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Magnetic proximity effect (MPE) in a topological insulator (TI)/magnetic insulator (MI) heterostructure induces magnetization to the TI's electronic states driven by means of magnetic exchange coupling from the MI layer [1–14]. As long as the MPE-induced magnetic order is realized through the Dirac electrons, this effect is expected to have applications in low-energy-consumption electronic and spintronic devices [1,2,15–17]. One prominent application is the quantum anomalous Hall effect (QAHE), where dissipationless chiral edge current can be harbored without external magnetic field [18–24]. Comparing with the other approach to magnetize the Dirac surface state (DSS) by doping the TI with transitional-metal ions [20,21,25–29], the MPE approach has the advantage that it can result in a uniform magnetization over the entire TI layer without creating any impurity sites nor destroying the nontrivial band structure [2,8,14,30]. While the QAHE to date has been realized in Cr- and V- doped TI systems [18–20,22,23,27], no QAHE has been hitherto demonstrated in a MPE-induced ferromagnetism system in spite of extensive studies. This brings up a few fundamental questions: Is the MPE actually happening to the DSS of TI? If so, what is the nature of the MPE-induced ferromagnetic order in DSS? For the first question, MPE in principle can happen to any electronic states of TI, including bulk states [31]. As to the second question, if, for instance, the MPE is realized through coupling with free carriers of the TI, the MPE will be enhanced by increasing the carrier density, hence hampering QAHE due to its carrier-free requirement [21]. Therefore, an experimental study on the correlation between the carrier density and the magnitude of MPE in TI will show whether the MPE through a MI is a viable route to realize QAHE.

Recently, the contribution of the bulk n-type carriers to MPE in a highly n-doped TI Bi2Se3/MI EuS heterostructure was studied through polarized neutron reflectometry (PNR) [9]. In this paper, by further equipping the PNR spectrometer with additional electrical transport capability and bottom-gate voltage (Vg), we are able to resolve the DSS’s contribution to the proximity-induced magnetism at the interface between TI (Bi0.2Sb0.8)3Te3 and MI EuS. Specifically, we found the maximum proximity-induced magnetism is realized when the Fermi level is close to the Dirac point.

The PNR experiments were carried out at beamline NG-D at the NIST Center for Neutron Research, at fixed temperature T = 5 K. PNR is a powerful technique for measuring the real-space magnetic structure of thin films [32]. The experimental setup is illustrated in Fig. 1(b), where the incident spin-polarized neutron beams are reflected by the MI/TI heterostructure (red and blue spheres), while the spin nonflip reflectivity signals from spin-up and spin-down components, R↑ and R↓, are collected alternatively. The PNR is integrated with a custom-built sample holder (Fig. 1b) and see Supplemental Material [33] for the image of the experimental setup, the x-ray diffraction (XRD), the interpretation of spin asymmetry, fitting scheme, and fixed-scattering length density (SLD) fitting for the image of the experimental setup, which allows in situ gate-dependent two-terminal longitudinal transport measurement while performing the PNR measurements. A μ0H = 0.7 T guide field is exerted at all times to prevent possible neutron depolarization. Figure 1(a) is a schematic of the EuS/(Bi0.2Sb0.8)3Te3 heterostructure. Both perpendicular magnetization, which causes the surface state band-gap opening [21], and the in-plane magnetization, which results in the shift of the Dirac cone [24], are capable of...
inducing QAHE. In the present setup, we focus on the in-plane magnetization, yet there might still be a canting effect which allows the spin rotation toward the perpendicular direction [9]. The reason for choosing a Eu-based element as proximity layer is its extremely high neutron absorption cross section [$\sigma_A(E_n) \approx 4530$ barn], which helps to identify the location of the proximity layer and has been implemented in a few recent studies [8,9]. Moreover, an ambipolar behavior of MPE is revealed, that for both $n$- and $p$-doped TI, the MPE is reduced when the carrier density is increased. This strongly suggests that the MPE in a TI/MI heterostructure is not originated from the free carriers and hence enables the MPE-based QAHE. The reduced proximity-induced magnetism at increased carrier density can be understood as a diamagnetic screening effect. We have developed a phenomenological model to explain this effect qualitatively. Our study sheds light on further exploring the direction of MPE toward next-generation dissipationless electronic and spintronic applications.

