Dynamical Quantum Anomalous Hall Effect in Strong Optical Fields

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Topological insulators (TIs) are characterized by the quantum anomalous Hall effect (QAHE) on the topological surface states under time-reversal symmetry breaking. Motivated by recent experiments on the magneto-optical effects induced by the QAHE, we develop a theory for the dynamical Hall conductivity for subgap optical frequency and intense optical fields using the Keldysh-Floquet Green’s function formalism. Our theory reveals a nonlinear regime in which the Hall conductivity remains close to $e^2/2h$ at low frequencies. At higher optical fields, we find that the subsequent collapse of the half quantization is accompanied by coherent oscillations of the dynamical Hall conductivity as a function of field strength, triggered by the formation of Floquet subbands and the concomitant inter-subband transitions.

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Introduction.— Topological quantum phases of matter are one of the most intriguing paradigms in contemporary condensed matter physics. Recently, intensive researches have been focused on the interplay between topological order and dynamics. This is well exemplified by the dynamical synthesis and manipulation of topological quantum phases, which include Floquet Chern insulators, Floquet TIs, Floquet Majorana fermions, and Floquet Weyl semimetals.

Topological Hall quantization is a hallmark signature of two-dimensional (2D) quantum anomalous Hall insulators. The crucial ingredients to realize the quantum anomalous Hall state are strong spin-orbit coupling and a magnetic Zeeman gap $\Delta$. With magnetic doping, these can be realized on the TI surface, single-layer or bilayer graphene, and HgTe quantum well.

Under a D.C. bias, a robust Hall plateau corresponding to the quantized Hall value has already been observed in magnetically doped TI films. While topological transport as a property of linear response has been largely focused on the high-frequency, off-resonance regime; the effects of low-frequency A.C. driving in the adiabatic regime, where the frequency is small compared to either the bandwidth or the band gap, remain not well understood. To remedy this situation, we numerically study the Hall conductivity as a function of the optical field strength and frequency without any expansion in powers of optical field strength. This approach allows us to address the full nonperturbative effects of the optical field on the electronic bands and transport properties of the system, even at low frequencies.

For concreteness, we take the massive Dirac model describing TI surface states with broken time-reversal symmetry and QAHE as our prototypical system of investigation. We expect the conclusions from our theory to be broadly applicable to other materials that host massive Dirac fermions such as Chern insulators and graphene-like materials with broken spatial-inversion symmetry.

2D massive Dirac fermions and QAHE.— We consider the quantum anomalous Hall state realized on the surface of a TI film with broken time-reversal symmetry, e.g., by interfacing with a magnetic substrate or by doping with magnetic adatoms. The film is sufficiently thick so that tunneling between the top and bottom surface states can be ignored. Focusing on a single surface, a low-energy electron at the $\Gamma$ point in the Brillouin zone is described by the 2D massive Dirac Hamiltonian $H = \sum_k \psi_k^{\dagger} H_k \psi_k$ with $H_k = d_k \cdot \tau$. Here, $\psi_k = (c_{k \uparrow}, c_{k \downarrow})^T$ with $c_{k \alpha}$, the electron annihilation operator with momentum $\hbar k$ and spin $\alpha(=\uparrow, \downarrow)$, $\tau = (\tau_x, \tau_y, \tau_z)$ consists of the...
Pauli matrices, and $d_k = (v h k_x, v h k_y, m_0)$ with $v$ being the Dirac velocity. In the presence of the Dirac mass $m_0$, or equivalently, the band gap $\Delta = 2m_0$, which is generated by the exchange field due to the magnetic substrate or adatoms, the bulk energy spectrum for the surface state becomes insulating with the energy dispersion $\varepsilon_k = |d_k| = [(v h)^2(k_x^2 + k_y^2) + m_0^2]^{1/2}$ for the conduction band and $-\varepsilon_k$ for the valence band. In the presence of a time-independent $D.C.$ electric field, 2D massive Dirac fermions give rise to the QAHE consisting of a robust one-half quantization of the Hall conductance in units of $\sigma_0 = e^2/h$ due to the half-kyrmion configuration of $d_k$ \[ \sigma_{xy} / \sigma_0 = (1/4\pi) \int d^2k \ d_k \cdot (\partial_k, d_k \times \partial_k) d_k = \text{sgn}(m_0)/2, \] where $d_k = d_k/|d_k|$, and ‘sgn’ denotes the signum function.

