Weighing Dirac fermions by nonlinear Hall effect

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Breaking the time-reversal symmetry on the surface of a topological insulator can open a gap for the linear dispersion and make the Dirac fermions massive. This can be achieved by either doping a topological insulator with magnetic elements or proximity-coupling it to magnetic insulators. While the exchange gap can be directly imaged in the former case, measuring it at the buried magnetic insulator/topological insulator interface remains to be challenging. Here, we report the observation of a large nonlinear Hall effect in iron garnet/Bi₂Se₃ heterostructures. Besides illuminating its magnetic origin, we also show that this nonlinear Hall effect can be utilized to measure the size of the exchange gap and the magnetic-proximity onset temperature. Our results demonstrate the nonlinear Hall effect as a spectroscopic tool to probe the modified band structure at magnetic insulator/topological insulator interfaces.

Dirac fermions [1] exist in nature as elementary particles with spin 1/2. Although massless Dirac fermions are rare in particle physics, they can emerge as quasiparticles in condensed matter systems such as graphene [2] and topological insulators (TIs) [3, 4]. Due to strong spin-orbit-coupling-induced band inversion, threedimensional (3D) TIs [5, 6] host gapless linear dispersion on the surface, protected by time-reversal symmetry (TRS). As a result, the effective mass of the electrons and holes is zero. When TRS is broken by introducing magnetic atoms into or on the surface of the TI, the Dirac dispersion opens a gap and acquires a parabolic shape, and the fermions become massive [7-10]. When the Fermi level is further tuned into the exchange gap, scientifically exotic as well as technologically promising phenomena like quantum anomalous Hall effect (QAHE) [11] and topological magnetoelectric effect [12] can arise.

The QAHE has been experimentally achieved in the $3D TI (BiSb)_2Te_3$ through both doping [13–15] and proximity-coupling to magnetic insulator (MI) [16] approaches at sub-Kelvin temperatures. Compared with the former, the magnetic proximity effect (MPE) method has the advantage of allowing optimizing the $T_{\rm C}$ of the MI and the bulk gap of the TI independently [17]. In order to raise the low QAHE observation temperature. measuring the exchange gap and understanding the MPE mechanism are crucial. However, the buried MI/TI interface evades surface-sensitive spectroscopic methods like angle-resolved photoemission spectroscopy (ARPES) [8– 10] and spectroscopic imaging scanning tunneling microscopy (SI-STM) [18], and the size of the gap and even the onset temperature of the MPE remain elusive and controversial [19,20]. In this Letter, we show that the MPE in MI/TI heterostructures can give rise to a large nonlinear Hall effect (NLHE) which is absent in nonmagnetic TIs. It can be further used to measure the MPE onset temperature and the size of the exchange gap, or equivalently, the mass of the Dirac fermions.



FIG. 1. Physical mechanism of the MPE-induced NLHE. (a) An ideal TI grown on a magnetic substrate has a linear Dirac dispersion. It does not exhibit the NLHE whether an IP field is applied or not. (b) When the TI is proximity-coupled to a magnetic layer, an exchange gap is opened for the dispersion and v becomes k-dependent. When there is no magnetic field, $j_y^{(2)}$ is still zero. (c) When a finite B is applied in the x direction, v is shifted in the y direction and $j_y^{(2)}$ becomes finite.

The physical mechanism of the MPE-induced NLHE is illustrated in Fig. 1. By semiclassical Boltzmann equation with constant relaxation time approximation, the second-order transverse charge current is

$$j_y^{(2)} = -\frac{e^3 E_x^2 \tau^2}{4\pi^2 \hbar^2} \int d\mathbf{k} v_y \frac{\partial^2 f_0}{\partial k_x^2},\tag{1}$$

