Magneto-optical and magnetoelectric effects of topological insulators in quantizing magnetic fields

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We develop a theory of the magneto-optical and magnetoelectric properties of a topological insulator thin film in the presence of a quantizing external magnetic field. We find that low-frequency magneto-optical properties depend only on the sum of top and bottom surface Dirac-cone filling factors $\nu_T$ and $\nu_B$, whereas the low-frequency magnetoelectric response depends only on the difference. The Faraday rotation is quantized in integer multiples of the fine structure constant and the Kerr effect exhibits a $\pi/2$ rotation. Strongly enhanced cyclotron-resonance features appear at higher frequencies that are sensitive to the filling factors of both surfaces. When the product of the bulk conductivity and the film thickness in $e^2/h$ units is small compared to $\alpha$, magneto-optical properties are only weakly dependent on accidental doping in the interior of the film.

I. INTRODUCTION

Topological insulators (TIs) are a recently identified new class of materials. Three-dimensional TIs have insulating bulks and metallic surfaces with an odd number of Dirac cones that are responsible for most unique TI properties. Angle-resolved photoemission spectroscopy experiments have established that several strongly spin-orbit coupled materials exhibit TI properties. In this Rapid Communication, we develop a theory of the magneto-optical and magnetoelectric properties of TI thin films in the presence of a perpendicular external magnetic field.

Our work is motivated in part by potential advantages of magneto-optical over transport characterization in isolating TI surface properties from bulk contamination due to unintended doping. Since Landau-level (LL) quantization of the TI’s surface Dirac cones has recently been established by scanning tunneling microscope experiments, it should be possible to detect surface quantum Hall effects optically, even when parallel bulk conduction is present. In the quantum Hall regime, we find that the low-frequency Faraday effect is quantized in integer multiples of the fine structure constant while the Kerr effect displays the same giant $\pi/2$ rotation relative to the incident polarization direction that appears when time reversal is broken by exchange coupling. At higher frequencies, we find strong cyclotron-resonance features in both Faraday and Kerr spectra.

One goal of our work is to clarify how the magnetoelectric effects peculiar to TIs (Refs. 5–9) are reflected in their thin-film magneto-optical properties. We show that low-frequency TI magneto-optical response in the quantum Hall regime depends on the sum of top and bottom surface filling factors whereas the magnetoelectric response of film polarization to an external magnetic field depends on the filling factor difference. We argue that coupling between electric and magnetic fields in the presence of a TI material is most usefully regarded as a property of its surfaces, not of its bulk.

II. LOW-FREQUENCY MAGNETOELECTRIC AND MAGNETO-OPTICAL RESPONSE

The response of a TI thin film to an external magnetic field is dominated by its Dirac-cone surface states. When the Dirac cone’s quantum Hall effect is well developed, its Hall conductivity $\sigma_{xy}$ has quantized plateau values with half-odd-integer values in $e^2/h$ units. The longitudinal resistivity vanishes on the Hall plateaus but is nonzero on the risers between Hall plateaus. The risers between the $(n -1/2)(e^2/h)$ and $(n +1/2)(e^2/h)$ plateaus occur near integer values of the filling factor $n = n_B \in \mathbb{Z}$. The Dirac cone’s quantum Hall effect has so far been cleanly observed in two-dimensional graphene, in which four separate Dirac cones conduct in parallel.

The Streda formula

$$\sigma_{xy} = e c (\partial N / \partial B) = e c (\partial M / \partial \mu),$$

which is valid at Hall plateau centers, implies a relationship between surface conductivities and the magnetoelectric response of a TI thin film. In Eq. (1) $N$ is the two-dimensional electron density, $B$ the external magnetic field, $M$ the orbital magnetization, and $\mu$ the chemical potential. We define the electronic polarization $P$ per unit volume of a TI thin film in terms of the difference between the surface charge densities accumulated on the top $(T)$ and bottom $(B)$ surfaces $P = e(N_T - N_B)/2$. It follows that for plateaus characterized by $\nu_T$ and $\nu_B$ the magnetoelectric susceptibility

$$\chi_{ME} = 4 \pi \sigma_{xy} / B = (\nu_T - \nu_B) \alpha,$$

depends only on the filling factor difference between surfaces ($\alpha = e^2/h$ is the fine structure constant). Recent experiments have demonstrated the possibility of tuning the surface carrier densities systematically by surface doping or gating. At a fixed magnetic field, the top and bottom surfaces need not be on the same Hall plateau. Provided that the bulk resistance is sufficiently large, $\chi_{ME}$ can be measured by contacting top and bottom surfaces separately and detecting voltages induced by magnetic field variation. If sample quality can be improved sufficiently, TI thin films might be useful as magnetic field sensors.

