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Signature of gate-controlled magnetism and localization effects at Bi₂Se₃/EuS interface

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Proximity of a topological insulator (TI) surface with a magnetic insulator (MI) can open an exchange gap at the Dirac point leading to exploration of surface quantum anomalous Hall effect. An important requirement to observe the above effect is to prevent the topological breakdown of the surface states (SSs) due to various interface coupling effects and to tune the Fermi level at the interface near the Dirac point. In this work, we demonstrate the growth of high-quality *c*-axis oriented strain-free layered films of TI, Bi₂Se₃, on amorphous SiO₂ substrate in proximity to an MI, europium sulfide (EuS), that show stronger weak anti-localization response from the surface than previous studies with epitaxially interfaced heterostructures. Importantly, we find gate and magnetic field cooling modulated localization effects in the SSs, attributed to the position of interface Fermi level within the band gap that is also corroborated from our positron annihilation spectroscopy measurements. Furthermore, our experiments provide a direct evidence of gate-controlled enhanced interface magnetism in EuS arising from the carrier mediated Ruderman–Kittel–Kasuya–Yosida interactions across the Bi₂Se₃/EuS interface. These findings demonstrate the existence of complex interfacial phenomena affecting the localization response of the SSs that might be important in proximity engineering of the TI surface to observe surface quantum Hall effects.

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INTRODUCTION

The process of breaking time reversal symmetry by introducing out-of-plane magnetism in the topologically protected surface states (SSs) of a 3-dimensional (3D) topological insulator (TI)^{1,2} leads to opening of an exchange gap (EG) at the Dirac point³. Such a mechanism support several new quantum phenomena, such as quantum anomalous Hall effect (QAHE)^{4,5}, half-integer quantum Hall effect (H-IQHE)^{1,6}, topological magneto-electric effect^{7,8}, and image magnetic monopoles⁹. In realizing these effects, the main challenge is to successfully demonstrate the presence of the EG with the Fermi level (E_F) lying within the gap. Currently, this is made possible in magnetic TIs such as in Cr- and V-doped compensated TIs, where perpendicular magnetism is developed in the bulk showing QAHE^{5,10}. However, in comparison, inducing local magnetism at TI surface through the short range nature of magnetic proximity effect (MPE) can provide a number of advantages over bulk doping^{3,5,11}; these include lower bulk and surface defect density due to improved film crystallinity, better controllability of the SSs, and independently controlling the magnetic state at each surface of TI to build switchable topological devices¹².

Conventionally, MPE can be achieved by using magnetic insulators (MIs). However, requirement of an out-of-plane magnetization at the TI surface makes this study difficult since most MIs naturally possess in-plane anisotropy. Also, new hybridization states¹³ that may form at the TI/MI interface near the E_F can contaminate the topological properties of the Dirac SSs^{14–16}. As a result, observing these effects through transport studies can become nontrivial.

In this regard, MPE studies in the 3D TI, Bismuth Selenide (Bi₂Se₃), with an MI, europium sulfide (EuS), is extensively

researched^{17–19}. Different research groups confirm the presence of an out-of-plane component or canted magnetization at the Bi₂Se₃/EuS interface with the magnetism existing at temperature much above the magnetic transition temperature (T_c) of EuS (~17 K)^{18,20}. Our magnetometry studies on Bi₂Se₃(10 nm)/EuS (1 nm) also confirm the existence of enhanced interface magnetism with a second transition close to 232 K (refer to Supplementary Fig. 2 and Supplementary Note 2). Unlike other TI/MI interfaces^{14,15,21,22}, recent theoretical calculation of the above interface also suggests induced magnetism, localized within the first quintuple layer (QL) of Bi₂Se₃²³, with the SS retaining most of the topological character compared to a pristine Bi₂Se₃ SSs²⁰. Furthermore, an induced EG of ~9 meV in the surface Dirac cone is suggested²³. Despite these reports, experimental observation of the EG opening at this surface has been elusive²⁴. Possible reasons for the above include: (a) inherent intrinsic doping from large Se vacancies leading to bulk dominated conduction with E_F pinning inside the conduction band (CB), and (b) loss of topological character/signal due to interface hybridization, surface-bulk coupling effects²⁵, and/or due to reduction in thickness (in an effort to probe the surface contribution over the bulk conduction) that weaken the robustness of bulk bands to preserve the SSs.