Dynamical response to the optical field.— We now consider linearly polarized light illuminated in the normal direction to the surface of the TI film. Choosing the polarization direction along the $z$ axis and the propagation direction along the $x$ axis, the incident light with electric field amplitude $E$ and frequency $\Omega$ is described by the vector potential $A(t) = -(cE/\Omega)\zeta(t)\sin(\Omega t)\hat{z}$. Here, a switching protocol is encoded in the function $\zeta(t) = e^{i/\tau}(\Theta(t)-\Theta(t+\tau))$, with $\tau$ being a switch-on time scale and $\Theta(x)$ the step function, which satisfies $\zeta \to 0$ for an equilibrium state at $t \to -\infty$ and $\zeta = 1$ for a nonequilibrium steady state (NESS) at $t \geq 0$. In the semiclassical treatment of the optical field, the Peierls substitution $\hbar k \to \hbar k + eA(t)/c$ in the original Hamiltonian leads to the time-dependent Hamiltonian $\hat{H}_k(t) = H_k + V(t)$, with the perturbation of the form $V(t) = -\hbar \gamma_0(\zeta(t)\sin(\Omega t)\tau_x + \gamma_0 eE\nu/\Omega)$ playing the role of Rabi frequency of the two-band system. The relative strength of the Rabi frequency to the photon energy defines the dimensionless coupling parameter $\lambda = \gamma_0/\hbar\Omega$, according to which the system is in a regime commonly classified as weak coupling ($\lambda \ll 1$) and strong coupling ($\lambda \gtrsim 1$) in quantum optics.

For the purpose of formulating a nonperturbative theory of dynamical response, we start with a generic form of the surface electric current density, $\mathbf{J}(t) = -eS^{-1} \sum_k \langle \mathbf{v}_k(t) \rangle$, where $S$ is the normalization area, $\mathbf{v}_k(t) = \psi_k^*(t) \nabla \psi_k(t)/h|\psi_k(t)|$ is the single-electron velocity operator, and $(\mathcal{M}) = \text{Tr}(\rho_0 \mathcal{M})$ is the canonical ensemble average of the operator $\mathcal{M}$ in terms of the initial density matrix $\rho_0 = e^{-H/k_B T}/\text{Tr}(e^{-H/k_B T})$, which depends on the switching protocol as discussed later. The time evolution of $\mathbf{J}(t)$ is expressed by using the lesser Green’s function, $[h\mathcal{G}^{<}_k(t,t')]_{\alpha\beta} = i\langle \hat{c}^\dagger_{k\alpha}(t) \hat{c}_{k\beta}(t) \rangle$. In this work, if we focus on a NESS, the system recovers time translational symmetry, i.e., $\hat{H}(t) = \hat{H}(t+\tau)$ with periodicity $\tau = 2\pi/\Omega$, and is thus governed by the Floquet theorem \[ \mathcal{G}^{<}(t) = \sum_{s=\pm} \mathcal{G}^{<}(t) e^{i\omega(t+\tau)/2} e^{-i\omega(t-\tau)/2}, \] where $\omega(t) = \int_{-\hbar\Omega/2}^{\hbar\Omega/2} d\omega \mathcal{G}^{<}(\omega)_{\alpha\beta;mn}/2\pi$, we find the surface current density $J_x(t) = \sum_{s=\pm} \text{Re} \left[ \mathcal{G}^{<}_{k\alpha}(t) E e^{-i\omega(t+\tau)/2} \right]_{\alpha\beta;mn}$, where the $s$-th harmonics of the field-dependent dynamical longitudinal and Hall conductivities are given by

