UC Riverside

UC Riverside Previously Published Works

Title

Direct observation of proximity-induced magnetism and spin reorientation in topological insulator on a ferrimagnetic oxide

Permalink

<https://escholarship.org/uc/item/7ht878zd>

Journal

Applied Physics Letters, 114(8)

ISSN

0003-6951

Authors

Yang, CY
Lee, YH
Ou Yang, KH
et al.

Publication Date

2019-02-25

DOI

10.1063/1.5083931

Peer reviewed

Observation of Interfacial Antiferromagnetic Coupling between Magnetic Topological Insulator and Antiferromagnetic Insulator

Fei Wang,^{†,‡} Di Xiao,[†] Wei Yuan,^{§,||} Jue Jiang,[†] Yi-Fan Zhao,[†] Ling Zhang,[†] Yunyan Yao,[§] Wei Liu,[‡] Zhidong Zhang,[‡] Chaoxing Liu,[†] Jing Shi,^{||} Wei Han,^{§,⊥} Moses H. W. Chan,[†] Nitin Samarth,[†] and Cui-Zu Chang^{*,†,||}

[†]Department of Physics, Pennsylvania State University, University Park, Pennsylvania 16802, United States

[‡]Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, China

[§]International Center for Quantum Materials, School of Physics, Peking University, Beijing 100871, China

^{||}Department of Physics, University of California, Riverside, California 92521, United States

[⊥]Collaborative Innovation Center of Quantum Matter, Beijing 100871, China

Supporting Information

ABSTRACT: Inducing magnetic orders in a topological insulator (TI) to break its time reversal symmetry has been predicted to reveal many exotic topological quantum phenomena. The manipulation of magnetic orders in a TI layer can play a key role in harnessing these quantum phenomena toward technological applications. Here we fabricated a thin magnetic TI film on an antiferromagnetic (AFM) insulator Cr_2O_3 layer and found that the magnetic moments of the magnetic TI layer and the surface spins of the Cr_2O_3 layers favor interfacial AFM coupling. Field cooling studies show a crossover from negative to positive exchange bias clarifying the competition between the interfacial AFM coupling energy and the Zeeman energy in the AFM insulator layer. The interfacial exchange coupling also enhances the Curie temperature of the magnetic TI layer. The unique interfacial AFM alignment in magnetic TI on AFM insulator heterostructures opens a new route toward manipulating the interplay between topological states and magnetic orders in spin-engineered heterostructures, facilitating the exploration of proof-of-concept TI-based spintronic and electronic devices with multifunctionality and low power consumption.

KEYWORDS: Topological insulators, antiferromagnetic insulators, exchange coupling effect, antiferromagnetic coupling, exchange bias effect

Topological insulator (TI), a material in which the interior is insulating but the electrons can travel along its surface/edge conducting channels, has radically changed the research landscape of condensed matter physics and material science in the past decade.^{1,2} The nontrivial Dirac surface/edge states of a TI are induced by the strong spin–orbit coupling of the material and protected by time-reversal symmetry (TRS).^{3–6} Breaking the TRS of a TI with a magnetic perturbation can lead to a variety of exotic quantum phenomena such as the quantum anomalous Hall (QAH) effect,^{7–9} topological magnetoelectric effect,^{8,10} and image magnetic monopole.¹¹ The QAH effect has been experimentally demonstrated in magnetically doped TI thin films, specifically Cr- and/or V- doped $(\text{Bi,Sb})_2\text{Te}_3$.^{12,13} To date, the critical temperature of the QAH state (T_{QAH}) which we define as the temperature below which the Hall resistance is larger than $0.97 \hbar/e^2$ at zero magnetic field, in magnetically doped TI films is still ~ 2 K.^{12–15} A low T_{QAH} impedes both the exploration of fundamental physics and meaningful techno-

logical applications based on this exotic phenomenon. A direct route to increase T_{QAH} is to increase the magnetic doping level in the TI film to enhance the Curie temperature (T_{C}). However, this process invariably degrades the quality of TI films and can even make their behavior trivial.¹⁶