In literature, the topological nature of SSs and its response to MPE are probed using magnetoconductance measurements by analyzing the weak anti-localization (WAL) or weak-localization (WL) signal from the spin-momentum locked SSs. In Bi₂Se₃, this signal is weak due to the dominant bulk contribution to conductivity, thereby making them harder to detect electronically. Reducing bulk conduction in Bi₂Se₃ films is achieved either by studying thinner films, or by reducing the bulk conductivity. Previous studies with epitaxially interfaced Bi₂Se₃/EuS films¹⁷,

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Fig. 1 XRD, TEM, and device structure. **a** XRD pattern in $\theta - 2\theta$ scan of Bi_2Se_3 films grown on sapphire (0001) and a-SiO_2 substrate. **b** Cross-sectional TEM image (zoomed out view) of Bi_2Se_3 films grown on a-SiO_2 . Length of the scale bar is 200 nm. **c** Zoom-in-view of cross-sectional TEM image of Bi_2Se_3 on a-SiO_2 (top) and sapphire (0001) (bottom). Length of the scale bars is 5 nm. **d** Schematic of our TI/MI Hall bar devices with bottom gate electrode.

Fig. 2 MIT and WAL response. **a** Thickness dependence of normalized longitudinal resistivity ρ_{xx} vs. T for Se-capped (hollow symbols) and EuS-capped device (solid symbols) with 12 nm (triangles) and 16 nm (circles) thickness of Bi_2Se_3 . Corresponding temperature value of the resistivity upturn is also indicated. Inset shows the temperature dependence of μ_{Hall} of Se- and EuS-capped 16-nm-thick Bi_2Se_3 devices. **b** Change in magnetoconductance, ΔG vs. B of $\text{Bi}_2\text{Se}_3/\text{Se}$ (hollow square) and $\text{Bi}_2\text{Se}_3/\text{EuS}$ (hollow circle) devices fitted with HLN formalism (solid line). **c** Temperature dependence of HLN fitting parameters, $\tilde{A}$ (solid) and B_ϕ (hollow) for $\text{Bi}_2\text{Se}_3/\text{Se}$ (square) and $\text{Bi}_2\text{Se}_3/\text{EuS}$ (circle) devices.

including the study with thinner films (<8 nm), report anomalous Hall effect with no evidence of WAL or WL signal after EuS growth. Raman study of these interfaces also reveal strong charge transfer²⁶ that may suggest interface hybridization to weaken and destroy the topological signature of SSs. This may not be a surprise since lattice-matched interfaces can sometimes be detrimental for proximity engineering, as also observed in the study of proximity-induced superconductivity²⁷.

RESULTS AND DISCUSSION

Sample preparation

In this article, we adopt a different approach to reduce the bulk conductivity by growing out-of-plane textured Bi_2Se_3 films on amorphous SiO_2 (a-SiO_2) substrate having a hexagonal surface atomic structure that is found essential for promoting the out-of-plane magnetization of EuS ¹⁸. We achieve such a film order by depositing the films at 250°C in our molecular beam epitaxy system with a base pressure of 5×10^{-10} mbar using a co-evaporation technique. After deposition, films are cooled gradually under Se environment and a capping layer is grown at room temperature. X-ray diffraction (XRD) measurements in Fig. 1a reveal (003) family of peaks suggesting a highly c -axis oriented layered growth of Bi_2Se_3 films that is found to be structurally as good as our epitaxially grown films on c -cut sapphire substrate. Cross-sectional transmission electron microscope (TEM) images (Fig. 1c) also reveal a layered growth supporting the hexagonal structure of 0.95 nm thickness of the QL with no interfacial strain compared to the films grown on sapphire. Our reflective high-energy electron diffraction study suggested in-plane non-textured growth of the films. Similar growth findings were also noted in the work of Bansal et al.²⁸. The structural phase of the Bi_2Se_3 films was confirmed using Raman spectroscopy (see Supplementary Fig. 1 and Supplementary Note 1). For transport measurements, thicker Bi_2Se_3 films in 10–16 nm range were grown and, subsequently, a 3 nm film of EuS was deposited in a sulfur-rich environment at room temperature, without breaking vacuum, followed by a 2-nm-capping layer of AlO_x ($x \sim 1.3$ –1.5). Our XRD analysis showed no EuS peaks, suggesting its amorphous growth.

Metal-to-insulator transition (MIT) and weak anti-localization response

Hall bar structures with length, L and width, W were patterned by mechanical etching or by in-situ shadow mask (Fig. 1d) as optical lithography techniques degraded the device mobility (see Fig. S3). Hall mobility (μ_{Hall}) of our 16 nm Bi_2Se_3 films was found to be $\sim 450 \text{ cm}^2/\text{Vs}$ (inset of Fig. 2a). Figure 2a shows the temperature dependence of normalized longitudinal resistivity (ρ_{xx}) for 12 and 16 nm Bi_2Se_3 films, grown under the same conditions, with two different capping layers, viz., 2 nm Se and 3 nm EuS. Both devices show a predominant metallic signature during cool down with the decrease in ρ_{xx} for the EuS-capped devices being relatively gradual than the Se-capped devices, a trend observed in different samples with different W (200 μm to 2 mm). At lower temperatures, a weak upward trend in ρ_{xx} is observed with a stronger

