



Emerging weak localization effects on a topological insulator–insulating ferromagnet (Bi_2Se_3 -EuS) interface

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Thin films of topological insulator Bi_2Se_3 were deposited directly on insulating ferromagnetic EuS. Unusual negative magnetoresistance was observed near the zero field below the Curie temperature T_C , resembling the weak localization effect; whereas the usual positive magnetoresistance was recovered above T_C . Such negative magnetoresistance was only observed for Bi_2Se_3 layers thinner than $t \sim 4$ nm, when its top and bottom surfaces are coupled. These results provide evidence for a proximity effect between a topological insulator and an insulating ferromagnet, laying the foundation for future realization of the half-integer quantized anomalous Hall effect in three-dimensional topological insulators.

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A topological insulator (TI) has a full energy gap in the bulk, and contains gapless surface states that cannot be destroyed by any nonmagnetic impurities. Because of time reversal symmetry, the surface states cannot be back-scattered by nonmagnetic impurities.^{1,2} When a thin magnetic layer is applied on the surface, a full insulating gap is opened, and an electric charge close to the surface is predicted to induce an image magnetic monopole.^{3,4}

Probably the most extensively studied three-dimensional TI (3D-TI) has been bismuth selenide (Bi_2Se_3),⁵⁻⁷ exhibiting crystal structure that consists of atomic quintuple layers (QLs), with three QLs forming a unit cell. As made, uncompensated samples typically have a Fermi level above the Dirac point and intersecting the bulk conduction band.^{8,9} In particular, low temperature transport measurements on ungated and uncompensated TI films show positive magnetoresistance (MR) at low magnetic fields and in a wide range of film thicknesses.¹⁰⁻¹² This was explained in terms of weak antilocalization (WAL) that results from spin-momentum locking on the surface state Dirac cone.^{10,13} While the inability to account for the bulk bands (presumably because of their low mobility) has challenged this simple assignment, the discovery of weak localization (WL) effects at higher fields^{12,14} and the ability to accurately separate quantum oscillation effects¹⁵ in high-quality films may provide a first step towards a more comprehensive understanding of transport in these systems.

By adopting topologically nontrivial Hamiltonians to describe these materials, various recent theories predict WL or negative MR near the zero field in a TI as a result of gap opening at its surface state Dirac point. The negative MR may arise from the surface state when the Fermi level is sufficiently close to the top of the gap.¹⁶ Alternatively, it can also be produced by bulk conduction and can only be observed when the surface conduction is sufficiently suppressed.^{17,18} Such a phenomenon was reported in cases of gated ultrathin Bi_2Te_3 films¹⁹ and magnetically doped Bi_2Se_3 films.²⁰

To further elucidate the uniqueness of transport in the surface state of TI materials, and as an initial step towards realizing half-integer quantized anomalous Hall effect (QAHE) and other applications, we studied the interface between a thin film TI (Bi_2Se_3) and an insulating ferromagnet (IF) (EuS). While above the Curie temperature T_C of the EuS we find positive MR that is observed ubiquitously in similar films, below T_C the MR becomes negative near the zero field, clearly indicating a proximity effect between the TI and the IF.

All bilayer (BL) samples presented in this Rapid Communication were grown using a pulsed laser deposition (PLD) method. For the bottom layer, high quality thin films of the well-studied insulating ferromagnet EuS with thicknesses 20–200 nm were grown on Al_2O_3 (0001) using targets made by spark plasma sintering (SPS) of high purity EuS powder. Atomic force microscopy (AFM) indicated better than 2 Å surface smoothness, and the crystallinity of the films was confirmed with X-ray diffraction (XRD).²¹ Magnetic properties of the films were studied with a superconducting quantum interference device (SQUID) magnetometer and a Sagnac interferometer.²² Their magnetizations were observed to have an appreciable perpendicular component [Fig. 1(a)] as required to realize the QAHE. T_C is estimated from a Curie-Weiss fit [Fig. 1(b)] to be 15.7 K. The local Kerr effect of the films at 10 K, using a scanning-Sagnac interferometer with 0.9 μm spatial resolution,^{22,23} indicates highly uniform magnetization [Fig. 1(e)]. Electric insulation ($R_{\square} > 20$ M Ω) was verified for 2 K $< T < 300$ K. Bi_2Se_3 thin films were then grown as top layers with quality that compares to that of films grown on nonepitaxial substrates such as Al_2O_3 . While more details on properties of PLD-grown Bi_2Se_3 and EuS films will be given elsewhere,^{21,24} we note here that the important features necessary for a reliable study of the TI-IF proximity effect, which are an insulating ferromagnet with a perpendicular anisotropy component, and a well defined interface to the TI material, are met in all our films. Specific