Since the purpose of this measurement is to detect the MPE’s possible variation, which can be considered as a second-order effect if the ferromagnetism in MI layer is zeroth order and the induced MPE is a first-order effect, the sample quality becomes essential. High-quality MI 5-nm EuS/4 quintuple layers (QLs) intrinsic TI (Bi$_2$Sb$_2$Te$_3$)$_x$Te$_{1−x}$ heterostructure is grown by molecular-beam epitaxy under a base vacuum $\sim 5 \times 10^{-10}$ Torr. The Bi:Sb ratio is carefully optimized to locate the Fermi level close to the Dirac point based on the method reported in Ref. [34]. The 4QL TI thickness is chosen to make sure no hybridization gap is formed on the surface states and to facilitate the bias-gate voltage tunability due to the low bulk-carrier concentration [35]. The TI thin film is grown on top of heat-treated 0.5-mm-thick SrTiO$_3$ (STO) (111) substrate for back-gating purpose.

FIG. 1. (a) Atomic configuration (left) and schematic band structure (right) at the MI/TI interface. When the ferromagnetic Eu ions magnetize the surface state of the TI (red arrow in left figure), the original Dirac cone (purple dotted lines) is shifted away from $\Gamma$ point (purple solid lines) through exchange coupling, causing MPE. (b) Schematics of the gate-dependent neutron reflectometry system, where the incident neutron momentum, reflected neutron momentum, and the momentum change are denoted as $k_i, k_f$, and $Q (Q \equiv k_f − k_i)$, respectively. The two neutron spins switched by a spin flipper shine onto the sample alternatively. See Sec. I of the Supplemental Material [33] for an image of the setup.

Upon fitting, the fitted curves [solid lines in Fig. 2(a)] show excellent agreement with the experiment data (filled points), with logarithmic figure of merit $\sim 5 \times 10^{−2}$. To eliminate any hysteresis effect caused by the ferroelectricity of STO [38], the whole sample was warmed up to room temperature and cooled back down under high vacuum during the polarity change from $+200$ to $−200$ V. For this reason, a very small thickness and density variation is allowed in the refinement process to obtain better fitting quality (see Sec. III of the Supplemental Material [33] for fitting scheme). A global fitting of all spectra simultaneously with strictly fixed thickness and SLD...
is performed independently using the REFLLD package \cite{39}, showing an identical qualitative trend that the proximity magnetism is higher at low gate voltage (see Sec. IV of the Supplemental Material \cite{33} for fixed-SLD fitting).

The refined SLD profiles from the fitted reflectivity curves at a few representative \( V_g \) are plotted in Fig. 3. The resulting nuclear scattering length density (NSLD) agrees well with the expected cross-section calculation that the STO substrate gives \( -3.6 \times 10^{-2} \text{ nm}^{-2} \), TI layer \( 1.8 \times 10^{-4} \text{ nm}^{-2} \), EuS \( 1.9 \times 10^{-4} \text{ nm}^{-2} \), and amorphous Al\(_2\)O\(_3\) capping layer \( 4 \times 10^{-4} \text{ nm}^{-2} \). From the magnetic scattering length density (MSLD), it can be seen clearly that at \( V_g = +20 \text{ V} \) the proximity MSLD is increased to \( 2.5 \mu_B/\text{unit cell} \), comparing with \( \sim 1.2 \mu_B/\text{unit cell} \) at \( V_g = +200 \text{ V} \), which is comparable with a previous report in Ref. \cite{8}, where TI\(_{\text{Sb}_2\text{Te}_3}\) was used. The absorption scattering length density (ASLD) indicates the region of neutron absorption; in this system, the ASLD is solely coming from Eu atoms. The warming and cooling of the sample between \( V_g > 0 \) measurements and \( V_g < 0 \) measurements causes some change of STO/TI interfacial roughness (Fig. 3(a) vs Fig. 3(b)), making the fixed-SLD analysis valid for \( V_g > 0 \) only (see Sec. III of the Supplemental Material \cite{33} for fitting scheme). Despite that two independent PNR refinement strategies and SA from raw data point to the same conclusion of peak proximity, we do not intend to fully exclude other possible SLD configurations due to the nuance effect caused by electric field.

