Proximity-induced magnetism and an anomalous Hall effect in Bi$_2$Se$_3$/LaCoO$_3$: a topological insulator/ferromagnetic insulator thin film heterostructure†

Shanna Zhu, a,b Dechao Meng, c,d Genhao Liang, c Gang Shi, a,b Peng Zhao, a,b Peng Cheng, a Yongqing Li, a Xiaofang Zhai, c,e Yalin Lu, c,e,f Lan Chen, a,b* and Kehui Wu*a,b,g

Inducing magnetism in a topological insulator (TI) by exchange coupling with a ferromagnetic insulator (FMI) will break the time-reversal symmetry of topological surface states, offering possibilities to realize several predicted novel magneto-electric effects. Seeking suitable FMI materials is crucial for the coupling of heterojunctions, and yet is challenging as well and only a few kinds have been explored. In this report, we introduce epitaxial LaCoO$_3$ thin films on a SrTiO$_3$ substrate, which is an insulating ferromagnet with a Curie temperature of $T_C \sim 85$ K, to be combined with TIs for proximity coupling. Thin films of the prototype topological insulator, Bi$_2$Se$_3$, are successfully grown onto the (001) surface of LaCoO$_3$/SrTiO$_3$ forming a high-quality TI/FMI heterostructure with a sharp interface. The magnetic and transport measurements manifest the emergence of a ferromagnetic phase in Bi$_2$Se$_3$ films, with additional induced moments and a suppressed weak antilocalization effect, while preserving the carrier mobility of the intrinsic Bi$_2$Se$_3$ films at the same time. Moreover, a signal of an anomalous Hall effect is observed and persists up to temperatures above 100 K, paving the way towards spintronic device applications.

1. Introduction

Three-dimensional (3D) topological insulators (TIs), as represented by Bi$_2$Se$_3$ family compounds, are a category of electronic materials characteristic of bulk valence and conduction bands connected by conducting surface states. The exotic gapless surface states are topologically protected and with locked spin-momentum, making the TIs fascinating in both theoretical research and potential electronic applications. Various intriguing consequences of the TME effect have been proposed, including the image magnetic monopole induced by a point charge, giant magneto-optical Kerr effect and universal Faraday effect, half-integer surface quantum Hall effect, and so on.

The key to opening an energy gap in the surface state of TIs and thus observing the topological magnetoelectric response lies in the breakdown of time-reversal symmetry (TRS) at the surface of TIs. Experimentally, breaking the TRS can be accomplished either by doping the TIs with magnetic atoms or by exchange-coupling with magnetic layers through a proximity effect. The former method has been widely explored with magnetic dopants from 3d transition metals (Mn, Fe, V, and Co) or rare-earth metals (such as Gd and Dy), where ferromagnetism in TIs has...
been observed. However, the strong scattering brought by random magnetic dopants inevitably introduces crystal defects and lowers the carrier mobility.\textsuperscript{30,32}

On the other hand, fabricating TI/magnetic insulator (MI) heterostructures is an alternative way to acquire magnetic order in TIs via the proximity effect, without introducing any extra impurities.\textsuperscript{12,17,18,38} The magnetic proximity effect is a well-known phenomenon that originates from interfacial exchange coupling.\textsuperscript{50} Compared with doping, the advantages of the heterostructure approach include better controllability of the electronic states,\textsuperscript{17} preservation of the TI's original crystalline structure,\textsuperscript{17} and bulk properties,\textsuperscript{34} enhanced temperature of the magnetic ordering,\textsuperscript{7,34,50} and so on. Moreover, some effects such as the half-integer quantum anomalous Hall effect (QAHE) can be observed only in a TI/MI heterostructure.\textsuperscript{15,18} Recently, a number of studies have been devoted to this method, including several theoretical studies\textsuperscript{12,16,17,31,52} and experimental demonstrations combining TIs with MI materials of ferromagnetic EuS,\textsuperscript{32–35} and GdN,\textsuperscript{36} ferrimagnetic Y$_2$Fe$_2$O$_5$(YIG),\textsuperscript{37–41} BaFe$_2$O$_4$,\textsuperscript{42,43} Fe$_3$O$_4$,\textsuperscript{44} CoFe$_2$O$_4$,\textsuperscript{45} and Tm$_3$Fe$_5$O$_{12}$(TIG),\textsuperscript{46} as well as antiferromagnetic MnSe,\textsuperscript{47} CrSb,\textsuperscript{48} and NiO.\textsuperscript{49} In spite of the above progress, the exploration is limited to a small number of materials. New kinds of ferromagnetic insulators (FMIs) with preferable properties are desirable for the fabrication of TI/FMI heterostructures. Unfortunately, the existing ferromagnetic insulators (FMIs) are very rare, and usually have in-plane magnetization for a thin film configuration.\textsuperscript{53} The main challenge resides in finding better magnetic materials, which can form a high-quality interface with TIs and, at the same time, provide a strong exchange coupling at the interface.\textsuperscript{12,51,52}