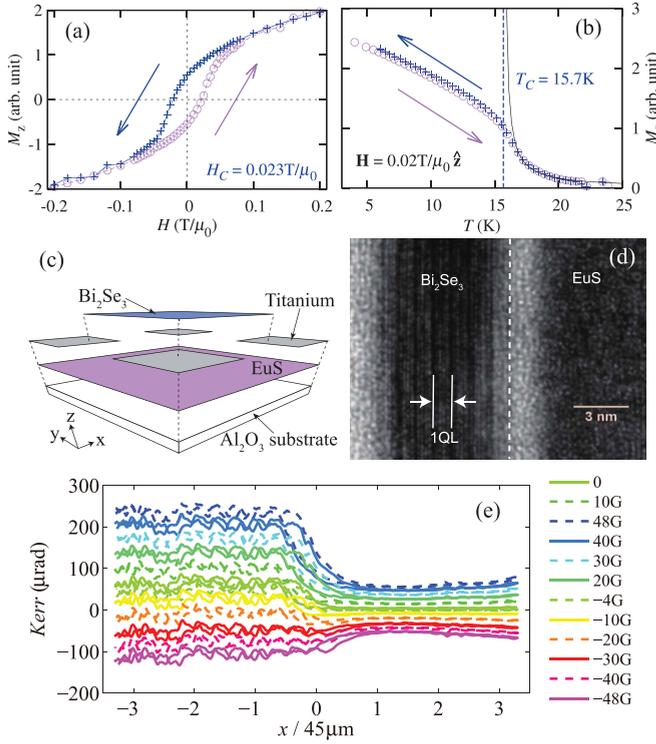


FIG. 1. (Color online) (a) Magnetization of an EuS thin film in perpendicular magnetic fields and (b) its dependence on temperature. The black solid curve is a fitting to the Curie-Weiss law. (c) Schematic of a bilayer device: an EuS film was deposited on an Al_2O_3 (0001) substrate, on top of it 15 nm titanium contacts with gradual height on the edges and a Bi_2Se_3 layer were sequentially deposited. (d) TEM image of the cross section of a bilayer device, showing the QL of Bi_2Se_3 and a smooth TI-IF interface. (e) Kerr angles measured at 10 K by a scanning Sagnac interferometer across the edge of the Bi_2Se_3 layer, where the $x < 0$ region is bare EuS and $x > 0$ is Bi_2Se_3 on top of EuS.

for the transport measurements, 15 nm-thick titanium was evaporated on corners of the device to serve as Ohmic contacts [Fig. 1(c)]. For shaping the layers, we used shadow masks to produce gradual height profiles and smooth overlapping near the edges of titanium contacts. The effectiveness was confirmed with scanning electron microscopy (SEM). The cross-section transmission electron micrographs [Fig. 1(d)] indicate smooth interface and excellent layering of the Bi_2Se_3 . For comparison, Bi_2Se_3 -only samples were made in each of the PLD sessions in which the Bi_2Se_3 of BLs were deposited, with Ohmic contacts between Bi_2Se_3 and bare Al_2O_3 (0001) substrates, i.e., an identical configuration to Fig. 1(c) except for the absence of an EuS layer.

