circular polarization basis, and 2\( \sigma_{xy} = \sigma^+ - \sigma^- \). The transmission Fresnel coefficients in the circular polarization basis, and 2\( \sigma_{xy} = \sigma^+ - \sigma^- \) are related to the induced circular dichroism similar to cyclotron resonance measurements. The real part, thought of as the angle of polarization rotation, is related to the reactive part of the conductivity. When the scattering rate is much less than the cyclotron frequency, \( \Im(\theta_F) \) (\( \Re(\theta_F) \)) shows a simple Lorentzian resonance (antiresonance) centered at \( B_{\text{res}} \). Contrary to CR measurements, n and p-type carrier resonances will simultaneously appear in both polarities of magnetic field but with opposite sign.

The actual model used to simulate the data is derived from a general solution to the transmission Fresnel coefficients for light normally incident on a stack of slabs which may have multiple conducting films at interfaces. The formalism is given in Reference [11] Appendix I.

Maxwell’s equations inside media with appropriate
boundary conditions are used to derive two matrices which can be viewed as transfer functions for the electric field vector. The $2 \times 2$ matrix $M[n_i, n_{i+1}, \sigma]$ transfers the electric field across an interface consisting of a film(s) with a net 2D conductivity $\sigma$ from a slab of index $n_i$ to a slab of index $n_{i+1}$. The matrix $K[n_i, d_i]$ propagates the electric field through a particular slab of thickness $d_i$ with index $n_i$.

Since the parylene coating and In$_2$Se$_3$ capping layer are thin (compared to wavelength) non-absorbing dielectric films in our frequency range, they do not affect the optical signals so we leave them out. The Bi$_2$Se$_3$ film is idealized as a conducting 2D film on top of an insulating Bi$_2$Se$_3$ dielectric slab that has optical absorptions due to phonons. Therefore the first boundary is between air and the idealized Bi$_2$Se$_3$ insulating slab, and between them is a NiCr film (used as a gate and characterized by the dc sheet resistance $R_g$) and another conductor associated with the Bi$_2$Se$_3$ free carrier response represented as $\sigma^\pm$. All contributions from all the free carriers in the Bi$_2$Se$_3$ film are lumped into $\sigma^\pm$. The second interface between the Bi$_2$Se$_3$ insulating slab and sapphire substrates is considered to have no free carriers. The last interface is sapphire into air with a NiCr film, an anti-reflection coating characterized by sheet resistance $R_{AR}$.

Using the transfer matrices, the Fresnel transmission coefficients $t^\pm$ are derived for the entire stack of slabs. All optical quantities presented in this paper are calculated normalized to the zero gate transmittance and is given by $\Gamma_{norm}$. Unpolarized gated FTIR measurements are reported normalized to the zero gate voltage and Fermi level $E_F$. However, the differential optical model that is insensitive to the raw data at the highest negative gate voltages.

The cyclotron resonance transmission is given by $T^+[V_g] = t^+[T]$. The FTIR transmission spectra, although unpolarized, is given by the same expression with $B = 0$ and $\sigma^+$ replaced by the longitudinal polarization expression $\sigma_{xx}$. The Faraday expression is given by $\theta_F[V_g] = i\frac{\alpha}{\pi}$. For the differentially gated measurements, $\Delta$-CR is given by

$$\frac{\Delta T^+}{T_{avg}} = \frac{T^+[V_g + \Delta V/2] - T^+[V_g - \Delta V/2]}{\frac{1}{2}[T^+[V_g + \Delta V/2] + T^+[V_g - \Delta V/2]]}$$

Unpolarized gated FTIR measurements are reported normalized to the zero gate transmittance and is given by (where $B$ is set to zero) $T^+[V_g]$, $\Delta T^+$, $\Delta \theta_F$ is given by $\Delta \theta_F[V_g] = \theta_F[V_g + \Delta V/2] - \theta_F[V_g - \Delta V/2]$.

There are two prominent phonons in Bi$_2$Se$_3$ over our spectral range of interest which can be written in terms of the complex index of refraction $n_{Bi_2Se_3} = \sqrt{\varepsilon}$ where

$$\varepsilon = \varepsilon_0 + \sum_{i=1}^{2} \varepsilon_i \quad \text{and} \quad \varepsilon_i = \frac{\omega_i^2}{\omega_0^2 - \omega_0^2 - i \omega_0 \gamma_i}.$$  