response in the EuS-capped devices. In the case of Se-capped films, the upturn in ρ_{xx} occur at ~ 18 K for 12 nm films and decrease to ~ 15 K for 16 nm films. While, in the case of EuS-capped films, a stronger upturn in ρ_{xx} occurs at a temperature of 19 K, that remains unchanged in both the 12 and 16 nm $\text{Bi}_2\text{Se}_3/\text{EuS}$ devices. In general, a weak upturn in film resistivity arises due to localization effects^{29,30} in the 2D limit originating from the film disorder. However, observation of a stronger upturn in EuS-capped devices at a higher temperature (near T_c of EuS) than the reference films, while showing no dependence on the Bi_2Se_3 film thickness, suggests an additional interface mechanism at play in our EuS-capped devices. Furthermore, our observation of similar μ_{Hall} in the EuS-capped and Se-capped devices of Fig. 2a, also strengthen our above claim.

To analyze the influence of the above interface effect on the WAL response of the SSs, magnetoconductance ($G(B)$, B is the magnetic field) study in the low-field range ($B < 0.2$ T, applied perpendicular to the film) was performed at low temperatures (< 30 K). In Fig. 2b, a strong upward cusp in $\Delta G(B) = G(B) - G(0)$ is measured at zero field in both the EuS-capped and Se-capped devices with $W/L \sim 1$, providing a clear signature of the presence of 2D WAL channel in our films. The above magnetoconductance plots are fitted with the Hikami–Larkin–Nagaoka (HLN) formalism³¹, given by $\Delta G(B) = \tilde{A}e^2/2\pi h [\ln(B_\phi/B) - \psi(1/2 + B_\phi/B)]$, where $\psi(x)$ is the digamma function, e is electronic charge, and h is the Planck's constant. The temperature dependency of the fitting parameters, dephasing field (B_ϕ) and prefactor $\tilde{A}$, that denote the effective number of 2D conducting channels contributing to WAL is plotted in Fig. 2c. In this low-field range, the contribution of bulk states to the WAL response, determined by performing in-plane magnetoconductance measurements³², was found to be negligible without significantly affecting the parameters extracted from the HLN fitting. In TI, we have two 2D WAL channels corresponding to the bottom and top SSs. Proximity of one of the surface (top) with MI, which theoretically opens an exchange gap, can considerably weaken its WAL response. For the case of Se-capped devices, we find $\tilde{A} \sim 1.6$, which indicates good decoupling between the top and bottom 2D WAL channels. However, in the EuS-capped devices, we notice a drop in WAL channel by ~ 0.8 at 1.5 K suggesting additional localization response due to EuS capping (see Fig. 2c).

Our above findings of localization at the top $\text{Bi}_2\text{Se}_3/\text{EuS}$ interface can arise due to different possible reasons. To rule out its origin to any trivial response from the EuS layer, temperature dependence of resistivity of a trivial metal, Pt (5 nm), was measured that showed no change in behavior due to its proximity with EuS (2 nm) (see Supplementary Fig. 11). These observations limit our explanation for the origin of the above response to possible electron–electron interactions (EEI)³³ or MPE-induced EG opening¹⁹. In the case of EEI, the large g-factor of the SSs can activate a large Zeeman splitting from the exchange field of EuS and contribute to corrections in the conductivity. In the following discussions, we refer to the above localization in EuS-capped films, causing an upturn in film resistivity, to an MIT response of the SSs.

Interface band-bending effects

In TIs, the position of E_F at the surface can greatly depend on the relative concentration of bulk and surface defect density^{29,34}. A large mismatch in the surface and bulk E_F can lead to charge transfer and bending of bulk bands near the surface, resulting in a depleted (an accumulated) surface region in films with lower (higher) surface defect density^{35,36} (refer to Supplementary Note 4). Such surface band-bending response, creating interfacial electrostatic fields that shift the Dirac cone and position of the E_F at the interface, has been observed using angle-resolved photo-emission spectroscopy (ARPES)³⁷ and gated terahertz cyclotron resonance measurements³⁸. In our films, the high bulk carrier

Fig. 3 PAS and interface band-bending effects. **a** S-parameter as a function of positron beam energy for reference Bi_2Se_3 film and EuS-capped Bi_2Se_3 film. The mean implantation depth probed by the positron beam is shown on the top axis. VEPFIT deduced S vs. E_p curves are shown by solid lines. Inset shows schematic of band-bending profile near the surface of Bi_2Se_3 . The error bar in the figure shows the standard error of the mean. **b** R_{xx} vs. V_g of the $\text{Bi}_2\text{Se}_3(16\text{ nm})/\text{EuS}(3\text{ nm})$ device measured at different T_s . A second hysteresis response appears in positive V_g at and below 25 K (refer to Supplementary Fig. 6). The plots are shifted vertically for clarity.

density ($> 10^{19}/\text{cm}^3$) is expected to pin the bulk E_F in the CB. However, understanding surface band-bending effects can help locate the position of E_F at the interface and to qualitatively probe the origin of the above interfacial MIT response.