LaCoO$_3$ (LCO) is a transition metal oxide-based perovskite material with peculiar properties. Although bulk LCO is diamagnetic at low temperatures, LCO thin films epitaxially grown on SrTiO$_3$(STO) or (La,Sr)(Al,Ta)O$_3$ (LSAT) exhibit a ferromagnetic ground state below 85 K, due to a spin-state transition induced by the tensile strain.\textsuperscript{54–56} Luckily, the strained LCO thin film is a good insulator below its Curie temperature.\textsuperscript{55–57} In addition, the magnetism from the d electrons (Go$^{3+}$) in LCO is expected to have a stronger exchange coupling with the p electrons in TIs compared with the f electrons in rare earth elements (such as Eu$^{2+}$, Gd$^{3+}$, and Tm$^{3+}$),\textsuperscript{12} and thus may be beneficial for the proximity interaction.

In this work, we report the fabrication and magnetotransport properties of a new TI/FMI heterostructure combining Bi$_2$Se$_3$ and strained LaCoO$_3$ thin films on a SrTiO$_3$ (001) substrate. We show that Bi$_2$Se$_3$ thin films with a uniform morphology and high carrier mobility, similar to those grown on a bare STO substrate, can be successfully grown on the surface of LCO/STO by molecular beam epitaxy (MBE). The transport measurements reveal that the weak antilocalization effect (WAL) of Bi$_2$Se$_3$ is much suppressed by the induced magnetism through proximity coupling with LCO, and the anomalous Hall resistance is observed to persist up to over 100 K. Furthermore, the magnetization data confirm that there are extra induced magnetic moments in the Bi$_2$Se$_3$ layer which can survive to a temperature higher than the Curie temperature of LCO/STO. Such enhanced ferromagnetism in TI/FMI heterostructures will be promising for high-temperature device applications.

2. Experimental methods

2.1 Thin film growth

LaCoO$_3$ thin films were grown epitaxially on the (001)-oriented SrTiO$_3$ substrates by pulsed-laser deposition at 750 °C. Commercial undoped STO (001) single crystals with a size of 5 × 5 × 0.5 mm$^3$ (one-side polished) were used as the initial substrates. They were treated by a standard cleaning procedure, including acid-etching and ultrasonic rinse steps followed by annealing in a pure O$_2$ atmosphere to obtain an atomically smooth surface without inducing noticeable oxygen vacancies. The as-treated STO (001) crystals were then used for the growth of LaCoO$_3$ films and Bi$_2$Se$_3$ films subsequently.

The growth of Bi$_2$Se$_3$ thin films was carried out by using a home-made MBE system in an ultrahigh vacuum (UHV) chamber with a base pressure less than 2.0 × 10$^{-10}$ Torr, equipped with the reflection high-energy electron diffraction (RHEED) facility. The as-prepared LCO/STO (001) samples were transferred \textit{ex situ} into the UHV chamber and were degassed at 300 °C for about 1 h before growth. During growth, high purity Bi (99.997%) and Se (99.999%) sources were co-evaporated from the standard Knudsen cells, while the substrate temperature was set in the range from 140 °C to 280 °C. The fluxes of Bi and Se were calibrated with a quartz crystal microbalance thickness monitor and the flux ratio of Bi/Se was fixed to 1/10, in order to reduce the amount of Se vacancies. The typical growth rate of the Bi$_2$Se$_3$ thin films was about 0.3 nm per minute for this work. \textit{In situ} real-time RHEED patterns were recorded during the growth process. In addition, Bi$_2$Se$_3$ films were also grown on the as-treated STO (001) substrates under the same growth conditions, as control samples for comparison.