where e is the electron charge, τ the relaxation time, \hbar

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the reduced Planck constant, v_y the y-component electron velocity, f_0 the equilibrium Fermi distribution function, and the electric field E_x is applied in the x direction. For a TI standing on a nonmagnetic substrate [Fig. 1(a)], the Hamiltonian for the topological surface states (TSS) is $H = \hbar v_F (k_x \sigma_y - k_y \sigma_x)$ where $\sigma_i \ (i = x, y, z)$ are the Pauli matrices. It gives an ideal linear dispersion and a constant group velocity $v = \pm v_{\rm F}$ (\pm stands for $\pm k$ branches). As a result, $j_{y}^{(2)}$ is zero after the integration and it remains zero even when an in-plane (IP) magnetic field is applied. When the TI is coupled to a magnetic material with out-of-plane (OP) magnetic moments, a mass term Δ is introduced to the Hamiltonian $H = \hbar v_F (k_x \sigma_y - k_y \sigma_x) + \Delta \sigma_z$, the dispersion acquires a nonlinear form $\varepsilon = \sqrt{\hbar^2 v_{\rm F}^2 k^2 + \Delta^2}$, and the velocity becomes k-dependent. When the magnetic field B_x is zero, there is no NLHE due to symmetry constraint [21] (Fig. 1(b)). However, when a finite B_x is applied in the x direction, the Hamiltonian takes the form

$$H = \hbar v_{\rm F} (k_x \sigma_y - k_y \sigma_x) + \frac{g \mu_{\rm B} B_x}{2} \sigma_x + \Delta \sigma_z, \quad (2)$$

with g being the Landé g-factor and $\mu_{\rm B}$ the Bohr magneton, and the velocity becomes

$$v = \frac{\hbar v_{\rm F}(k_y - aB_x)}{\sqrt{\hbar^2 v_{\rm F}^2 [k_x^2 + (k_y - aB_x)^2] + \Delta^2}} v_{\rm F}, \qquad (3)$$

where $a \equiv \frac{g\mu_{\rm B}}{2\hbar v_{\rm F}}$. As can be seen in Fig. 1(c), with the shifted v_y , the contributions to $j_y^{(2)}$ from $\pm k$ branches no longer cancel each other, and a nonzero $j_y^{(2)}$ is generated, giving rise to the NLHE.

Based on previous spectroscopic results [8–10] on *n*doped Bi₂Se₃, the exchange gap Δ is typically a few tens and the Fermi energy $\varepsilon_{\rm F}$ a few hundreds of meV. As a result, for the small field (~0.1 T) and low temperature (<50 K) regime investigated in this study, the two relations $g\mu_{\rm B}B \ll \hbar v_{\rm F}k_{\rm F}$ and $k_{\rm B}T \ll \varepsilon_{\rm F}$ are well satisfied, and a simple analytical expression can be obtained for the nonlinear Hall current density as (see Supplemental Note 1 for derivations [22])

$$j_y^{(2)} = \sigma_{xy}^{(2)} E_x^2 = \frac{e^3 \tau_{\rm bs}^2 v_{\rm F}}{8\pi\hbar^2} \left(\frac{\Delta}{\varepsilon_{\rm F}}\right)^2 \frac{g\mu_{\rm B} B_x}{\varepsilon_{\rm F}} E_x^2, \quad (4)$$

where $\sigma_{xy}^{(2)}$ represents the nonlinear Hall conductivity and $\tau_{\rm bs}$ is the relaxation time of the bottom surface. Similarly, the longitudinal conductivity under a *y*-direction field is derived as $\sigma_{xx}^{(2)} = -\sigma_{xy}^{(2)}$ [22]. This is distinct from the previous observed NLHEs in TIs [23, 24] where $|\sigma_{xx}^{(2)}| = 3|\sigma_{xy}^{(2)}|$ was found.