\begin{align}
\text{Re}[\mathcal{G}^{<}_{xx}(t)] &= \text{Im}[\mathcal{G}^{<}_{x\bar{x}}(t)], \\
\text{Im}[\mathcal{G}^{<}_{yx}(t)] &= \text{Im}[\mathcal{G}^{<}_{y\bar{x}}(t)],
\end{align}

for the dissipative (incoherent) components, and

\begin{align}
\text{Re}[\mathcal{G}^{<}_{yx}(t)] &= \text{Re}[\mathcal{G}^{<}_{x\bar{x}}(t)], \\
\text{Im}[\mathcal{G}^{<}_{yx}(t)] &= -\text{Re}[\mathcal{G}^{<}_{y\bar{x}}(t)],
\end{align}

for the reactive (coherent) components, and $\mathcal{G}^{<}_{x\bar{x}}(t)$ reads

\[ \mathcal{G}^{<}_{x\bar{x}}(t) = -2E_0/E \left( \frac{v h}{\Delta} \right)^2 \sum_{\kappa \in \mathbb{Z}} \sum_{\nu \in \mathbb{Z}} \int_{-\hbar\Omega/2}^{\hbar\Omega/2} d\omega \left\{ \mathcal{G}^{<}_{\kappa\kappa}(\omega) \right\}_{\nu;\kappa+n,s,n \pm [\mathcal{G}^{<}_{\kappa\kappa}(\omega) \right\}_{\nu;\kappa+n,s,n+1} \right. \]
Green’s function can be found by numerically inverting Eq. (20). Then, the result is inserted into Eq. (4) through the relation $G_{kk}^R = (G_{kk}^R - G_{kk}^G + G_{kk}^A)/2$.

In Eq. (11), the initial NESS was not specified yet. We assume that the optical field is adiabatically switched on with the switch-on time $\tau$ long enough compared with other time scales in the system, so as to restore the condition of thermal equilibrium at $t = 0$.

Under this condition, the initial NESS is described by the equilibrium Green’s function with the components: $G_{kk}^R(\omega) = U_k \tilde{G}_{kk}^R(\omega) U_k^\dagger$, where $\tilde{G}_{kk}^R(\omega) = [\mathcal{L}^2(\mathcal{L}^{\infty + i\eta})]_{\mathcal{L}^2} - \mathcal{E}_k \tau_z \otimes \mathcal{I}_\infty$ and $\tilde{G}_{kk}^R(\omega) = [\tilde{G}_{kk}^R(\omega)]^\dagger$. $\tilde{G}_{kk}$, satisfying the fluctuation-dissipation relation [33], is given by $\tilde{G}_{kk}^R(\omega) = [\tilde{G}_{kk}^R(\omega) - \tilde{G}_{kk}^R(\omega)]_{\mathcal{L}^2} \otimes [\mathcal{I}_\infty - 2\mathcal{F}(\omega)]$. Here, $\mathcal{I}_n$ is the $n \times n$ identity matrix, $\eta$ is the band broadening width due to elastic scattering of electrons, $(\Omega)_{mn} = n\Omega\delta_{mn}$, and $(\mathcal{F}(\omega))_{mn} = \int_{R} (\omega + n\Omega)\delta_{mn}$ with $\mathcal{F}_R(\omega) = (\epsilon_{\omega + kn}^R + 1)^{-1}$. The unitary operator $U_k = (\sin(\vartheta_k/2) / \sin\varphi_k) \tau_z + \sin(\varphi_k/2) \tau_x) \otimes I_\infty$ transforms the original spin representation of $\mathcal{H}_k$ into the band representation, with $\vartheta_k = \cos^{-1}([\Delta/(2\mathcal{E}_k)])$.

**Linear-response and near-resonance regimes.**— Our formalism provides a generic framework to investigate the dynamical response of massive Dirac electrons to strong optical fields. Before applying our theory to the full non-perturbative regime, here we consider in particular (i) the linear-response and (ii) the near-resonance coherent regimes, and see whether our framework reproduces established results in these well-known limits.