The TI’s surface quantum Hall effect can also be probed by Faraday and Kerr angle measurements. In the low-frequency regime the electromagnetic wavelength is much longer than the film thickness. Faraday’s and Ampère’s laws...
then imply that the electric field is spatially constant across the film, while the magnetic field jumps by a value proportional to the current integrated across the TI film. Allowing for a bulk conductivity \( \Sigma \) due to unintended doping and assuming a small bulk Hall angle we find that the low-frequency quantum Hall regime Faraday and Kerr angles depend on the TI surface properties only through \( \nu_T + \nu_B \):

\[
\tan \theta_F = \frac{(\nu_T + \nu_B) \alpha}{1 + 2\pi \Sigma d/c},
\]

\[
\tan \theta_K = \frac{4(\nu_T + \nu_B) \alpha}{1 - (1 + 4\pi \Sigma d/c)^2 - [2(\nu_T + \nu_B) \alpha]^2}.
\]

The bulk carriers enter as an effective longitudinal surface conductivity \( \Sigma d \), where \( d \) is the film thickness. When the bulk conductivity is sufficiently small so that \( \Sigma d/(e^2/h) \ll \alpha \), the Kerr angle exhibits a universal full-quarter rotation \( \theta_K = \pm \pi/2 \). When \( \Sigma d/(e^2/h) \ll 1/\alpha \), the Faraday angle is quantized in integer multiples of the fine structure constant

\[
\theta_F = (\nu_T + \nu_B) \alpha.
\]

Given the rapid progress in TI film quality, these regimes should be within reach experimentally; in particular, the less stringent condition for Faraday angle quantization should be currently accessible. Since the magnetoelectric polarizability Eq. (2) yields the filling factor difference and the Faraday angle Eq. (5) yields the filling factor sum, measurement of both quantities could allow the filling factors \( \nu_T, \nu_B \) to be extracted individually.

### III. DIRAC-CONE AC CONDUCTIVITY

Outside of the long-wavelength limit, TI thin-film magneto-optical properties depend on the finite-frequency Dirac-cone conductivity which we now evaluate microscopically. The high-frequency signal consists of resonances at inter-LL transition frequencies. We neglect optical phonon contributions to the conductivity which are not expected to be significantly dependent on magnetic field strength. In an external magnetic field the Dirac-cone Hamiltonians for the top and bottom surfaces are \( H = (-1)^{s} (i\tau \cdot \nabla + eA/c + V/2) + \Delta \tau_z, \) where \( \tau \) is the spin Pauli-matrix vector, \( A = (0, Bx) \) is the vector potential, \( \Delta = g_{\mu B} B/2 \) is the Zeeman coupling, \( V \) accounts for a possible potential difference between top and bottom surfaces due to doping or external gates, and \( L = 0, 1 \) for the top (0) and bottom (1) surfaces. The LLs are labeled by integers \( n \) and for \( n \neq 0 \) have eigenenergies [relative to the Dirac point energies \( (-1)^{s} V/2 \)]

\[
\epsilon_n = \text{sgn}(n) \sqrt{2n^2} |n|/\ell_B^2 + \Delta^2,
\]

where \( \ell_B = |c/eB| \) is the magnetic length. In the \( n = 0 \) LL spins are aligned with the perpendicular field and \( \epsilon_0 = -\Delta \). For convenience we rewrite the LL index as \( n = sm \), where \( m = 0, 1, 2, \ldots, N_c \) and \( s = \pm 1 \) for electronlike and holelike LLs. \( N_c = \ell_B^2/(2e^2/c^2) \) is the largest LL index with an energy smaller than the ultraviolet cutoff \( e_c \). We choose \( e_c = E_g/2 \), where \( E_g \) is the bulk band gap.