In our experiments, we used the second-harmonic voltage (SHV) method to detect the NLHE. With Equation (4), the measured second-harmonic Hall resistance $R_{yx}^{2\omega} \equiv V_y^{2\omega}/I_x$ is calculated as (Supplemental Note 2)

$$R_{yx}^{2\omega} = \frac{1}{w} \frac{\sigma_{xy}^{(2)}}{r \left[\sigma_{xx}^{(1)}\right]^3} I_x = \frac{8\pi^2 \hbar^4 v_{\rm F} \tau_{\rm bs}^2 \Delta^2 g \mu_{\rm B} B_x}{r w e^3 \tau_{\rm avg}^3 \varepsilon_{\rm F}^6} I_x, \quad (5)$$

where I_x is the current, w is the width of the Hall bar, r accounts the ratio of the current going through the bottom surface, and in order to show explicitly the Fermi energy dependence, we adopt an effective 2D form for the total linear conductivity $\sigma_{xx}^{(1)} = \frac{1}{R_{xx}} \frac{l}{w} = \frac{e^2 \tau_{\text{avg}} \varepsilon_{\text{F}}}{4\pi\hbar}$ with l and R_{xx} being the length and resistance of the sample and τ_{avg} an average relaxation time for all the bulk and two surface conduction channels. Besides exhibiting the linear dependence on electric field (or current) and magnetic field, Eq. (5) also shows that the size of the Dirac mass gap Δ can be inferred from the NLHE once ε_{F} , τ_{bs} and current distribution r are determined.

To investigate the TRS breaking effect on the NLHE in TI, we grew 8 quintuple-layer (QL) Bi_2Se_3 (BS) films on magnetic insulator $Tm_3Fe_5O_{12}$ (TmIG, 30 nm) and $Y_3Fe_5O_{12}$ (YIG, 2.5 μm and 100 nm) films with OP and IP easy-axis respectively, and nonmagnetic $Gd_3Ga_5O_{12}$ (GGG) substrate with a low-temperature Se-buffer layer growth method [25]. As seen in Fig. 2(a), streaky reflection high-energy electron diffraction (RHEED) patterns were observed from the very first QL, and x-ray diffraction (XRD) results of all the Bi_2Se_3 films exhibit clear (0,0,3n) peaks, indicating the *c*-axis growth orientation and the high and similar crystalline quality. After fabricating the films into $200 \times 100 \ \mu m$ Hall bar devices by photolithography [Fig. 2(b) inset], we sent a low frequency a.c. current in the x direction, and measured the second-harmonic Hall voltage (SHV) with lock-in technique while sweeping the external magnetic field also in the x direction. The results at 4.5 K for three samples having similar carrier densities $(n_{\rm s} \sim 3 \times 10^{13} {\rm ~cm^{-2}})$ are plotted in Fig. 2(b). Here and after, YIG refers to the 2.5 μ m thick sample unless otherwise noted. Consistent with above theoretical analysis, the NLHE in timereversal invariant GGG/BS is negligible, while in TmIG and YIG/BS the gapped surface states give rise to a linear-in-B NLHE [Eq. 5]. The YIG/BS sample also exhibits an extra hysteresis close to zero field corresponding to the reversal of the IP magnetization. The origin of this hysteretic NLHE [24] will be investigated elsewhere. In this study, we focus on the linear-in-B one. We note that although the easy axis of YIG is IP, the Dirac-electronmediated exchange interaction [7, 26] tilts the magnetic moments to OP direction at the interface [27, 28], which in turn opens a gap for the TSS. The presence of MPE in TmIG/BS and YIG/BS samples is also evidenced by the suppression of weak-antilocalization (Supplemental Fig. S3). As seen in Fig. 2(c) inset, the linear current dependence of $R_{yx}^{2\omega}/B$ demonstrates the nonlinear nature of the Hall effect $(V_y^{2\omega} = I_x R_{yx}^{2\omega} \propto I_x^2)$. The deviation



FIG. 2. NLHE in MI/TI heterostructures. (a) XRD results of 8 QL Bi₂Se₃ films grown on TmIG, YIG and GGG substrates. Inset: RHEED images after 1 and 8 QL growth. (b) $V_y^{2\omega}$ as a function of the *x*-direction field for the three samples. Lower inset: a device image with illustrated experimental setup. Scale bar, 100 μ m. (c) The nonlinear longitudinal $R_{xx}^{2\omega}$ and transverse resistance $R_{yx}^{2\omega}$ under *x*- and *y*-direction field scans. Lower inset: the current dependence of $R_{yx}^{2\omega}/B_x$.

from linear dependence at large current densities is due to Joule heating.