In our above effort, we performed depth-resolved positron annihilation spectroscopy (PAS) measurements at room temperature using variable low-energy positron beam that allowed profiling the vacancy-type defects^{39,40}, i.e., primarily the Se vacancies (see “Methods”). From the Doppler-broadened positron annihilation spectrum, S-parameter, defined as the ratio of the counts in 511 ± 1 keV peak region to the total counts in 511 ± 10 keV region, is deduced. The S-parameter characterizes the annihilation of positrons with low-momentum valence electrons, which is directly related to the concentration of vacancy defects. In Fig. 3a, we plot the S-parameter, extracted from the positron annihilation spectrum, vs. positron beam energy (E_p)/mean implantation depth for the reference Bi_2Se_3 and the $\text{Bi}_2\text{Se}_3/\text{EuS}$ films having a capping layer of AlO_x . Here, the effect of variation in the capping layer thickness for the two films has been included in the PAS analysis (see Supplementary Note 7). We observe that for very low positron energies, the S-parameter is higher for the reference Bi_2Se_3 films than the EuS-capped films. As the energy increases beyond ~ 1.25 keV, corresponding to an implantation depth of ~ 9 nm, the S-parameter for the two samples merge suggesting similar bulk response of the Bi_2Se_3 films (see

Fig. 4 Gate tunable interface magnetism with enhancement in T_c . **a** R_{xx} vs. T of $\text{Bi}_2\text{Se}_3(16\text{ nm})/\text{EuS}(3\text{ nm})$ device ($L = 1.5\text{ mm}$, $W = 1\text{ mm}$) measured at different V_g s. T_c^{bulk} denotes the T_c of the bulk EuS film, measured at negative V_g (depleted carriers at interface). Top left inset shows increase in T_{MIT} (square; shown by vertical dashed lines in main figure) at positive V_g s which is corroborated by the increase in n_{Hall} (circle). Enhanced interface magnetism, attributed to indirect exchange at positive V_g s, leads to large $\Delta R = R(1.5\text{ K}) - R(T_{\text{MIT}})$ peaking at $V_g = 50\text{ V}$ (bottom-right inset). **b** R_{xx} vs. T of $\text{Bi}_2\text{Se}_3(16\text{ nm})/\text{Se}(2\text{ nm})$ device at different V_g s. **c** Warm-up and cool-down plot of R_{xx} vs. T at $V_g = 50\text{ V}$ showing hysteresis response for $T < 19\text{ K}$ and $T > 25\text{ K}$ attributed to different T_c of bulk (19 K) and interface (33 K) EuS, respectively.

Supplementary Note 7 for detailed analysis). Variable energy positron fit (VEPFIT) software analysis⁴¹ was carried out by assuming a two-layer fitting, comprising of the Bi_2Se_3 film and the underneath SiO_2 substrate layer (see Supplementary Note 7). The above analysis to the experimental S vs. E_p curve yielded best fits by invoking a mean internal electric field in the Bi_2Se_3 film within the depth of 9 nm from the top TI surface. In the reference Bi_2Se_3 samples, a large positive interfacial electric field of $6.1 \times 10^{-4}\text{ V/nm}$ was extracted supporting the formation of a downward band bending (top right inset of Fig. 3a), as also observed in ARPES study of Bi_2Se_3 surfaces³⁷. On the contrary, in the EuS-capped films, an order of magnitude drop in the mean electric field is observed ($\sim 0.9 \times 10^{-4}\text{ V/nm}$). Considering the interfacial electric field to be proportional to the carrier density, we find that the surface carrier density at the $\text{Bi}_2\text{Se}_3/\text{EuS}$ interface (compared to bulk value of $\sim 4 \times 10^{13}/\text{cm}^2$) can fall by nearly an order of magnitude to $\sim 10^{12}/\text{cm}^2$. These interpretations are in agreement with density functional theory (DFT) studies that reveal Eu–Se bond formation at the $\text{Bi}_2\text{Se}_3/\text{EuS}$ interface to be more favorable²³ leading to a lower surface defect density. Such a condition support a nearly flat-band condition moving the interface E_F closer to the band edge (bottom inset of Fig. 3a, also see Supplementary Note 4).