2.2 Characterization

The surface morphology of the LCO/STO (001) and Bi$_2$Se$_3$ thin films were characterized with an atomic force microscope (AFM) in tapping mode. The crystal structure was detected by X-ray diffraction (XRD) using a single crystal X-ray diffractometer (PANalytical) with Cu K$_\alpha$ radiation of wavelength 1.5406 Å in the $\omega$–$2\theta$ scan. The magnetization data were measured with a superconducting quantum interference device (SQUID) magnetometer.

For transport measurements, the Bi$_2$Se$_3$/LCO/STO(001) samples were patterned into Hall bars with a channel width of about 500 μm. Platinum wires pasted with silver epoxy were used as the electrical contact. The fabricated devices were then placed in a cryostat system with a temperature down to $\sim$1.7 K and a magnetic field up to $\pm$9 T. The current was along the channel (x axis) and the magnetic field was applied in a perpendicular direction (z axis). Several batches of samples grown
under different conditions were tested, and the typical results are given below. A thickness of 20 QL was chosen for the Bi$_2$Se$_3$ films, well above the 6 nm threshold, below which the top and bottom surfaces of the TIs may be coupled by quantum tunneling and result in a hybridization gap near the Dirac point.58 We also carried out transport measurements of the Bi$_2$Se$_3$/STO control samples, to provide the reference data with non-magnetic substrates.

3. Results and discussion

The schematic of our TI/FMI heterostructure device is depicted in Fig. 1(a). Three layers from top to bottom are the TI (Bi$_2$Se$_3$) film, FMI (LaCoO$_3$) film, and the SrTiO$_3$ (001) substrate, respectively, with their thickness shown in the graph. Fig. 1(b) shows the typical AFM image of the surface morphology of a 7.5 nm-thick LaCoO$_3$ film. Large-scale, homogeneous surface and atomically flat terraces are presented. The root-mean-square (RMS) roughness of the entire surface measured in Fig. 1(b) is 0.11 nm, with each terrace at an average height of 3.76 Å, corresponding to the height of a LaCoO$_3$ unit cell (u.c.). Fig. 1(c) shows the AFM image of a 20 nm Bi$_2$Se$_3$ film grown on LCO/STO (001) under the optimal growth conditions, at a substrate temperature of about 150 °C. Characteristic pyramid-shaped islands, with quintuple-layer (QL) steps and terraces are observed. Another sample of Bi$_2$Se$_3$ on LCO/STO(001) with a reduced thickness of 10 nm is shown in Fig. 1(d), with a similar morphology but flatter islands. Although the triangular terraces are not very large because of the relatively low growth temperature used here, the integral morphology is well-ordered and uniform from a large-scale scan (see Fig. S1(a) in the ESL†). Moreover, this morphology is almost the same as that of the Bi$_2$Se$_3$ samples grown on the bare STO (001) substrates under the same conditions, as shown in Fig. S1(b).† A QL-by-QL growth mode is observed from the RHEED pattern by a periodic oscillation of the diffraction intensity during film growth. Bright and straight stripes in Fig. 1(e) indicate a clean and flat surface of LCO/STO(001) before Bi$_2$Se$_3$ growth, while clear and sharp stripes in Fig. 1(f) indicate the highly crystalline and smooth surface of Bi$_2$Se$_3$ films.

For the XRD patterns shown in Fig. 2, four samples, including Bi$_2$Se$_3$ on STO(001), Bi$_2$Se$_3$ on LCO/STO(001), the as-prepared LCO/STO(001) and as-treated STO(001), are displayed on a log plot by a vertical offset from up to down. In the figure, the Bi$_2$Se$_3$ film exhibits the (00$l$) type of reflections over the 10°–80° scan range without the other sets of peaks of bulk Bi$_2$Se$_3$, indicating high crystallinity along the growth direction. The Bragg peak positions of the Bi$_2$Se$_3$ film grown on LCO/STO(001) agree very well with the Bi$_2$Se$_3$/STO(001) control sample as well as the ICSD standard card for Bi$_2$Se$_3$ single crystals, suggesting that the Bi$_2$Se$_3$...
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The initial electron densities are in the range of $2.5 \times 10^{13}$ cm$^{-2}$ (corresponding $n_{2D} = 1.4 \times 10^{19}$ cm$^{-2}$) and a mobility of $\mu = 659$ cm$^2$ V$^{-1}$ s$^{-1}$. It is important to note that the carrier mobility is as high as those of the non-magnetic control samples, indicating that the pristine good transport properties of the undoped TI has been preserved in our TI/FMI heterostructure. This is in good contrast to those doped TI samples with magnetic atoms where the carrier mobilities are seriously damped by scattering with dopants. The mobility is also improved compared to some other reported TI/FMI systems.