To examine the relevance of surface conduction, samples with different thicknesses for the Bi_2Se_3 layer were fabricated and transport properties of two representative thicknesses are shown in Fig. 2. As ubiquitously seen in TI thin films, the PLD-grown Bi_2Se_3 -only samples show positive MR at low fields regardless of thicknesses, which broadens monotonically with increasing temperature [Figs. 2(a) and 2(b)], consistent with weak antilocalization (WAL). Fittings to the standard Hikami-Larkin-Nagaoka (HLN) formula describing the WAL magnetoconductance^{13,25,26} yield dephasing lengths

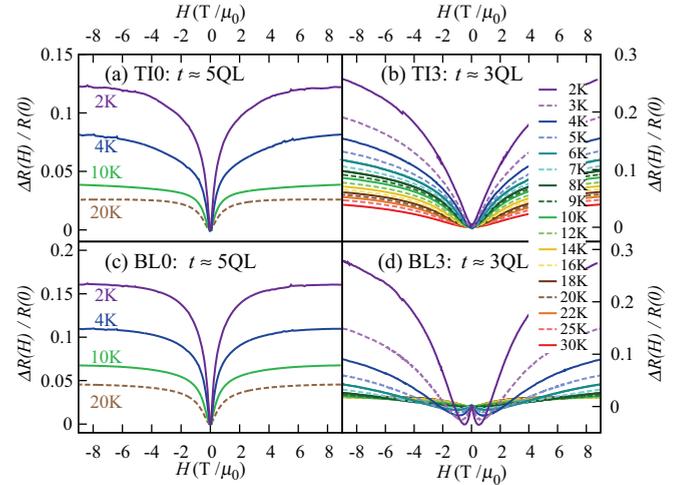


FIG. 2. (Color online) MR and its temperature dependence of PLD-grown Bi_2Se_3 thin films (TI) and Bi_2Se_3 -EuS BL devices. (a), (b) The Bi_2Se_3 -only samples have positive MR from WAL regardless of thicknesses. (c), (d) The BL devices with TI-layer thicknesses $t \gtrsim 4$ QL behave similarly to the TI-only films, whereas with TI-layer thicknesses $t \lesssim 4$ QL a distinctive WL-like negative MR is observed at low fields below the Curie temperature of EuS. The thickness limit coincides with the occurrence of coupling between the top and the bottom surfaces of a TI thin film.

l_ϕ comparable to molecular-beam epitaxy (MBE)-grown samples with the same thicknesses.^{11,14} For thicknesses of the Bi_2Se_3 layer greater than ~ 4 QL, the TI-IF bilayers have similar low-field MR features to their TI-only counterparts [Fig. 2(c)]. By contrast, for thicknesses $t \lesssim 4$ QL, the bilayers show distinctive negative low-field MR at low temperatures [Fig. 2(d)], resembling WL effects. Such negative MR features are clearly distinguishable well below the Curie temperature ($T_C = 15.7$ K) of the IF [Figs. 3(c) and 3(b)], whereas positive MR from WAL appears at higher fields. Below and close to T_C [Fig. 3(b)], the negative MR can no longer be directly observed. However, its remnant contribution reverses the thermal broadening of the overall positive MR. This suggests that the WL effect is reduced rapidly close to the ferromagnetic transition. Above T_C [Fig. 3(a)], the positive MR is eventually broadened when increasing the temperature, similar to common WAL features in TI-only thin films. The agreement between T_C and the temperatures at which WL becomes dominant strongly indicates a proximity effect between the IF and the TI. In Figs. 2 and 3 we presented the negative MR in the same sample, whereas four bilayer samples (labeled as BL1–4) with $t \lesssim 4$ QL from different growth batches all demonstrated such a proximity effect in a consistent manner. The thickness criterion ($t \lesssim 4$ QL) coincides with the thickness when the two surfaces of a Bi_2Se_3 film are observed to be coupled,⁹ suggesting a surface-originated WL mechanism.