In Fig. 3b, we show temperature dependence of longitudinal resistance (R_{xx}) vs. back-gate voltage (V_g) of the $\text{Bi}_2\text{Se}_3/\text{EuS}$

devices. Interestingly, a hysteresis response in R_{xx} is observed at all temperatures below 125 K, with similar response observed in four different sets of samples and with different film thickness (see Supplementary Note 6 and Supplementary Fig. 6). In comparison, the Se-capped devices showed no such behavior (see Supplementary Note 5 and Supplementary Fig. 5). Presence of such hysteresis response observed only in $\text{Bi}_2\text{Se}_3/\text{EuS}$ devices at low temperatures further supports our PAS analysis that the interface E_F lies near the band edge and drops into the band gap with cool down due to freezing of charge carriers. With the position of interface E_F in the band gap, V_g modulation leads to pinning/depinning effects at the band edge, responsible for the observed hysteresis in Fig. 3b. These findings may therefore suggest the origin of our interfacial MIT response to the localization affects around the Dirac point of the top SSs.

Effect of gate voltage on interface magnetism

The MIT response in our $\text{Bi}_2\text{Se}_3/\text{EuS}$ devices was further investigated in Fig. 4a by measuring R_{xx} vs. T at different V_g s. Here, at negative V_g , an insulating transition was observed at temperature, $T_{\text{MIT}} \sim 19\text{ K}$, which was enhanced to $\sim 32\text{ K}$ at positive V_g with a corresponding increase in the carrier density (top left inset of Fig. 4a). In comparison, the Se-capped devices showed no such response (Fig. 4b). Such an increase in T_{MIT} at positive V_g , which also makes the device conducting (seen by the downward shift of plots), at first seems counter-intuitive. However, this observation may be suggestive of conduction-electron-mediated ferromagnetic exchange between the Eu atoms strengthening the interface magnetism, with an enhancement in T_c of the EuS “interface layer”. This enhanced interface magnetism in our devices is resulting in a larger increase in resistance below T_{MIT} , peaking at $V_g \sim 50\text{ V}$ (bottom-right inset of Fig. 4a). Comparing with the warm-up data of R_{xx} vs. T (Fig. 4c), we observe a clear hysteresis in the low- and high-field range that supports the existence of different magnetic transition temperature of bulk (T_c^{bulk}) and interface ($T_c^{\text{interface}}$) EuS layer. This feature is also revealed in resistance plots ($V_g > 15\text{ V}$) in Fig. 4a with a point of inflection at 19 K, below which, the magnetic transition of EuS layers away from the interface causes the resistance to increase further. Similar increase in T_c of EuS, however, due to bulk electron doping has been previously reported^{42,43}. It is interesting to note that at large negative V_g (depleted interface), we observe a higher value of $T_{\text{MIT}} \sim 19\text{ K}$, i.e., 2 K above the bulk T_c of EuS (17 K) which may indicate the role of high spin-orbit strength of the Bi_2Se_3 SSs²⁰.

In EuS, magnetism arises from the overlap of the Eu 4f orbitals with the valence band below the E_F that leads to a weaker exchange coupling⁴⁴. DFT studies by Kim et al.²⁰ show that the SSs that weakly interconnect the conduction and valance bands can mediate exchange coupling between Eu atoms through the Ruderman–Kittel–Kasuya–Yosida (RKKY) type indirect interactions^{45–47}, with the exchange constant being sensitive to the position of E_F near the Dirac point. The observed gate-controlled tuning of T_{MIT} in our devices provides evidence for such an indirect exchange mechanism existing at the $\text{Bi}_2\text{Se}_3/\text{EuS}$ interface. Also, cooling the device in perpendicular magnetic fields (H_{FC}), at $V_g = 50\text{ V}$ showed a surprising decrease in T_{MIT} (or $T_c^{\text{interface}}$), with the increasing strength of H_{FC} (see Fig. 5). Such a response may arise because, here, the external magnetic field interacts not only with the localized 4f spins of Eu by direct interaction, but also through the conduction electrons at the interface. Such a theoretical framework of RKKY interactions that include the sign and value of the interaction potential between the localized spins and the polarized conduction electrons in external magnetic field has not been extensively explored⁴⁸. In addition, EEs in presence of Zeeman field can also greatly affect the above mentioned indirect interactions causing a reduction in $T_c^{\text{interface}}$ ⁴⁹.

Fig. 5 Effect of field cooling on the MIT response. R_{xx} vs. T of the $\text{Bi}_2\text{Se}_3(16 \text{ nm})/\text{EuS}(3 \text{ nm})$ device measured at $V_g = 50 \text{ V}$ with varying perpendicular H_{FC} s. The plots are shifted vertically for clarity. Bottom-right inset shows lowering of T_{MIT} with H_{FC} attributed to the weakening of the indirect exchange at the interface.