In Fig. 3(a), we plot the temperature-dependent carrier mobility of a typical Bi$_2$Se$_3$/LCO sample grown under optimal conditions, as well as the corresponding longitudinal resistance in a zero magnetic field ($H = 0$). The longitudinal resistance decreases gradually when the sample is cooled down from 300 K, and has a minimum resistivity of 0.7 m$\Omega$ cm around 10 K. This metallic-like behavior at high temperatures is widely observed in TI materials. While the carrier density is not sensitive to temperature when measured below 100 K, the mobility has a clear dependence on temperature. From the relation $\mu(T) = R_{xx}(T)/\rho_{xx}(T)$, where $R_{xx}$ is the Hall coefficient, the mobility is inversely proportional to the longitudinal resistivity $\rho_{xx}$, resulting in the nearly inverse dependence of mobility to temperature.

The magnetoresistance (MR) data provide another means to probe the TI’s properties. Fig. 3(b) depicts the MR of a Bi$_2$Se$_3$/LCO sample with the highest mobility that is grown at an optimal $T_{sub}$ of 150 °C. The MR, defined as $MR = [\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0)$, is positive for all curves measured here. For the curve taken at $T = 1.7$ K, the MR is characterized by a cusplike shape near the zero field due to the weak antilocalization (WAL) effect, which is characteristic of strong spin–orbit coupled TI materials. While the temperature rises, MR gradually decreases, and the cusp-shaped feature is weakened as well, and completely disappears at $T = 100$ K, leaving a parabola-shaped MR which reduces to the classical quadratic relationships. The trend of a weakened WAL effect with increasing temperature is similar to that of the Bi$_2$Se$_3$/STO samples, indicating the preservation of the intrinsic characteristics of TIs in our TI/FMI heterostructures.

Fig. 3(c) shows the normalized MR data of several Bi$_2$Se$_3$/LCO samples grown at different $T_{sub}$ together with two Bi$_2$Se$_3$/STO control samples measured at 1.7 K. They have an overall positive cusp-shaped MR in the low-field range of ±20 kOe. However, compared with the sharper cusp of non-magnetic control samples, the MR of the Bi$_2$Se$_3$/LCO samples is significantly reduced, meaning that the WAL effect is suppressed due to the coupling with the underlying magnetic layer. It is worth mentioning that for all the samples with growth conditions in a wide range from 150 °C to 225 °C, the reduced MR of Bi$_2$Se$_3$/LCO samples is always observed with comparison to their non-magnetic counterparts, while their difference becomes even larger at the higher $T_{sub}$ range (see the light blue line and dark blue line plotted in the figure). In addition, the MR curves of the Bi$_2$Se$_3$/LCO and optimal $T_{sub} = 150$ °C, we have a carrier density of $n_{2D} = 2.7 \times 10^{13}$ cm$^{-2}$ (corresponding $n_{1D} = 1.4 \times 10^{19}$ cm$^{-2}$) and a mobility of $\mu = 659$ cm$^2$ V$^{-1}$ s$^{-1}$. It is important to note that the carrier mobility is as high as those of the non-magnetic control samples, indicating that the pristine good transport properties of the undoped TI has been preserved in our TI/FMI heterostructure. This is in good contrast to those doped TI samples with magnetic atoms where the carrier mobilities are seriously damped by scattering with dopants. The mobility is also improved compared to some other reported TI/FMI systems.

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![Image](https://example.com/image.png)

**Fig. 2** X-ray diffraction spectrum of a 20 nm Bi$_2$Se$_3$ film on LaCoO$_3$/SrTiO$_3$(001) (blue line) and the data of three other samples for comparison. The inset is a zoom-in view of the (006) peak of the Bi$_2$Se$_3$ sample, which shows well-defined Kiessig fringes, indicating a smooth surface of the Bi$_2$Se$_3$ layer and a sharp interface between the film and the substrate.

