Magneto-optical and Magneto-electric Effects of Topological Insulators in Quantizing Magnetic Fields

Wang-Kong Tse and A. H. MacDonald
Department of Physics, University of Texas, Austin, Texas 78712, USA

We develop a theory of the magneto-optical and magneto-electric 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 magneto-electric 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.

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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 (ARPES) experiments have established that several strongly spin-orbit coupled materials \(^2\) exhibit TI properties. In this paper we develop a theory of the magneto-optical and magneto-electric 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 \(^2\) 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 STM experiments \(^3\), it should be possible to detect surface quantum Hall effects optically, even when parallel bulk conduction is in 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 \( \pi/2 \) 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 magneto-electric effects peculiar to TIs \(^3\) 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 magneto-electric 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.

Low-frequency Magneto-electric 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 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 non-zero 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 \( \nu = n \in \mathbb{Z} \). The Dirac cone’s quantum Hall effect has so far been observed \(^1\) only in two-dimensional graphene, in which four separate Dirac cones conduct in parallel.

The Streda \(^12\) formula,

\[
\sigma_{xy} = ec(\partial N/\partial B) = ec(\partial M/\partial \mu),
\]

which is valid at Hall plateau centers, implies a relationship between surface conductivities and the magneto-electric 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 electric 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 magneto-electric susceptibility

\[
\chi_{ME} = 4\pi \partial P/\partial B = (\nu_T - \nu_B)\alpha,
\]

depends only on the filling factor difference between surfaces \( \alpha = e^2/hc \) is the fine structure constant. Recent experiments have demonstrated \(^14\) 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.
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 magneto-electric 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 \( \nu_{T,B} \) to be extracted individually.

**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 (T) and bottom (B) surfaces are

\[
H = \langle -1 \rangle B \left( \nu \tau \cdot \left( -i \nabla + e \mathbf{A}/c \right) + V/2 \right) + \Delta \tau_z,
\]

where \( \mathbf{A} \) is the spin Pauli matrix vector, \( \mathbf{A} = (0, Bx) \) 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 \( ( -1 )^{L/2} E / 2 \))

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

where \( \ell_B = \sqrt{c |e| B} \) is the magnetic length. In the \( n = 0 \) LL spins are aligned with the perpendicular field and \( \varepsilon_0 = -\Delta \). For convenience we rewrite the LL index as \( n = s m \), where \( m = 0, 1, 2, \cdots N_c \) and \( s = \pm 1 \) for electron-like and hole-like LLs. \( N_c \approx \ell_B^2 (\varepsilon_c^2 - \Delta^2)/2e^2 \) is the largest LL index with an energy smaller than the ultraviolet cut-off \( \varepsilon_c \). We choose \( \varepsilon_c = E_g/2 \) where \( E_g \) is the bulk band gap.

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

\[
\sigma_{\alpha\beta}(\omega) = \frac{\omega^2}{2\pi\ell_B^2} \sum_{s,s'}^N \left| \Gamma_{ss'}^{\alpha\beta} (m, \omega) \right|^2 \left| \mathcal{C}_0^{s}(m, \omega) \right|^2 \left| \mathcal{C}_1^{s}(m, \omega) \right|^2 \left| \mathcal{C}_0^{s'}(m, \omega) \right|^2 \left| \mathcal{C}_1^{s'}(m, \omega) \right|^2 \left( \frac{1}{\omega - \varepsilon_{sm} - \varepsilon_{s'm} + i/2\tau} - \frac{1}{\omega + \varepsilon_{sm} - \varepsilon_{s'm} + i/2\tau} \right).
\]

In Eq. (7), the LL eigenspinors are \( \mathcal{C}_0^s = 0 \), \( \mathcal{C}_{10} = \sqrt{2} \), and for \( m \neq 0 \) \( \mathcal{C}_{1sm} = s \sqrt{\varepsilon_m + \Delta^2} \sqrt{\varepsilon_m + \Delta^2} \), and \( \mathcal{C}_{1sm} = \sqrt{\varepsilon_m - \Delta^2} \sqrt{\varepsilon_m - \Delta^2} \). Eqs. (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.