First, we are able to recover the linear-response result when $\nu_0 \ll \Delta$. In this regime, Eq. (11) can be expanded analytically up to the linear order in $\nu_0$, and we recover the Kubo formula result for dynamical conductivity [21] (see Supplementary Material for details). In Fig. 1a), we also numerically confirm that the linear-response behavior of the dynamical Hall conductivity is recovered as the optical field strength is decreased (the reference plot of the Kubo formula result is indicated by the red solid line) [33]. In the low-frequency regime, we notice that the one-half Hall quantization remains robust against weak fields.

Secondly, we examine the near-resonance coherent regime, where the optical frequency is close to the band gap with the detuning $\delta = (\Delta - \Omega(0))$ satisfying $\Delta \gg \delta \gg \eta$. For the Dirac model, a theory based on the Bloch equation has been developed within the rotating wave approximation (RWA) [20] (see Supplementary Material for details). The result for the dynamical Hall conductivity from this theory is plotted in Fig. 1(b) as the red solid line. For weak fields ($E/E_0 \lesssim 0.2$), as $\eta$ decreases below $\delta$ approaching the coherent regime ($\eta \to 0$), we find close agreement between the Keldysh-Floquet and the Bloch-RWA results. This regime corresponds to weak coupling with $\lambda \ll 1$. On the other hand, for stronger fields ($E/E_0 \gtrsim 0.2$), the Keldysh-Floquet result differs noticeably from the Bloch-RWA result, because $n$-photon excitation processes ($n \geq 2$) that becomes important at strong fields are ignored in the RWA [41], but are exactly captured in the Keldysh-Floquet approach.

**Dynamical QAHE.**— Our main interest in this work is to investigate the robustness and the dynamical breakdown of the QAHE without restricting ourselves to the linear-response or near-resonance coherent regimes. Here, we focus our discussion on $\text{Re}[\sigma^{\pm \pm}_{xx}(E)]$ and $\text{Re}[\sigma^{\pm \pm}_{xx}(E)]$, which are sufficient to capture the QAHE in the low-frequency regime. Figure 2 shows the dynamical breakdown of the QAHE as a function of optical field strength for different frequencies. Both the longitudinal and Hall conductivities exhibit a consistent pattern underlain by the following two main features. First, there is a clear signature for the robustness of the half-quantized Hall regime. In Fig. 2(b), for $h\Omega/\Delta = 0.2$, low enough to capture the QAHE, the regime is robust up to a threshold optical field strength $E_{th}/E_0 \approx 0.12$. For $E < E_{th}$, the longitudinal counterpart is suppressed, and the generalized Hall angle $\theta_H$, which is defined by $\tan^{-1}[\text{Re}(\sigma^{+\pm}_{xx})/\text{Re}(\sigma^{\pm \pm}_{xx})]$, attains the maximum value $\pi/2$ [see Fig. 2(a) and (c)]. Secondly, in the regime of $E > E_{th}$, the QAHE dynamically collapses, and the conductivities exhibit an oscillatory behavior, which becomes more prominent with decreasing optical field frequency or increasing field strength.

In Fig. 3 we show the full dependence of the dynamical Hall conductivity on both the optical frequency and field strength. The lower left corner of the plot...
labeled by “QAHE” corresponds to the low-field adiabatic regime in which the quantized Hall conductivity remains robust. The region with $E < E_{th} \approx 0.12E_0$ and $\hbar \Omega \in (0, \Delta)$ corresponds to the linear-response regime where the Kubo theory is valid; whereas the region with $\hbar \Omega/\Delta \approx 0.2$ and $E/E_0 \ll 1$. This regime with low frequency and high electric field is characterized by a strong light-matter coupling, since $\lambda = (E/E_0)/(\hbar \Omega/\Delta)^2 \gg 1$. Numerical calculations in this regime is challenging because of the large number of Floquet modes involved due to a small frequency. To understand the behavior in this regime, we treat the time-independent part of the Hamiltonian $H_k$ as a perturbation \[^{42}\] valid when $V_0 \gg \varepsilon_k \geq \Delta/2$. To linear order in $\varepsilon_k/V_0$, we find that $\sigma_{yx}^{\alpha \beta}(E) \propto \sum n \in \mathbb{N} Q_n W_n$, where $Q_n$ and $W_n$ are weight functions corresponding to the Floquet state with $n$-photon excitations ($n \geq 1$), labeled by “$F_n$” in Fig. 3.