Ferromagnetic (FM) order can also be introduced into a TI layer through proximity to an FM insulator layer. By not introducing magnetic ions into the TI, the sample quality, in particular the carrier mobility, is expected to be much higher.¹⁷ Experimental efforts along this line have demonstrated proximity induced interfacial magnetization in TIs with a few FM insulators, including EuS ,^{18,19} GdN ,²⁰ $\text{BaFe}_{12}\text{O}_{19}$,²¹ $\text{Cr}_2\text{Ge}_2\text{Te}_6$,²² ferrimagnet yttrium/thulium iron garnet (YIG/

Received: January 3, 2019

Revised: March 20, 2019

Published: April 3, 2019

Figure 1. The magnetic TI Cr-doped Sb_2Te_3 /AFM Cr_2O_3 heterostructure. (a) Schematic atomic structure of the Cr-doped Sb_2Te_3 /AFM Cr_2O_3 heterostructure. The magnetic moments of Cr-doped Sb_2Te_3 and the surface spins of the Cr_2O_3 layer are AFM aligned. (b) STEM image of the Te layer-capped 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ /35 UC Cr_2O_3 heterostructure grown on a sapphire substrate, accompanied by an EDS map of Al, Cr, Sb, and Te of the sample.

TIG),^{23–26} and (Ga,Mn)As.²⁷ Because the magnetic proximity effect is a short-range magnetic exchange interaction, an antiferromagnetic (AFM) insulator layer with uncompensated surface spins could play the same role as a FM insulator.²⁸ Antiferromagnetic (AFM) insulators have a number of advantages compared with FM insulators, such as their insensitivity to perturbing magnetic fields, the high THz operating frequencies, and the negligible stray fields. These are attractive properties for spintronic applications.^{29–31} As the Néel temperature (T_N) of AFM insulator is usually well above the room temperature, it may be possible to induce a FM order with much higher Curie temperature (T_C) in a TI. Recently, a transport cum neutron scattering experiment has found the interfacial spin texture modulation and an enhanced T_C in a magnetically doped TI interfaced with an AFM metal CrSb.³² In view of the metallic property of CrSb, it is not possible to single out the transport property of the TI layer in such heterostructures. Therefore, an insulating AFM substrate (i.e., AFM insulator) would be a better candidate to induce magnetic orders in the TI layer.

In this Letter, we grew AFM insulator Cr_2O_3 layers with different thicknesses on heat-treated sapphire (0001) substrate to be followed with four quintuple layers (QL) thick magnetic TI $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ layer to form $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ /AFM Cr_2O_3 heterostructures. We used the anomalous Hall (AH) effect of the magnetic TI to probe the field- and temperature-dependence of its magnetization and the interfacial exchange coupling with the insulating AFM layer. We note that in these initial experiments the magnetic TI film (i.e., Cr-doped Sb_2Te_3) is not in the QAH insulator regime. Its chemical potential crosses the bulk valence bands and thus it is highly p-doped. We demonstrated that when the thickness of the Cr_2O_3 layer (m) is ≤ 3 -unit cell (UC, 1 UC ~ 1.36 nm), the AH hysteresis loops of the magnetic TI layer show a crossover from a negative exchange bias to a positive exchange bias along the axis of the magnetic field ($\mu_0 H_{\text{CF}}$) used for field cooling of the heterostructures. This crossover is revealed by systematically varying the magnitude of $\mu_0 H_{\text{CF}}$ and indicates an interfacial AFM coupling between the magnetic moments of the magnetic TI layers and the surface spins of the AFM Cr_2O_3 layer. The crossover of negative to positive exchange bias disappears for $m \geq 4$ UC because the T_N of the thicker Cr_2O_3 layer is higher than the T_C of the magnetic TI layer and this makes the surface spins of the AFM layer aligned randomly and smears out the

appearance of the exchange bias. Upon further increase of the thickness of the Cr_2O_3 layer, the T_C of $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ layers is progressively enhanced from ~ 39 K without the Cr_2O_3 layer to ~ 50 K for Cr_2O_3 layer thicker than 14 UC (see [Supporting Information](#)). The T_C enhancement also confirms the existence of an interfacial exchange coupling between the magnetic TI and the Cr_2O_3 layers.