A similar NLHE was previously observed in 20 QL Bi_2Se_3 grown on Al_2O_3 substrate. It was attributed to the intrinsic nonlinear features in the Dirac dispersion, i.e., the k^3 hexagonal warping and the k^2 particle-hole

asymmetry terms. These effects can be ignored here due to the following reasons. First, if these contributions are dominant, the NLHE in GGG/BS, TmIG/BS and YIG/BS samples should be in the same order of magnitude because of the similar carrier density and crystalline qualities. However, the SHV response in GGG/BS is negligible while the magnitude of the NLHE in TmIG/BS and YIG/BS scaled by the coefficient $\gamma_y \equiv \frac{R_{yx}^{2\omega}}{R_{xx}IB}$ reach 0.025 and 0.031 respectively, which are two orders of magnitude larger than that in Al₂O₃/Bi₂Se₃ ($\gamma_y = 1 \times 10^{-4}$) [23]. Second, as seen in Eq. (5) and data in the following, the MPE-induced NLHE scales inversely with carrier density (or Fermi level), while the warping-induced NLHE is most pronounced at high carrier concentrations. And third, as exemplified by the result for the TmIG/BS sample [Fig. 2(c)], the ratio between the transverse and longitudinal nonlinear conductivities $\frac{\sigma_{xy}^{(2)}}{\sigma_{xx}^{(2)}} = \frac{R_{yx}^{2\omega}}{R_{xx}^{2\omega}} \frac{l}{w}$ in our TmIG/BS and YIG/BS samples are 0.89 and 0.82 respectively, which are close to the theoretical value 1 but away from the value 1/3 calculated for the warping-induced NLHE [23]. Similarly, Nernst effect can be ruled out as a dominant contribution to the SHV due to the absence of it in the GGG/BS sample. Thus, Fig. 2(b) unambiguously demonstrates the magnetic origin of the NLHE in MI/TI heterostructures. Fig. 2(c) also shows that under a y-direction field scan, $R_{yx}^{2\omega}$ becomes much smaller than that in an x-direction field, consistent with the angular dependence predicted by Eq. (5).

After establishing the origin of the NLHE, we now turn to use it to estimate the Dirac mass gap. Fig. 3(a) displays the temperature dependence of the NLHE in the TmIG/BS sample. From 4.5 to 200 K, $R_{yx}^{2\omega}$ at a fixed field decreases sharply and becomes negligible above 30 K, reflecting the weakening and finally disappearance of the MPE. In Eq. (5), g and $v_{\rm F}$ of Bi₂Se₃ can be found from literature as 19.5 [29] and 5×10^5 m/s [6] respectively. The surface electron relaxation time for TIs was calculated to be 230-550 fs [30, 31]. Here, to account the suppression of WAL from MPE, we take a reduced value of 100 fs. The bulk Fermi level 0.28 (0.26) eV of the TmIG (YIG) sample is estimated from the carrier density 3.5 (3.0)×10¹³ cm⁻², and τ_{avg} is calculated from the known R_{xx} and $\varepsilon_{\rm F}$. The major uncertainty lies in estimating r, the amount of current going through the bottom surface. Because of the large dielectric constant (~ 12.5) of garnets [32], it is likely that there is an upward band-bending (BB) at the bottom surface, just like the upward BB formed on the top surface of Bi_2Se_3 in contact with the high-k dielectric HfO_2 [33]. This causes depletion of the bottom surface electrons and effectively lowers the Fermi level relative to the Dirac point. This BB picture is supported by the one-order-of-magnitude reduction in surface carrier density in YIG/Bi₂Se₃ films as compared with Si/Bi_2Se_3 films [34]. Assuming the top and bottom surface carrier densities are $n_{\rm s}^{\rm ts} = 5 \times 10^{12}$