<table>
<thead>
<tr>
<th>Sample $T_{sub}$ (°C)</th>
<th>Bi$_2$Se$_3$/LCO/STO</th>
<th>Bi$_2$Se$_3$/STO</th>
</tr>
</thead>
<tbody>
<tr>
<td>150</td>
<td>175</td>
<td>225</td>
</tr>
<tr>
<td>200</td>
<td></td>
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<tr>
<td>225</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$n_{2D}$ ($\times 10^{13}$ cm$^{-2}$)</td>
<td>2.70</td>
<td>2.19</td>
</tr>
<tr>
<td>$\mu$ (cm$^2$ V$^{-1}$ s$^{-1}$)</td>
<td>659</td>
<td>534</td>
</tr>
</tbody>
</table>

**Table 1** Carrier density ($n_{2D}$) and mobility ($\mu$) of several Bi$_2$Se$_3$/LCO/STO samples grown at different substrate temperatures (denoted by $T_{sub}$), ranging from 150 °C to 225 °C, as well as the two control samples of Bi$_2$Se$_3$/STO at $T_{sub} = 150$ °C and 225 °C, respectively. All data were measured at 1.7 K.


Bi$_2$Se$_3$/STO samples have different curvatures up to a high-field range, which further distinguishes them in magneto-transport properties. In order to gain further insight into the suppression of the WAL effect in our Bi$_2$Se$_3$/LCO samples, we carried out a quantitative analysis of the low-field magneto-conductance (MC) data. Magnetoconductivity is defined as 

$$\Delta \sigma(H) = \sigma_{xx}(H) - \sigma_{xx}(0),$$

where the 2D sheet conductance ($\sigma_{xx}$) is calculated by the relation

$$\sigma_{xx} = \frac{\rho_{xx}}{\rho_{yx}^2 + \rho_{xx}^2}.$$  

As proposed in the literature, the WAL quantum corrections to the low-field magnetoconductivity of Bi$_2$Se$_3$ thin films can be described by the Hikami-Larkin-Nagaoka (HLN) equation:

$$\Delta \sigma(H) \approx -\frac{e^2}{2\pi\hbar} \left[ \psi\left(\frac{1}{2} + \frac{H_{\phi}^2}{H^2}\right) - \ln\left(\frac{H_{\phi}^2}{H^2}\right) \right],$$

where $\psi(x)$ is the digamma function, $H_{\phi} = \frac{B_{\phi}}{\mu_0} = \frac{1}{\mu_0} \frac{\hbar}{4eI_{\phi}^2}$ is the dephasing field, and $I_{\phi}$ is the dephasing length. The coefficient $\alpha = 1/2$ is used for a system with a single coherent channel, which is also expected for the electron transport on one surface of a 3D TI with a single Dirac cone.

As plotted in Fig. 3(d), the magnetoconductivity of the samples has an inverted shape as that in Fig. 3(c), with the cusp pointing upwards. The overall negative magnetoconductivity is a key signature of the WAL effect. The two Bi$_2$Se$_3$/STO control samples have sharper apices near the zero field and separated far from the four Bi$_2$Se$_3$/LCO samples with the suppressed WAL effect. Fitting the data with the HLN equation, we find that $\alpha$ takes a value very close to 0.5 for Bi$_2$Se$_3$/STO, whereas for Bi$_2$Se$_3$/LCO, $\alpha$ distributes in the range of 0.35 to 0.43. The smaller $\alpha$ suggests more magnetic scattering at the interface of Bi$_2$Se$_3$/LCO, while in the limit of strong magnetic scattering, $\alpha \sim 0$ is expected. For the second parameter $I_{\phi}$, the fitted values for the magnetic and non-magnetic samples are more different. For Bi$_2$Se$_3$/LCO, with $T_{\text{sub}}$ increasing from 150 °C to 225 °C, $I_{\phi}$ takes the values of 259 nm, 218 nm, 197 nm, and 147 nm, respectively. While for the two Bi$_2$Se$_3$/STO control samples, $I_{\phi}$ yields 417 nm and 799 nm, much bigger than those of the magnetic samples. The significantly reduced coherence length in the Bi$_2$Se$_3$/LCO samples suggests that the WAL effect is suppressed by the magnetic proximity coupling to a great extent.