The phonon parameters for epitaxial films are experimentally determined to be $\varepsilon_0 = 25.6$, $\Omega_1 = 615 \text{ cm}^{-1}$, $\omega_1 = 63.1 \text{ cm}^{-1}$, $\Gamma_1 = 1.8 \text{ cm}^{-1}$, $\Omega_2 = 80 \text{ cm}^{-1}$, $\omega_2 = 133 \text{ cm}^{-1}$, and $\Gamma_2 = 2 \text{ cm}^{-1}$. Similar values for bulk crystals were previously reported. The free carrier response of the Bi$_2$Se$_3$ film is modeled as the sum of three Drude terms $\sigma^\pm = \sum_{i=1}^{3} \frac{\sigma_{ps,i}^2}{\varepsilon_0 - \varepsilon - \sigma_{ps,i}^2} + \frac{\gamma_i}{\varepsilon_0 - \varepsilon - \sigma_{ps,i}^2}$, where $\sigma_{ps,i} = eB/m(c,i)$ is the cyclotron frequency, $\sigma_{ps,i}^2 = Z_{ps}e^2/m(c,i)$ is the two dimensional plasma frequency, $\gamma_i$ is the inverse transport lifetime, $m(c,i)$ is the cyclotron mass, $n_i$ is the two dimensional carrier density, and $Z_0$ is the impedance of free space.

The parameters for the Drude term labelled BSAL (bottom surface accumulation layer) in the main text is found to be $n_{BSAL} = 1 \times 10^{13} \text{ cm}^{-2}$, $\gamma_{BSAL} = 50 \text{ cm}^{-1}$, and $m_{BSAL} = 0.25 m_0$ where $m_0$ is the mass of a free electron. For the Drude term labelled TSS (top surface topological state), $n_{TSS}$ and $\gamma_{TSS}$ are set equal to the values based upon ARPES digitized TSS dispersion such that $n_{TSS} = k_F^2/(4\pi)$ and $\gamma_{TSS} = h\gamma_F/v_F$ where $k_F$ and $v_F$ are functions of $E_F$. $E_F$ is a function of gate voltage determined by the Thomas-Fermi screening model. The relation between gate voltage $V_g$ and Fermi level $E_F$ is severely constrained by the small mass observed in the raw data at the highest negative gate voltages. The TSS scattering rate $\gamma_{TSS}$ is a free parameter which is measured.

The surface admittance associated with the carriers labelled IB (interim bulk) is assumed to be much smaller than the BSAL contribution in this experiment. In this case, the value chosen for the carrier density associated with the IB carriers at the Dirac point is insignificant in the overall differential optical response. Only the change in carrier density is relevant. We therefore assume for convenience that $n_{IB} = 0$ at the Dirac point. All changes in carrier density of the IB carriers are then completely determined by the band bending model. The scattering rate $\gamma_{IB}$ and cyclotron mass $m_{IB}$ are free parameters.

NiCr films are used as a gate and antireflection (AR) coating since NiCr has a very high scattering rate. Therefore, the admittance is frequency independent at THz frequencies. The optical response is then completely characterized by the dc sheet resistance where $R_{AR} = 275 \Omega$, and $R_g = 400 \Omega$. Other parameters are the index of refraction of sapphire $n_{sapphire} = 3.1$ and vacuum $n_0 = 1$, the thickness of the sapphire substrate $d_{sapphire} = 0.05 \text{ cm}$, and the Bi$_2$Se$_3$ film thickness $d_{BiSe_3} = 60 \text{ nm}$.

The amount of band bending at zero gate voltage associated with defects can be conceptualized as caused by fixed surface charge. It is a free parameter, but highly constrained by the observed small mass at the highest negative gate voltage combined with knowing the amount of charge transferred from a gate into the surface state, and is found to be $2.8 \times 10^{12} e cm^{-2}$ for the top surface. It is not a priori obvious that three Drude terms should suffice in reproducing the data since numerous conductivity contributions could be associated with bulk and accumulated carriers. For the BSAL, there exists many subband states, and the lowest subbands are likely Rashba split. Although the mass of the IB carriers (0.21) is expected to be between the conduction band edge mass (0.15-0.22) and the BSAL mass (0.25), it is quite large and likely a result of a convolution involving an average over bulk carriers and low binding energy BSAL states. However, the differential optical model that is insensitive to
the BSAL parameters impressively reproduces the data sets with effectively two Drude terms allowing clean extraction of the TSS parameters.

III. GATED OPTICAL DATA ADDENDA AND OPTICAL MODEL DISCUSSION

Gated optical data addenda

Figures S2(a,b) report ∆-CR data that are similar to Figure 2(a) except the measurements were performed at different frequencies. Figures S2(d,e) are the modeled optical responses that use the same parameters as reported in Figure 3.

Differential Fourier transform spectroscopy data (∆-FTIR) and modeled response are reported in Figures S2(c) and (f). The oscillations present in the data are from Fabry-Perot interference occurring in the substrate due to an imperfect antireflection coating. The increase in transmission with negative gate voltage is evidence of dominant n-type carriers. If a single Drude n-type carrier existed and a gate changed only the carrier density, then the data set would be fully symmetric about zero gate bias. The asymmetry is an indication of surface carrier contributions changing scattering rate and/or mass.

A 7 mm spot size was used for the FTIR measurements. Such a large spot size relative to the sample allows the maximum of throughput power required to measure to low frequencies. However, some leakage light occurred through un gated parts of the film due to the shadow masking procedure. The measured ∆-FTIR signals are therefore reduced. Although the photometrics differ from the model, the qualitative behavior is reproduced. The apparent asymmetric response observed in the transmission associated with the phonon resonance in Figures S2(c) and S2(c) is reproduced by a simple Lorentzian dielectric response function and requires no other exotic explanation.