At this stage, the effect of carrier density tuned through a \( V_g \) to the proximity-induced magnetism becomes clear. The MPE strength as a function of \( V_g \) is plotted in Fig. 4(a), together with the sheet longitudinal resistance \( R_{xx} \). It can be seen clearly that the MPE shows a peak at \( V_g \approx 0 \text{ V} \), which coincides with the behavior of the \( R_{xx} \), indicating that the highest MPE is realized near the Dirac point. Moreover, a finite carrier density—both \( n \) and \( p \) type—could reduce the MPE to the normal value as reported in Ref. \cite{8}. On the other hand, the \textit{ex situ} magnetoresistance (MR) is performed as well. For longitudinal MR measurement (Fig. 4(b)), a typical weak antilocalization (WAL) behavior is revealed at almost all voltages \cite{40,41}. In the weak \( H \) regime, the MR shows a steeper increase when the Fermi level is closer to the Dirac point. This behavior is consistent with a previous report \cite{42}. Besides, the Hall measurements are also performed (Fig. 4(c)), from which we could verify the carrier type and estimate the carrier density \cite{43}. The lowest carrier density \( n_{2D} = -2.8 \times 10^{12} \text{ cm}^{-2} \) is located at \( V_g = +20 \text{ V} \). Since the carrier densities at \( V_g = -200, -50, +50, \) and \( +200 \text{ V} \) are \( n_{2D} = 2.64 \times 10^{13} \text{ cm}^{-2} \), \( 8.0 \times 10^{12} \text{ cm}^{-2} \), \( -5.0 \times 10^{12} \text{ cm}^{-2} \), and \( -2.75 \times 10^{13} \text{ cm}^{-2} \), respectively, while the maximum Dirac electron carrier density accommodated in the spin-polarized Dirac cone of a TI gives \( \sim 1.6 \times 10^{13} \text{ cm}^{-2} \) (assuming \( E_{\text{Fermi}} = 0.3 \text{ eV} \) and \( v_F = 5 \times 10^6 \text{ m/s} \)), we can see that the Fermi level is already tuned from the bulk valence band to the bulk conduction band even at low \( V_g \). This is consistent with the report in Ref. \cite{44} that the critical carrier density of the Dirac cone is \( \sim 5.0 \times 10^{12} \text{ cm}^{-2} \), and is useful to simplify the theory discussed below. In addition, we

![FIG. 3. (a)–(c) The SLD profiles at \( +50 \), \( +0 \), and \( -100 \text{ V} \), respectively. The NSLD contains the information of sample thickness and interfacial roughness, which agrees well with the TEM result in (d). From the MSLD, we can see that the magnetic proximity effect (yellow circled region) can be tuned through \( V_g \), and is reduced for both \( n \)- and \( p \)-type doping. The ALSLD signal solely comes from the Eu ions; hence, the lack of ASLD \( \sim 4 \text{ nm} \) in the TI side indicates that the proximity-induced magnetism is not coming from the interdiffused Eu ions, which is further confirmed from the sharp TI/EuS interface in the TEM image. On the other hand, there are diffused Eu ions to the Al\(_2\)O\(_3\) cap, which can be seen from both ASLD \( \sim 9 \text{ nm} \) at rough interface of EuS/Al\(_2\)O\(_3\) from the TEM.](image)
notice that the peak of MPE ($V_g \sim +25$ V by fitting) and the peak of $R_{xx}$ ($V_g = +20$ V) have a slight mismatch, indicating that the transport and neutron scattering may have slightly different origins of the contributing carriers. One possible explanation is that the transport may have contributions from both bottom TI/STO interface and top TI/Mp interface while the spatially resolved neutron on only is sensitive to the top TI/Mp interface. Even so, we believe the transport and neutron results are directly comparable, since the gating is strong enough to tune the Fermi level of the top surface due to the high dielectric constant of TI materials ($\sim 100$) [20,27,45].