**FIG. 1:** (Color online). (a) Faraday rotation \( \theta_F \) versus frequency \( \omega/\varepsilon_c \) at equal filling factors on both surfaces \( \nu = 1/2 \) (green), \( \nu = 1 \) (red), \( \nu = 3/2 \) (blue), and \( \nu = 2 \) (black). The densities on both surfaces are \( N_{T,B} = 5 \times 10^{11} \text{ cm}^{-2} \). For this density the filling factor is given by \( \nu = 20.81/B \) [Tesla]. We choose a 30 nm-thick Bi2Se3 film as a prime example of TIs with a large bulk band gap \( E_g = 0.35 \text{ eV} \). The Fermi velocity is \( v = 5 \times 10^5 \text{ cm s}^{-1} \) and the dielectric constant \( \varepsilon = 29 \). (b) Kerr rotation \( \theta_K \) versus frequency at the same filling factors.
Fig. 2b illustrates the effect of varying $B$ to the optical thickness of the TI film and the substrate. It follows from Eq. (8) that 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 like SiO$_2$ or using free-standing films suitable for optical studies [14]. $\omega_K$ can also be increased by surface doping.

Discussion—TI's have interesting magneto-electric 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 $\nu = 1/2$ 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. 2a, 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 [5], the giant Kerr effect survives up to a relatively large frequency $\nu_K = 2 \times 10^{11}$ cm$^{-2}$. Cyclotron resonance features corresponding to particular transitions are allowed when the initial Landau level is at least partially occupied and the final Landau level is at least partially empty. In the weak-field semiclassical transport regime, $\sigma_{xx} \propto B$ and $\theta_K$ vanishes as $B \to 0$. (b) Kerr frequency $\omega_K$ (see text) as a function of magnetic field for $N_{T,B} = 10^{12}$ cm$^{-2}$ (dark/black) for free-standing 30 nm-thick Bi$_2$Se$_3$ ($\epsilon_s = 1$), with SiO$_2$ substrate ($\epsilon_s = 4$), and Si substrate ($\epsilon_s = 12$). Grey/red line shows the free-standing case with $5 \times 10^{11}$ cm$^{-2}$. The substrate thickness $d_s = 1 \mu$m.
Dirac cone filling factors, $\nu_T$ and $\nu_B$, 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 $\Sigma d/(e^2/h)$, 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 non-zero 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 magneto-electric response of film polarization to field depends only on the difference.

The influence of a bulk TI material on electromagnetic fields can be represented \[6\] by introducing an $\mathbf{E} \cdot \mathbf{B}$ term with coefficient $\alpha_{\rm ME}$ in the electromagnetic Lagrangian. The microscopic Poisson and Ampère equations then take \[16\] \[17\] the form

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

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

When both surfaces are on Hall plateaus, Eqs. \[7\] with $\rho$ and $\mathbf{J}$ set to zero is a valid course-grained description of the interface provided that we set

$$\text{d} \alpha_{\rm ME}/\text{d} z = (4\pi/c) \sigma_{xy} \delta(z) = 2\nu \delta(z). \quad (10)$$

Because $\alpha$ is $\nu$ can change by an integer without altering the bulk, only $\alpha_{\rm ME}$ modulo $2\alpha$ characterizes the bulk material. (When the bulk $\alpha_{\rm ME}$ is mapped to the interval $[0, 2\alpha]$, only $\alpha_{\rm ME} = \alpha$ is consistent with time-reversal invariance.) In the thin film geometry normally employed in experiment, however, a bulk value of $\alpha_{\rm ME}$ modulo $2\alpha$ is not sufficient to predict the result of a magneto-optical measurement \[15\] because it does not specify the Hall plateau index. Moreover, in real samples with non-zero disorder the Dirac-cone surface Hall conductivities will \[19\] be strongly suppressed when the external magnetic field is weak. The values of $\alpha_{\rm ME}$ 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 magneto-electric constant for vacuum, TI thin film, and substrate regions can be obtained by integrating Eq. \[10\] to obtain $\alpha_{\rm ME} = 0$ in the upper vacuum, $\alpha_{\rm ME} = 2\nu_T$ in the TI thin film, and $\alpha_{\rm ME} = 2\nu_T(\nu_T + \nu_B)$ in the substrate and lower vacuum. In both cases the appropriate values of $\alpha_{\rm ME}$ depend critically on surface properties. Although $\alpha_{\rm ME} \text{mod}(2\alpha)$ 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 magneto-electric response.

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\[10\] The influence of non-magnetic 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.

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