Although the total Hall resistance $R_{xy}$ has a nearly linear dependence on the magnetic field, by carefully examining the data, some abnormal signals have been revealed. Note that the estimated carrier density is much larger than $0.5 \times 10^{13}$ cm$^{-2}$, the limit that the two surfaces in the topological regime can only contain. This suggests that the Fermi level $E_F$ must be located inside the conduction band, such that a significant number of bulk electrons participate in the Hall measurement, responsible for the overall linear behavior of the $R_{xy}$.
analyze the signals behind it, we removed the dominant linear Hall background from the overall $R_{xy}$. The non-linear part of $R_{xy}$ can thus be extracted, as denoted by $R_{AHE}$ and plotted in Fig. 4(a). For the Bi$_2$Se$_3$/LCO sample, a clear nonlinear component is presented, whose shape resembles the magnetic hysteresis loops of LCO/STO shown in Fig. 4(b). The saturated maximum of $R_{AHE}$ measured at 1.7 K approaches $0.5 \Omega$, and the corresponding saturation field is consistent with that in the M–H loops of LCO/STO. This implies a direct correlation between the $R_{AHE}$ and magnetization of the LCO/STO substrate.

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Further discussion about the $R_{AHE}$ will be given later. In contrast, the extracted part of the Bi$_2$Se$_3$/STO control sample does not show any visible features of non-linearity even at 1.7 K. The distinct difference confirms that the nonlinear part of the Hall resistance comes from the induced magnetism in the Bi$_2$Se$_3$ layer through the interaction with the adjacent LCO FMI layer.

It is known that an ordinary Hall effect occurs in a nonmagnetic conductor, where the Hall resistance $R_{xy}$ is proportional to the applied perpendicular field $H_z$. While in a ferromagnetic (FM) conductor, the $R_{xy}$ initially increases steeply in weak $H_z$ but saturates at a larger value that is nearly $H_z$ independent. This is known as the anomalous Hall effect (AHE). An empirical relation for the total Hall effect in FM conductors is given by $R_{xy} = R_0 H_z + R_s M_z$, where the second term is the anomalous Hall resistance ($R_{AHE}$) which is due to spontaneous magnetization. The observed $R_{AHE}$ in Fig. 4(a) thus comes from the contribution of the AHE. However, it should be noted that there are other possibilities that might result in a nonlinear component of $R_{xy}$ other than the proximity-induced ferromagnetism in the TI layer. One possibility is the coexistence of multiple types of carrier. This would happen when the Fermi level is in the vicinity of the Dirac point where both electrons and holes are present. Thus, the scenario can be ruled out by the fact that $E_F$ locates inside the conduction band,
indicating that the Bi$_2$Se$_3$ film is in the single carrier type regime. The second possibility comes from the response of the underlying LCO/STO ferromagnetic layer, which might become conducting itself. This is also excluded since both the LCO and STO under-layers are well insulating with the resistivity $\rho > 10^5$ $\Omega$ cm under 100 K as reported previously $^{55-57}$ which is larger than that of the Bi$_2$Se$_3$ film by an order of 10$^7$. Besides, since the growth temperature is at $\sim$750 °C for LCO and below 300 °C for Bi$_2$Se$_3$, we do not expect any significant vacancies doped in LCO during the growth of Bi$_2$Se$_3$ to make it conducting. Therefore, we can ascribe the observed $R_{\text{AHE}}$ as the consequence of exchange coupling between the Bi$_2$Se$_3$ layer and the ferromagnetic LCO layer via the proximity effect.