Fig. 6 Gate voltage dependence of Hall resistance. R_{xy} vs. V_g for $\text{Bi}_2\text{Se}_3(16 \text{ nm})/\text{EuS}(3 \text{ nm})$ device at 1.5 K.

Gate voltage dependence of Hall signal

Apart from the enhancement in T_{MIT} for $V_g > 15 \text{ V}$ in Fig. 4a, we also observe a prominent and reproducible drop in device resistance preceding MIT. This sudden drop in resistance shows a maximum of $\sim 10 \Omega$ at $V_g = 50 \text{ V}$. Also, when cooled in higher magnetic fields, the resistance drop preceding T_{MIT} starts to vanish (Fig. 5). Although the increase in R_{xx} due to magnetic proximity is in agreement with the localization effects observed from our WAL studies, the concomitant drop in R_{xx} at the onset of interface magnetism is not well explained based on any spin disorder or EEI models supporting WL mechanisms^{16,33,50}. In Fig. 6, we plot Hall resistance (R_{xy}) vs. V_g of the device, measured at 1.5 K, that showed a hysteresis response with the value rising above $V_g \sim 15 \text{ V}$ in the positive sweep cycle reaching $\sim 70 \text{ m}\Omega$ and dropping down at -27 V in the negative sweep cycle of V_g . Comparing with the data of Fig. 4a where the resistance drop in R_{xx} preceding T_{MIT} is observed only for $V_g > 15 \text{ V}$, we notice an increase in R_{xy} in the same V_g window (during the positive V_g sweep cycle) in Fig. 6 that might suggest a common phenomenon responsible for the above two responses. We have analyzed the possibility to associate the above origin to a weak signature of H-IQHE driven by the movement of E_F in the EG (see Supplementary Note 8). However, the weak experimental signal, possibly due to the large bulk conductivity, canted magnetism of EuS and finite temperature makes this analysis inconclusive and open for future exploration.

In conclusion, we report favorable growth conditions of EuS-proximitized Bi_2Se_3 films on a- SiO_2 substrate providing evidence of lower vacancy-type surface defect density that support the movement of the interface E_F toward the Dirac point. Also, we

observe localization response in the proximitized SSs below the T_c of EuS with a possible topological origin. Most importantly, we demonstrate the possibility to modulate interface magnetism using V_g , driven by an indirect RKKY interaction at the $\text{Bi}_2\text{Se}_3/\text{EuS}$ interface. These findings offer us the possibility to design a more robust surface quantum Hall system in our quest to develop switchable topological electronic devices.

METHODS

Growth, device fabrication, and transport measurement

Film growth was performed in a custom designed molecular beam epitaxy system with a base pressure of 5×10^{-10} mbar with all crucial interfaces prepared without breaking vacuum. Hall bar devices were mechanically patterned under a microscope and contacts were made using wire bonding on a 44 leadless chip carrier (refer to Supplementary Note 3). In addition, Hall bar geometry devices were also fabricated using in-situ shadow mask technique. Low-temperature magneto-transport measurements were performed in a variable temperature insert cryostat with a base temperature of 1.5 K.

Positron annihilation spectroscopy

Depth-resolved Doppler broadening measurements were carried out on the samples using the variable low-energy positron beam. Using this approach, vacancy-type defects were studied in our films starting from the surface up to a depth of a few hundreds of nm by implanting positron beam of varying energy ranging from 235 eV to 2.735 keV in steps of 0.1 keV. The mean implantation depth follows an empirical relation: $z \text{ (nm)} = (40 \times E_p^{1.6})/\rho$ where E_p is the positron beam energy in keV and ρ is the density of the sample in g/cm^3 . For the PAS study, Doppler broadened 511 keV annihilation energy spectrum was obtained for each positron beam energy using HPGe detector having an energy resolution of 1.5 keV for 662 keV γ -ray of ^{137}Cs . From each annihilation spectrum, the S-parameter was deduced.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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REFERENCES