Intuitive description of Figure 2(d), ∆-CR model

The modeled optical signals in relation to the Drude parameters can be qualitatively understood most easily by considering the ∆-CR model curves of Figure 2(d). A CR is a Lorentzian-like absorption where \( n \) defines the weight, \( \gamma \) defines the width, and \( m_c \) defines the center of the resonance via \( B_{res} = (\epsilon \omega/e)m_c \). When a gate voltage is applied, these parameters can change. A single ∆-CR curve is the difference between pairs of Lorentzians, a pair for the TSS and IB contributions. It is convenient to ignore the BSAL completely since it does not change with gate voltage and therefore does not contribute to the differential response. Also note that the surface admittance associated with the BSAL contribution is much larger than the IB and TSS contribution, so the overall modeled differential optical signals are insensitive to the precise values of the BSAL parameters.

Near zero gate voltage, the mass and scattering rate of the TSS and IB carriers do not change much with gate voltage. Only the weights of the resonances change. Since \( \gamma_{TSS} \gg \gamma_{IB} \) and the change in carrier density \( \Delta n_{TSS} \approx \Delta n_{IB} \) as shown in Figure 3, the resulting line shape appears Lorentzian centered on the mass of the IB contribution. As the gate is increased and the Fermi level moves higher, the dominant response is the IB contribution that has a growing scattering rate and carrier density. Taking the difference between two Lorentzians that have slightly different widths (\( \gamma \)) will cause a suppression of the peak at the resonant B-field. A slight simultaneous increase in mass shifts one of the resonances in B-field relative to the other causing a slight skew in the difference curve. In this case, the part of the differential cyclotron resonance occurring at low B-fields are slightly more suppressed than the higher fields due to the changing IB mass. The gradual increase of the TSS scattering rate which is relatively large causes a much wider general suppression, most notably giving rise to the subtle suppression of the tails at large negative B-fields.

As one gates from zero voltage toward the Dirac point, only the carrier density of the IB contribution changes. Therefore, the ∆-CR of the IB carriers maintain an approximately constant Lorentzian-like line shape. The TSS response is superimposed on the IB background. A rapid decrease of the mass in the TSS contribution in combination with the rapidly diminishing spectral weight give rise to the peak-dip-hump structure. The change in scattering rate amplify these features.

At the highest negative gate voltages, the ∆-CR peak for the TSS becomes just the CR peak of the higher Fermi level since very little relative spectral weight exists in the lower Fermi level peak. This is most vividly illustrated by considering the hypothetical scenario where the lowest Fermi level is actually at the Dirac point where there are no carriers. The difference curve then would be exactly the TSS CR curve at the higher Fermi level, superimposed on the IB contribution. Therefore, the peak-dip-hump structure evolves to a single peak structure at the two highest negative gate voltages.

An understanding of all the ∆-CR curves can be summarized. The IB contribution is the dominant response at positive gate voltages. At negative voltages, the IB contribution to ∆-CR is roughly constant since only \( n \) is changing. The peak-dip-hump structure that develops at large negative gate voltages is due to the rapidly changing mass amplified by the changing scattering rate of the TSS. At the highest negative gate voltages, a single TSS cyclotron resonance is superimposed on a constant ∆-CR IB Lorentzian-like peak.
FIG. S2: Gated optical transmission measurements addenda (a-b) 10K ∆-CR measurements are the same as described in the caption of Figure 2 except measured at different frequencies labeled on each graph (c) 6 K zero magnetic field FTIR transmission measured at gate voltage $V_g$ normalized to the zero gate value. (d-f) the corresponding modeled optical responses using the same parameters reported in Figure 3 that reproduce the data of Figures 2(a-c) and Figures 1(c,d).

IV. TOPOLOGICAL INTERFACE STATES

The interface between two insulating materials with band gaps inverted with respect to each other can support topologically protected interface states. For the ARPES measurements on bulk topological insulating materials the vacuum takes the part of the topologically trivial insulator. The more general case has been solved within the two band model using $k \cdot p$ perturbation theory in reference 14. This theory shows that the Dirac spectrum is modified as a function of the gap of the trivial insulator and any potential step at the interface. Thus for a trivial insulator with a gap of 1.2 eV in contact with a topological insulator with a bulk gap of 300 meV in the presence of a potential step of 0.3 eV the Dirac point of the topological surface state shifts by 60 meV. This potential step can result from differences in the work function of the two materials or the presence of a dipole layer at the interface. While not enough is known about the In$_2$Se$_3$/Bi$_2$Se$_3$ interface to predict the potential step exactly, this calculation demonstrates that a shift of the right magnitude is expected to occur with reasonable values of a potential step. A more accurate theory together with experimental determination of the potential step is needed to confirm this result.