Using the Kubo formalism we find that in the quantum Hall regime \( (\Omega_\theta \tau \gg 1) \), where \( 1/\tau \) the quasiparticle lifetime broadening and \( \Omega_B = c/\ell_B \) is a characteristic frequency typical of the LLs spacing) the conductivity in \( e^2/h = \alpha c \) units is given by

\[
\sigma_{\alpha\beta}(\omega) = \frac{e^2}{2\pi f_B^2} \sum_{m=0}^{N_c-1} \sum_{s,s'=\pm 1} \frac{f_{sm} - f_{s'(m+1)}}{\epsilon_{sm} - \epsilon_{s'(m+1)}} \Gamma_{\alpha\beta} \left( m, \omega \right).
\]

Here \( \alpha, \beta = \{x, y\}, f_{sm} \) is a Fermi factor, and

\[
\Gamma_{\alpha\beta} \left( m, \omega \right) = \frac{1}{\omega - e_{sm} + e_{s'(m+1)} + i/2\tau} \pm \frac{1}{\omega + e_{sm} - e_{s'(m+1)} + i/2\tau}.
\]

In Eq. (7), the LL eigenstates are \( C_{i|0} = 0, C_{|0} = 1/\sqrt{2} \), and for \( m \neq 0 \) \( C_{|s} = \sqrt{e_m + s\Sigma}/\sqrt{2e_m} \), and \( C_{|s} = \sqrt{e_m - s\Sigma}/\sqrt{2e_m} \). Equations (6) and (7) express \( \sigma_{\alpha\beta} \) as a sum over interband and intraband dipole-allowed transitions which satisfy \( |n' - n| = \pm 1 \). In the \( \omega = 0 \) limit Eq. (6) yields correct half-quantized plateau values for the Hall conductivity.

### IV. TI THIN FILM CYCLOTRON RESONANCE

Resonances at transition energies are responsible for dips and peaks in transmission and reflection spectra, respectively, and give rise to associated characteristic features in Faraday and Kerr spectra. Figure 1(a) shows the frequency dependence of the Faraday rotation for several different filling factors. These results are obtained by combining the ac

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low-frequency giant Kerr effect, the resonant peaks which correspond to absorptive LL transitions are nonuniversal. We note that Zeeman coupling shifts the zeroth LL away from the Dirac point and therefore breaks the electron-hole (e-h) symmetry of the LL spectrum. This implies that the \( n = -1 \rightarrow 0 \) and \( 0 \rightarrow 1 \) transition frequencies, \( \omega_{n-1} = \frac{\sqrt{2}v^2}{l_B^2} + \Delta^2 + \Delta \) and \( \omega_{1-0} = \frac{\sqrt{2}v^2}{l_B^2} + \Delta^2 - \Delta \), become different. In general, \( \theta_K \) is an odd function of filling factor \( \nu \) if e-h symmetry is intact. When the surface filling factors \( \nu_T = \nu_H = 1/2 \) are opposite, e-h asymmetry implies that cyclotron-resonance peaks of opposite sign appear in \( \theta_K \) at the two transition frequencies. The Kerr effect thus provides a smoking gun to detect the absence of e-h symmetry.

In Fig. 2(a) we plot the Kerr angle \( \theta_K \) as a function of frequency and magnetic field for fixed surface carrier densities. As in the exchange coupling case,\(^7\) the giant Kerr effect survives up to a relatively large frequency which we refer to as the Kerr frequency,

\[
\omega_K = \frac{2\pi \sigma_{xx}^0(0)/c}{[(\varepsilon - \mu) + (\varepsilon - \mu_s)d_s] - 2\pi \sigma_{xx}^0(0)/c}.
\]

Here \( \sigma_{xx}, \sigma_{yy} \) are the total surface conductivities, \( \varepsilon, \mu, d \) are the dielectric constant, permeability (\( = 1 \)), and thickness of the TI film, \( \varepsilon, \mu_s, d_s \) are the corresponding substrate values and \( ' \) in \( \sigma_{xx}^0 \) denotes a frequency derivative. It follows from Eq. (8) that \( \omega_K \) is inversely proportional to the optical thickness of the TI film and the substrate. Figure 2(b) illustrates the effect of varying \( B \) on \( \omega_K \). There exists an optimal value of \( B \) for which \( \omega_K \) is in the terahertz range. High \( \omega_K \) values are most readily achieved on “low-\( \kappa \)” substrate materials such as SiO\(_2\) or using free-standing films suitable for optical studies.\(^14\) \( \omega_K \) can also be increased by surface doping.