- Hasan, M. Z. & Kane, C. L. Colloquium: topological insulators. *Rev. Mod. Phys.* **82**, 3045–3067 (2010).
- Qi, X. L. & Zhang, S. C. Topological insulators and superconductors. *Rev. Mod. Phys.* **83**, 1057–1110 (2011).
- Chen, Y. L. et al. Massive Dirac fermion on the surface of a magnetically doped topological insulator. *Science* **329**, 659–662 (2010).
- Yu, R. et al. Quantized anomalous Hall effect in magnetic topological insulators. *Science* **329**, 61–64 (2010).
- Chang, C.-Z. et al. Experimental observation of the quantum anomalous Hall effect in a magnetic topological insulator. *Science* **340**, 167–170 (2013).
- Gusynin, V. P. & Sharapov, S. G. Unconventional integer quantum Hall effect in graphene. *Phys. Rev. Lett.* **95**, 146801 (2005).
- Qi, X.-L., Hughes, T. L. & Zhang, S.-C. Topological field theory of time-reversal invariant insulators. *Phys. Rev. B* **78**, 195424 (2008).
- Essin, A. M., Moore, J. E. & Vanderbilt, D. Magnetoelectric polarizability and axion electrodynamics in crystalline insulators. *Phys. Rev. Lett.* **102**, 146805 (2009).
- Qi, X.-L., Li, R., Zang, J. & Zhang, S.-C. Inducing a magnetic monopole with topological surface states. *Science* **323**, 1184–1187 (2009).
- Mogi, M. et al. Tailoring tricolor structure of magnetic topological insulator for robust axion insulator. *Sci. Adv.* **3**, eaao1669 (2017).
- Hor, Y. S. et al. Development of ferromagnetism in the doped topological insulator $\text{Bi}_{2-x}\text{Mn}_x\text{Te}_3$. *Phys. Rev. B* **81**, 195203 (2010).
- Hou, Y., Kim, J. & Wu, R. Magnetizing topological surface states of Bi_2Se_3 with a CrI_3 monolayer. *Sci. Adv.* **5**, eaaw1874 (2019).
- Men'shov, V. N., Tugushev, V., Ereemeev, S., Echenique, P. & Chulkov, E. Magnetic proximity effect in the three-dimensional topological insulator/ferromagnetic insulator heterostructure. *Phys. Rev. B* **88**, 224401 (2013).