$$Q_n = \int_{-\infty}^{\infty} d\tilde{\omega} \int_{-\infty}^{\infty} d\tilde{\omega}' P \frac{1}{\tilde{\omega} - \tilde{\omega}' + \imath 0} \rho(\tilde{\omega}) \rho(\tilde{\omega}' + n\Omega)$$

$$\times \left[ f_{FD}(\tilde{\omega}) - f_{FD}(\tilde{\omega}' + n\Omega) \right],$$

$$W_n = \frac{E_0}{E} \frac{\varepsilon_n}{2} \left[ J_{n-1}(2\lambda)[J_n(2\lambda) - J_{n-2}(2\lambda)](1 - \delta_{n,1}/2),$$

where $P$ stands for the principle value integral, $\rho(\tilde{\omega}) = -\imath \pi \sum_k \sum_{\alpha \in \{\uparrow, \downarrow\}} [\hat{G}_k^R(\tilde{\omega})]_{\alpha \alpha} n_n/\pi$ is the time-averaged local density of states with $\tilde{\omega} = \omega + n\Omega$, and $J_n(x)$ is the Bessel function of the first kind (see Supplementary Material for details). We note that this result is closely connected to the tunneling current formula under A.C. bias voltage in the Tien-Gordon theory \[^{43}\]; in fact, the Hall conductivity with Eqs. (7)-(8) is in the form of the Kramers-Kronig counterpart of the Tien-Gordon tunneling conductivity. Our numerical results show that the field dependence of $Q_n$ is insignificant. Instead, the main effect is captured in the asymptotic form of $W_n$ for $\lambda \gg 1$,

$$W_n \approx \frac{1}{2\pi} \left[ (-1)^n + \frac{1}{2} \delta_{n,1} \right] \frac{E_0 \cos(4\lambda)}{E} \frac{\lambda}{\Omega},$$

from which we see that the Hall current oscillates with the optical field strength $E/E_0$ at a frequency $\sim (\Delta/\hbar \Omega)^2$. The oscillation frequency is therefore a direct probe of the light-matter coupling $\lambda$, with more frequent oscillations characterizing a stronger coupling.

For bismuth-based TIs with Dirac velocity $v \approx 5 \times 10^5$ m s$^{-1}$ and magnetically-induced gap $\Delta = 0.02 - 0.2$ eV \[^{14}\] \[^{16}\] \[^{26}\] \[^{27}\], we estimate that the required optical frequency and field strength for observing coherent oscillations of the Hall conductivity are $\Omega < 30.39 - 303.9$ THz and $E \leq 1.215 - 121.5$ MV m$^{-1}$, which are well within current experimental accessibility. The dynamical Hall conductivity in the TI film can be measured indirectly through magneto-optical Faraday and Kerr rotations, or directly in a standard Hall measurement geometry illu-
minated with the linear polarization of the normally incident light parallel to the length of the Hall bar. In addition to TIs, graphene or bilayer graphene doped with noble metal atoms offer an alternate class of systems with a band gap and associated topological Hall transport \[2,13\]. In this scenario, the valley degrees of freedom give rise to a quantized valley Hall conductivity. Illuminated by a strong optical field, dynamical valley Hall currents will be generated in the transverse direction, which can be measured in a nonlocal transport geometry \[43–47\].

In summary, we have developed a theory for the dynamical quantum anomalous Hall effect driven by intense optical fields. Our theory addresses the question of the robustness of topological Hall quantization in the non-linear electric field regime, and predicts a collapse of Hall quantization at high fields accompanied by coherent conductivity oscillations as a function of optical field strength. Our work sheds light on the problem of the nonequilibrium dynamical response of topological phases under a strong optical field, and our findings should offer new insights in nonequilibrium topological states particularly in the low-frequency, adiabatic regime.

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