**V. DISCUSSION**

TIs have interesting magnetoelectric and magneto-optical properties when time-reversal symmetry (TRS) is broken to open up a gap in its Dirac-cone surface states. TRS can in principle be broken by exchange coupling to an insulating ferromagnet, although it is not yet established that a sufficiently strong coupling can be achieved in practice. The circumstance discussed here in which a perpendicular magnetic field is applied to a TI thin film provides an experimentally simpler and phenomenologically richer method for producing TI thin films with weak TRS breaking. The special case in which the filling factor \( \nu = 1/2 \) on both surfaces yields the same surface Hall conductivities as in the exchange-coupled case and therefore the same\(^5\) low-frequency Kerr and Faraday angle results. In the magnetic field case, however, strong magneto-optical and magnetoelectric effects occur over a broad range of separately controllable carrier densities on both surfaces. The top and bottom surface Dirac-cone filling factors, \( \nu_T \) and \( \nu_H \), can be identified\(^15\) from the ratios of magneto-optical resonance frequencies and from the patterns they produce in Faraday or Kerr spectra. Bulk conduction has a quantitative influence on Faraday and Kerr spectra only.
when $\Sigma d/\epsilon^2 h$, the bulk contribution to the dimensionless effective surface conductivity, is larger than about $1/\alpha$ and $\alpha$, respectively. In the quantum Hall regime, the Kerr angle is large whenever the filling factors sum to a nonzero value and are away from one of the integer values at which longitudinal resistance peaks occur. On quantum Hall plateaus magneto-optical properties depend only on the sum of individual surface filling factors, whereas the magnetoelectric response of film polarization to field depends only on the difference.

The influence of a bulk TI material on electromagnetic fields can be represented by introducing an $E \cdot B$ term with coefficient $\alpha_{\text{ME}}$ in the electromagnetic Lagrangian. The microscopic Poisson and Ampère equations then take \cite{16,17} the form

$$\nabla \cdot E = 4\pi\rho - \nabla \alpha_{\text{ME}} \cdot B,$$

$$\nabla \times B - (1/c) \partial E/\partial t = (4\pi/c)J + \nabla \alpha_{\text{ME}} \times E.$$ \hspace{1cm} (9)

When both surfaces are on Hall plateaus, Eq. (9) with $\rho$ and $J$ set to zero is a valid coarse-grained description of the interface provided that we set

$$d\alpha_{\text{ME}}/dz = (4\pi/c)\alpha_{xy}\delta(z) = 2\alpha v\delta(z).$$ \hspace{1cm} (10)

Because $\alpha$ can change by an integer without altering the bulk, only $\alpha_{\text{ME}}$ modulo $2\alpha$ characterizes the bulk material. (When the bulk $\alpha_{\text{ME}}$ is mapped to the interval $[0,2\alpha]$, only $\alpha_{\text{ME}} = \alpha$ is consistent with time-reversal invariance.) In the thin film geometry normally employed in experiment, however, a bulk value of $\alpha_{\text{ME}}$ modulo $2\alpha$ is not sufficient to predict the result of a magneto-optical or magnetoelectric measurement \cite{18} because it does not specify the Hall plateau index. Moreover, in real samples with nonzero disorder the Dirac-cone surface Hall conductivities will be strongly suppressed when the external magnetic field is weak. The values of $\alpha_{\text{ME}}$ required to correctly reproduce the low-frequency magneto-optical signal will therefore become small and vanish with the external magnetic field. In the stronger-field quantum Hall regime, the appropriate magnetoelectric constant for vacuum, TI thin film, and substrate regions can be obtained by integrating Eq. (10) to obtain $\alpha_{\text{ME}} = 0$ in the upper vacuum, $\alpha_{\text{ME}} = 2\alpha v_T$ in the TI thin film, and $\alpha_{\text{ME}} = 2\alpha(v_T + v_S)$ in the substrate and lower vacuum. In both cases the appropriate values of $\alpha_{\text{ME}}$ depend critically on surface properties. Although $\alpha_{\text{ME}} \bmod (2\alpha)$ is indeed a bulk property of a disorder-free TI, its value does not normally provide enough information to predict either magneto-optical properties or magnetoelectric response.

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10. The influence of nonmagnetic matter on electromagnetic waves is completely captured by its frequency-dependent conductivity tensor. At frequencies well below the TI gap an external magnetic field has a negligible influence on the bulk conductivity. When carriers are present in the bulk due to unintended doping, magnetic field dependence is still expected to be weak when disorder is strong.


17. This form for the electromagnetic Lagrangian captures matter effects only in the adiabatic dissipationless limit.

18. In addition, the coupling $\alpha_{\text{ME}}$ is equivalent to the magnetoelectric susceptibility $\chi_{\text{ME}}$, only for TI geometries with one simply connected surface when $\chi_{\text{ME}} = 2\delta_{\text{surface}}\alpha$ such as a sphere or semi-infinite slab, but not for thin film.