At this point, one may wonder whether MPE is caused by Dirac electrons or if they merely have coincidental correlation. To further understand the implication in Fig. 4(a), we developed a simple phenomenological model to account for this ambipolar behavior from a perspective of magnetic susceptibility. It is well known that the bulk electronic bands for this ambipolar behavior from a perspective of magnetic susceptibility

\[ \chi_{\text{tot}}(q) = \frac{1}{2} \frac{\chi_s^B(q - \theta(q)) + \chi_s^D(q + \theta(q))}{\chi_s^B(q - \theta(q)) + \chi_s^D(q + \theta(q))}, \]

where $\alpha$ is the Heaviside step function. The spin susceptibility $\chi_s$ can be written as [31]

\[ \chi_s^B(q) = \frac{\mu_B^2}{4\pi^2} \sum_{m,n} \int d^2k \left[ f_0(E_{n,k}^{R,D}) - f_0(E_{m,k+q}^{R,D}) \right]. \]

where $E_{n,k}^{R,D}$ are the eigenvalues for the corresponding surface or bulk Hamiltonian; $f_0$ is the Fermi-Dirac distribution function. Here, by plugging the material-dependent eigenvalues [46] into Eq. (2), the susceptibility can be computed accordingly. For the particular EuS/(Bi$_2$Sb$_3$)$_3$Te$_3$ system, due to the lack of the corresponding effective Hamiltonian, here only a qualitative picture is provided, which is sufficient to demonstrate whether carrier density can cause a unipolar ($\chi$ changes monotonically with $E_F$) or an ambipolar effect ($\chi(E_F = 0)$ is a local minimum or maximum). Since the bias voltage is shown to be effective enough to tune $E_F$ into bulk bands even at low $V_g$, assuming $E_c \approx E_D$ (small paramagnetic region) and neglecting all anisotropy, which allows a much simpler dispersion relation $E_{m,k+q}^{B,D} = \sqrt{k^2 + q^2}$ (the resulting trend does not depend on the particular form of dispersion), an ambipolar effect is revealed [Fig. 4(d)] that when $E_F$ is away from the Dirac point, the diamagnetic screening gets stronger for both $n$- and $p$-type doping, hence reducing the MPE. This agrees with our experimental results in Fig. 4(a). At even higher $E_F$, the screening effect is saturated, since besides screening, other electronic states could directly contribute to the proximity-induced magnetism [31].

In summary, we showed the Dirac-electron-mediated MPE at the interface of a MI/TI heterostructure by means of gate-dependent PNR measurements. The tunability of the MPE enables future theoretical studies to utilize MPE to manipulate the interfacial spin texture in TI [47–49]. The maximum MPE, in particular, is realized when the Fermi level is approaching the Dirac point where carrier density is minimized. Since the realization of in-plane magnetization-induced QAHE requires both that the Fermi level is near the Dirac point together with the strong MPE, our study proves that the MPE is favored at low carrier concentration, which opens up the possibility for realizing in-plane magnetization-induced QAHE [24] at MI/TI heterostructure.

M.L., Q.S., and G.C. would like to acknowledge support by S3TEC, an Energy Frontier Research Center funded by US Department of Energy (DOE), Office of Basic Energy Sciences (BES) under Award No. DE-SC001299/DE-FGG02-09ER46577 and DARPA MATRIX Program Contract No. HR0011-16-2-0041. M.L. and C.Z.C thank A. Grutter and B. Kirby for the helpful discussion. Y.Z. and L.W. acknowledge the support from DOE-BES / Materials Science and Engineering (MSE) division under Contract No. DE-AC02-98CH10886. J.S.M. acknowledges the support from NSF Grant No. DMR-1207469, ONR Grant No. N00014-16-1-2657, and the STC Center for Integrated Quantum Materials under NSF Grant No. DMR-1231319. C.Z.C would like to acknowledge support from the startup grant provided by Penn State University.

M.L. and Q.S. contributed equally to this work.