Below 2 K [Fig. 3(d)], we observed a continuous sharpening of the low-field WL feature when lowering the temperature, as expected from diminishing thermal dephasing. Unexpectedly, the magnitude of negative MR was reduced when lowering the temperature below 1 K. This can be explained by the inhomogeneity observed in the Bi_2Se_3 layer grown on top

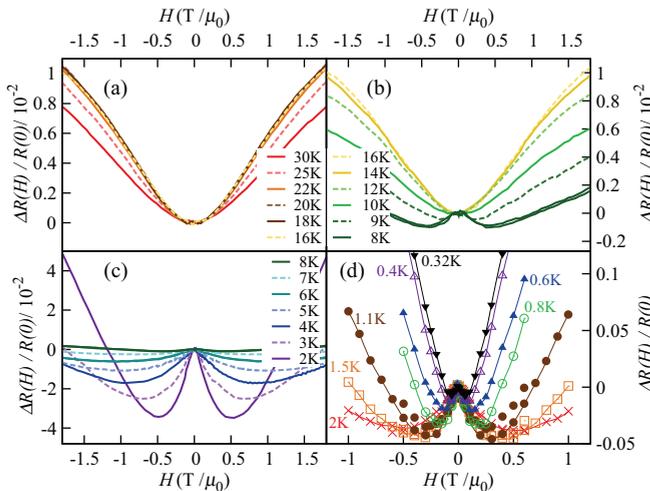


FIG. 3. (Color online) Low-field MR of a TI-IF bilayer device (BL3, $t \approx 3\text{QL}$) in magnetic fields perpendicular to the film. (a) Above T_C , a WAL-like positive MR sharpens with decreasing temperature. (b) Just below T_C , MR broadens with decreasing temperature. (c) Well below T_C , a WL-like negative MR emerges near the zero field. The emergence of WL below T_C of the EuS indicates a clear TI-IF proximity effect. (d) MR of BL3 below $T = 2\text{ K}$, with solid lines as illustrative guides. Resistance at $T \leq 0.8\text{ K}$ was measured with two-terminal configurations.

of EuS. While the Bi_2Se_3 thin films grown on bare substrates were verified by AFM and XRD to be adequately uniform in thickness, the TEM images taken at different locations on the cross section of the BL3 sample show large variations ($\pm 2\text{ QL}$) in thickness of the Bi_2Se_3 layer, with an estimated mean value consistent with the thickness of the Bi_2Se_3 -only film grown in the same PLD session. For the thicknesses of interest ($t \lesssim 4\text{ QL}$), both the resistance of Bi_2Se_3 films and its temperature dependence are known to change sharply with thickness at low temperatures.¹¹ Thus special difficulty is introduced when measuring the sheet resistance with a van der Pauw method, where the sample thickness is assumed to be uniform.^{27,28} With such inhomogeneous geometry, electric conduction is limited by the thinner and therefore more resistive parts of the sample. Indeed, the sheet resistance of the BL samples are one order of magnitude higher than that of the Bi_2Se_3 -only samples with similar thicknesses [Fig. 4(b)]. When lowering the temperature, the change in the zero-field resistance $R(0)$ is dominated by the sharp temperature dependence of the thinner parts of the sample. The low-field features in magnetoresistance, however, are mainly contributed by the thicker parts, where the dephasing length is known to be longer.¹¹ Therefore, after dividing by $R(0)$ as the normalization factor, $\Delta R(H)/R(0)$ appears with a reduced magnitude. On the other hand, measuring the Hall effect, which is insensitive to film inhomogeneity,²⁹ the sheet carrier density is calculated showing similar values ($n_{2D} \approx 2 \times 10^{13}\text{ cm}^{-2}$) for both TI-only films and TI-IF bilayers and are very close to reported values for MBE films.¹¹

So far in our discussions, we assumed that the low-field negative magnetoresistance has an orbital origin such as WL effects suggested by recent theories.^{16–18} The presence of a magnetic material brings possibilities of MR originated from