14. Luo, W. & Qi, X.-L. Massive Dirac surface states in topological insulator/magnetic insulator heterostructures. *Phys. Rev. B* **87**, 085431 (2013).
15. Ereemeev, S. V. et al. Magnetic proximity effect at the three-dimensional topological insulator/magnetic insulator interface. *Phys. Rev. B* **88**, 144430 (2013).
16. Ereemeev, S. V., Men'shov, V. N., Tugushev, V. V. & Chulkov, E. V. Interface induced states at the boundary between a 3d topological insulator Bi_2Se_3 and a ferro-magnetic insulator EuS. *J. Magn. Mater.* **383**, 30–33 (2015).
17. Wei, P. et al. Exchange-coupling-induced symmetry breaking in topological insulators. *Phys. Rev. Lett.* **110**, 186807 (2013).
18. Katmis, F. et al. A high-temperature ferromagnetic topological insulating phase by proximity coupling. *Nature* **533**, 513–516 (2016).
19. Yang, Q. I. et al. Emerging weak localization effects on a topological insulator–insulating ferromagnet Bi_2Se_3 -EuS interface. *Phys. Rev. B* **88**, 081407 (2013).
20. Kim, J., Kim, K.-W., Wang, H., Sinova, J. & Wu, R. Understanding the giant enhancement of exchange interaction in Bi_2Se_3 -EuS heterostructures. *Phys. Rev. Lett.* **119**, 027201 (2017).
21. Lang, M. et al. Proximity induced high-temperature magnetic order in topological insulator-ferromagnetic insulator heterostructure. *Nano Lett.* **14**, 3459–3465 (2014).
22. Liu, W. et al. Enhancing magnetic ordering in Cr -doped Bi_2Se_3 using high- T_c ferromagnetic insulator. *Nano Lett.* **15**, 764–769 (2015).
23. Lee, A. T., Han, M. J. & Park, K. Magnetic proximity effect and spin-orbital texture at the Bi_2Se_3 /EuS interface. *Phys. Rev. B* **90**, 155103 (2014).
24. Krieger, J. A. et al. Do topology and ferromagnetism cooperate at the EuS/ Bi_2Se_3 interface? *Phys. Rev. B* **99**, 064423 (2019).
25. Yang, X. et al. Light control of surface–bulk coupling by terahertz vibrational coherence in a topological insulator. *npj Quantum Mater.* **5**, 1–7 (2020).
26. Osterhoudt, G. B. et al. Charge transfer in EuS/ Bi_2Se_3 heterostructures as indicated by the absence of Raman scattering. *Phys. Rev. B* **98**, 014308 (2018).
27. Reeg, C., Loss, D. & Klinovaja, J. Metallization of a Rashba wire by a superconducting layer in the strong-proximity regime. *Phys. Rev. B* **97**, 165425 (2018).
28. Bansal, N. et al. Robust topological surface states of Bi_2Se_3 thin films on amorphous SiO_2/Si substrate and a large ambipolar gating effect. *Appl. Phys. Lett.* **104**, 241606 (2014).
29. Brahlek, M., Koirala, N., Bansal, N. & Oh, S. Transport properties of topological insulators: band bending, bulk metal-to-insulator transition, and weak anti-localization. *Solid State Commun.* **215**, 54–62 (2015).
30. Brahlek, M., Koirala, N., Salehi, M., Bansal, N. & Oh, S. Emergence of decoupled surface transport channels in bulk insulating Bi_2Se_3 thin films. *Phys. Rev. Lett.* **113**, 026801 (2014).
31. Hikami, S., Larkin, A. I. & Nagaoka, Y. Spin-orbit interaction and magnetoresistance in the two dimensional random system. *Prog. Theor. Phys.* **63**, 707–710 (1980).
32. He, H.-T. et al. Impurity effect on weak antilocalization in the topological insulator Bi_2Te_3 . *Phys. Rev. Lett.* **106**, 166805 (2011).
33. Wang, J. et al. Evidence for electron-electron interaction in topological insulator thin films. *Phys. Rev. B* **83**, 245438 (2011).
34. Veyrat, L. et al. Band bending inversion in Bi_2Se_3 nanostructures. *Nano Lett.* **15**, 7503–7507 (2015).
35. Benia, H. M., Lin, C., Kern, K. & Ast, C. R. Reactive chemical doping of the Bi_2Se_3 topological insulator. *Phys. Rev. Lett.* **107**, 177602 (2011).
36. Rakyta, P., Ujfalussy, B. & Szunyogh, L. Band bending at the surface of Bi_2Se_3 studied from first principles. *N. J. Phys.* **17**, 123011 (2015).
37. Hsieh, D. et al. A tunable topological insulator in the spin helical Dirac transport regime. *Nature* **460**, 1101–1105 (2009).
38. Jenkins, G. S. et al. Dirac cone shift of a passivated topological Bi_2Se_3 interface state. *Phys. Rev. B* **87**, 155126 (2013).
39. Amarendra, G., Viswanathan, B., Rao, G. V., Parimala, J. & Purniah, B. Variable low energy positron beams for depth resolved defect spectroscopy in thin film structures. *Curr. Sci.* **73**, 409–417 (1997).
40. Schultz, P. & Lynn, K. Interaction of positron beams with surfaces, thin films, and interfaces. *Rev. Mod. Phys.* **60**, 701–779 (1988).
41. Veen, A. V., Schut, H., de Vries, J., Hakvoort, R. A. & Ijpma, M. R. Analysis of positron profiling data by means of “VEPFIT”. *AIP Conf. Proc.* **218**, 171–198 (1991).
42. Holtzberg, F., McGuire, T. R., Methfessel, S. & Suits, J. C. Effect of electron concentration on magnetic exchange interactions in rare earth chalcogenides. *Phys. Rev. Lett.* **13**, 18 (1964).
43. Selinsky, R. S., Han, J. H., Pérez, E. A. M., Guzei, I. A. & Jin, S. Synthesis and magnetic properties of Gd doped EuS nanocrystals with enhanced Curie temperatures. *J. Am. Chem. Soc.* **132**, 15997–16005 (2010).
44. Coey, J. M. D., Viret, M. & Molnar, S. V. Mixed-valence manganites. *Adv. Phys.* **48**, 167–293 (1999).
45. Ruderman, M. A. & Kittel, C. Indirect exchange coupling of nuclear magnetic moments by conduction electrons. *Phys. Rev. C* **96**, 99 (1954).
46. Kasuya, T. A theory of metallic ferro- and antiferromagnetism on Zener's model. progress of theoretical physics. *Prog. Theor. Phys.* **16**, 45–57 (1956).
47. Yosida, K. Magnetic properties of Cu-Mn alloys. *Phys. Rev.* **106**, 893 (1957).
48. Balcerzak, T. The RKKY interaction in the presence of an external magnetic field. *Phys. Status Solid C* **3**, 212–215 (2006).
49. Ziener, C. H., Glutsch, S. & Bechstedt, F. RKKY interaction in semiconductors: effects of magnetic field and screening. *Phys. Rev. B* **70**, 075205 (2004).
50. Dzero, M., Xia, J., Galitski, V. & Coleman, P. Topological kondo insulators. *Annu. Rev. Condens. Matter Phys.* **7**, 249–280 (2016).

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AUTHOR CONTRIBUTIONS

S.M. and S.S. fabricated the devices and performed the measurements. A.B., S.S., and R.P. optimized and characterized the TI film growth. S.S. and S.C. setup the measurement facility. S.A. performed PAS measurements. B.S. performed TEM study and the analysis. S.S. and K.V.R. analyzed the data. K.V.R. conceived the idea. S.S. and K.V.R. wrote the paper. All authors contributed to the discussion.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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