FIG. 3. Inferring the mass of Dirac fermions. (a) The nonlinear Hall resistance $R_{yx}^{2\omega}$ under x-direction field scans at various temperatures. Inset, the temperature dependence of the slope $R_{yx}^{2\omega}/B$. (b) The calculated exchange gap Δ or mass $m = \Delta/v_{\rm F}^2$ as a function of temperature. The MPE onset temperature is determined as the intercept of two linear fit in the massless and massive regimes.

cm⁻² and $n_s^{\rm bs} = 5 \times 10^{11}$ cm⁻² [34, 35] and all electrons have the same mobility, r is roughly estimated to be 1.4 and 1.7% for the TmIG and YIG samples (Supplemental Note 6). After rescaling the local Fermi level by $n_s^{\rm bs}$, the exchange gap is calculated to be 22 and 17 meV for these two samples. Although due to the large uncertainties in $n_s^{\rm bs}$, r and $\tau_{\rm bs}$, these values are only rough estimations, the order-of-magnitude agreement with the gap size obtained previously by ARPES measurements in Mn- or Fe-doped Bi₂Se₃ (7-50 meV) [8] and SI-STM in Cr-doped (BiSb)₂Te₃ (9-48 meV) [18] demonstrates the validity of using the NLHE to probe the exchange gap at the buried MI/TI interface.

The inferred Dirac gap Δ or equivalently mass $m = \Delta/v_{\rm F}^2$ at different temperatures are summarized in Fig. 3(b). It unambiguously shows the evolution of the Dirac electrons from being massless to massive when tem-



FIG. 4. Carrier density dependence of the coefficient $\gamma_y \equiv \frac{R_{xx}^{2\omega}}{R_{xx}IB}$ in various devices. YIG is the same 2.5 μ m thick sample discussed in the main text. YIG2 is another 100 nm thick sample. The solid line is fit to $n_{\rm s}^{-3}$.

perature is lowered. The sharp increase of Δ below 30 K indicates that MPE is most pronounced at low temperatures. By extrapolating the intercept of two linear fit in the massive and massless regimes, we obtain the MPE onset temperature T_{MPE} for the TmIG and YIG samples as 27 and 28 K, respectively. The unexpectedly low and close T_{MPE} may suggest the key role of *coherent* Dirac electrons in mediating the MPE in MI/TI heterostructures [17, 26, 36].

The carrier density dependence of the NLHE is shown in Fig. 4, where γ_y is plotted as the *y*-axis to remove the effect from different resistance in different devices. The data fit qualitatively well to $n_{\rm s}^{-3}$, which agrees with our theoretical model [Eq. (5)] and distinguishes it from the NLHE due to hexagonal warping.

The nonreciprocal transport responses [37] in noncentrosymmetric systems depend sensitively on the band structure. As a special case, our results show that the MPE-induced massive Dirac dispersion enhances the NLHE on the TI surface by two orders of magnitude. We also demonstrate that inversely, this NLHE can be used as a scale to measure the mass of Dirac fermions. This scale can be further calibrated if the Fermi level is tuned into bulk band gap, e.g., by chemical doping or electric gating, so current distribution and surface electron relaxation time can be more accurately determined. Even when uncalibrated, this NLHE probe can accurately tell when does the exchange gap starts to form and how does it evolve with temperature, which is helpful for understanding the MPE mechanism and realizing higher-temperature QAHE in MI/TI heterostructures. It may also be applied to modulation-doped [14] or intrinsic magnetic [38, 39] TIs where only one surface is tuned to interact with magnetism.

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