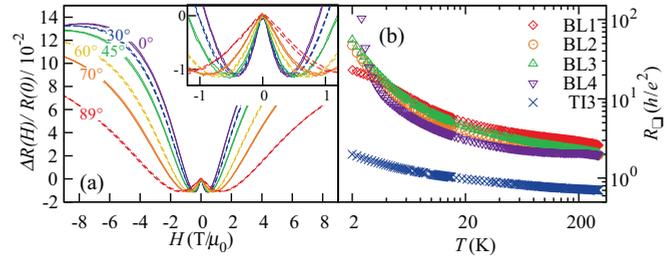


FIG. 4. (Color online) (a) The WL and WAL features in BL4 broaden with increasing angles between the magnetic field and the normal of the sample plane, confirming their orbital origins. The inset is a zoomed-in version of low-field features. (b) Sheet resistances of the four TI-IF bilayer samples with WL features (BL1–4) and a PLD-grown TI-only film (TI3) measured with the van der Pauw method. The nonvanishing MR near parallel fields and the high resistances of the BL samples are consistent with the observed inhomogeneity in sample thickness.

scatterings at localized spins.³⁰ However, the magnitude of any spin-introduced MR should be greater above T_C and should diminish rapidly below T_C as the spins being aligned during the ferromagnetic transition,³¹ which is contrary to what we observed. Moreover, we show in Fig. 4(a) the broadening of the negative MR as the sample is rotated from perpendicular to near-parallel applied magnetic field, clearly indicating its orbital origin. The fact that some negative MR features remain finite in the near-parallel field is likely due to the uneven thickness in the Bi_2Se_3 layer as a consequence of a locally slanted top surface, on which local electron transport may have a finite angle to the magnetic field that is parallel to the sample plane. In fact, when the magnetic field is scaled with an effective angle θ^* between the magnetic flux and the normal of local electron transport, both the WL at low fields and the WAL at higher fields are found to coincide well with the perpendicular-field MR. The difference between the effective angles θ^* and the nominal angles read from the instrument (θ) is small for most angles ($|\theta - \theta^*| \leq 3^\circ$ for $\theta \leq 60^\circ$) and increasing towards the parallel field ($\theta^* \approx 75^\circ$ for $\theta = 89^\circ$). This is consistent with expectations from a film with a smooth bottom surface and an uneven top surface.

Theory predicts a gap opened at the Dirac point as a result of the proximity between the TI and the IF.³² Such a gap opening is expected to result in WL only when the Fermi level is sufficiently near the gap.^{16–18} However, as is common in ungated and uncompensated Bi_2Se_3 , our carrier densities suggest a Fermi level intersecting the conduction band and, therefore, far away from the Dirac point.³³ Recent calculations suggest that surface charges on the IF layer may result in band bending at the interface,³⁴ hence providing a possible mechanism to move the gap towards the Fermi level. However, whether this is the case depends on the details of atomic arrangement at the interface,³⁵ which cannot be determined with available data. On the other hand, low levels of sulfur doping are known to modify the band structure of Bi_2Se_3 while preserving its structural phase.^{36,37} Such an effect may as well be present at the Bi_2Se_3 -EuS interface. Attempts to perform angle-resolved photoemission spectroscopy (ARPES) measurements to determine the band gap, the location of the Dirac point, and the Fermi level produced inconclusive results,

primarily due to the inhomogeneity in films thicknesses. We note that a recent study of $\text{Bi}_2\text{Se}_3/\text{EuS}$ bilayers shows a weak tendency towards the anomalous Hall effect, which was argued to indicate the emergence of a ferromagnetic phase in TI.³⁸ Samples in that study consisted of a bottom Bi_2Se_3 film and a top EuS film, making it impossible to determine whether the EuS is truly insulating. We contrast it with the present approach in which we first obtained high-quality, well characterized EuS films, on which the TI film was deposited. Since a true 3D TI will have one and only one surface state, irrespective of inhomogeneities and local defects, we prefer this approach and further argue that the unusual negative magnetoresistance

at low fields below the Curie temperature of the ferromagnet is a strong indication of a proximity effect between a topological insulator and an insulating ferromagnet.

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