The induced magnetism in Bi$_2$Se$_3$ films are also supported by the magnetization data. Fig. 4(c) plots the magnetic hysteresis loops of a typical 20 QL Bi$_2$Se$_3$ film on LCO/STO measured at 1.7 K. Its shape also resembles the M–H loops of LCO/STO that are shown in Fig. 4(b), with a similar coercive field seen in the inset. The magnitude of the coercive field in (c) is a little larger than that in (b), where $H_c = 350$ Oe. The small variation of the coercive field can be attributed to the interface exchange coupling that often results in a shifted hysteresis loop. $^{50}$

Moreover, the total magnetic moments at the saturation of the Bi$_2$Se$_3$/LCO sample is about 48% larger than that of the LCO film itself, indicating that there are additional induced moments in the heterostructure through the proximity effect at the interface. Because of the short-range nature of the ferromagnetic exchange interaction, we can expect the induced magnetic moments in the Bi$_2$Se$_3$ film to be canting arrayed in the bottom surface layer near the interface. $^{34,68}$ The measurements of the AHE and magnetization clearly demonstrate the existence of the induced moments in the Bi$_2$Se$_3$ layer with an out-of-plane component, by the strong coupling with the FMI layer through the proximity effect.

Further evidence of induced magnetism can be found from the temperature dependence of the field-cooled magnetization curve for Bi$_2$Se$_3$/LCO samples shown in Fig. 4(d). The plot is drawn on a log–log scale in order to magnify the transitions at a lower temperature. Below the Curie temperature of LCO/STO at $T_c \sim 85$ K, the magnetization $M$ increases rapidly as temperature $T$ decreases, as expected due to the ferromagnetic phase-transition. This is consistent with the $M$–$T$ curve of the LCO/STO film itself. While at temperatures higher than 85 K, the $M(T)$ of Bi$_2$Se$_3$/LCO exhibits much more interesting behaviors than that of LCO/STO, which is merely a near-zero straight line $^{55}$ (see also Fig. S4 in the ESI$^\dagger$). In the range of 85 K to 200 K, $M$ decreases but remains at a value higher than that of LCO, indicating that there are still some magnetic moments induced in the Bi$_2$Se$_3$/LCO heterostructure above 85 K. Moreover, as shown in Fig. S5 in the ESI,$^\dagger$ the $M$–$H$ hysteretic loop is also observed at 100 K for a thicker Bi$_2$Se$_3$/LCO sample. This is a striking result, which suggests a drastically enhanced Curie temperature of the Bi$_2$Se$_3$/LCO system. It can be inferred that, owing to the large spin–orbit interaction and the spin-momentum locking of the TI surface, the proximity-induced magnetic moments in the Bi$_2$Se$_3$ layer could be preserved up to a higher temperature than the Curie temperature of the LCO layer, due to the robustness of the TI nature. Moreover, it is possible for an exchange-coupling-motivated spin-state ordering and an induced strain through the interfacial interaction with the Bi$_2$Se$_3$ film to modify the ferromagnetism in the underlying LCO layer, which might give rise to a magnetic transition temperature higher than 100 K and result in an enhanced Curie temperature of the LCO film. $^{71,72}$

Combining the contribution from the two possible mechanisms, it is understandable why the AHE signals can survive above 100 K as mentioned before. Another surprising consequence is that, in the range of approximately 200 K to 300 K, a noticeable raise of $M(T)$ that is much larger than that expected from the paramagnetism alone emerges, and this could be attributed to the reoriented spins perpendicular to the interface in the absence of the large in-plane moments. $^{34}$ The extraordinary enhancement of ferromagnetism in relatively higher temperatures demonstrates the effectiveness of the proximity-coupling approach, which may also be applied to other similar systems with a broad variety of materials.

4. Conclusions

In conclusion, we have successfully synthesized a TI/FMI heterostructure by growing Bi$_2$Se$_3$ thin films on LCO/STO, which is shown to be a good platform for realizing a ferromagnetic topological insulating phase through proximity coupling. A well-ordered surface and smooth interface ensure a high-quality of the Bi$_2$Se$_3$ film with a mobility comparable to its pristine counterpart on the STO substrate. The suppressed WAL effect of Bi$_2$Se$_3$/LCO compared with that of Bi$_2$Se$_3$/STO, as well as the emergent AHE signals in transport measurements are both good indicators of the induced magnetism in TIs via exchange coupling with the ferromagnetic LCO layer. The observation of an unusual ferromagnetic phase persisting far above the Curie temperature of LCO/STO provides additional evidence of induced magnetism in the Bi$_2$Se$_3$ film. These findings will be both enlightening in further research studies and prospective in technological applications of integrated devices.

Conflicts of interest

There are no conflicts to declare.

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