Bulk Cr_2O_3 is a well-known AFM insulator with a T_N of 307 K,³³ whose linear magnetoelectric property has been used in voltage-controlled spintronic devices.^{34–36} In Cr_2O_3 , the spins are FM aligned along the (0001) direction within a single layer, whereas the spins of the adjacent layers are AFM coupled ([Figure 1a](#)). It is known that the ordering temperature T_C (T_N) of an FM (an AFM) material usually decreases in the 2D limit from the bulk value due to the finite size effect.^{37,38} Therefore, the T_N of AFM Cr_2O_3 films can be controlled by varying m . The Cr_2O_3 layers with different m (from 1 to 35 UC) were deposited at 500 °C by pulsed laser deposition (PLD) on heat-treated sapphire (0001) substrates.^{39,40} The growth process was monitored by in situ reflection high-energy electron diffraction (RHEED). The sharp and streaky “1 $\times$ 1” patterns indicate highly ordered Cr_2O_3 films with atomically flat surfaces ([Figure S1](#)). The high quality of the Cr_2O_3 films is also confirmed by atomic force microscopy and high-resolution X-ray diffraction (HR-XRD) measurements ([Figures S2 and S3](#)).

The growth of the 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ films on AFM Cr_2O_3 layers was carried out in a molecular beam epitaxy (MBE) chamber with a base pressure of 2×10^{-10} mbar. During the growth of the magnetic TI film, the Cr_2O_3 /sapphire substrate was maintained at ~ 240 °C. This low MBE growth temperature inhibits the diffusion of the Cr atoms into the Cr-doped Sb_2Te_3 layer. The 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ shows sharp “1 $\times$ 1” RHEED patterns, smooth surface morphology with a root-mean-square (RMS) roughness of ~ 0.9 nm over $5 \mu\text{m} \times 5 \mu\text{m}$ area in atomic force microscopy, and the sharp (00 n) peaks in the HR-XRD spectroscopy ([Figures S4–S6](#)). To avoid possible contamination (e.g., the degradation of the water and the carrier doping of oxygen), a 10 nm thick Te layer was deposited at room temperature on the magnetic TI film prior to the removal of the heterostructure samples from the MBE chamber for ex situ measurements.

Unlike the conventional diluted magnetic semiconductors, in which the FM order is usually mediated by the Ruderman–

Figure 2. AFM interfacial coupling between Cr-doped Sb_2Te_3 and Cr_2O_3 layers as revealed by the observation of crossover from negative to positive exchange bias. (a–c) The Hall resistance ρ_{yx} at $T = 2$ K of 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ layer grown on 3 UC (a), 2 UC (b), 1 UC (c) Cr_2O_3 layers field cooled with $\mu_0 H_{\text{CF}} = 0.05, 0.3, 0.8, 2$, and 7 T. (d) The exchange bias $\mu_0 H_{\text{E}}$ as a function of $\mu_0 H_{\text{CF}}$ at $T = 2$ K for 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ grown on 1–4 UC Cr_2O_3 layers. Note that $\mu_0 H_{\text{E}}$ is negligible in the 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ /4 UC Cr_2O_3 heterostructure since the T_{N} of 4 UC Cr_2O_3 layer is higher than T_{C} of 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ layer. The error bars reflect the standard deviations of the left and right coercive fields $\mu_0 H_{\text{CL}}$ and $\mu_0 H_{\text{CR}}$ used in the $\mu_0 H_{\text{E}}$ calculations. The inset shows the zoomed-in low $\mu_0 H_{\text{CF}}$ region. The horizontal intercepts denoted by the arrows become larger in heterostructures with the thicker Cr_2O_3 layer.

Kittel–Kasuya–Yosida (RKKY) mechanism (i.e., depends on the itinerant carriers),⁴¹ the FM order in magnetic TI film is mediated by the van Vleck mechanism.⁹ The van Vleck mediated FM order is independent of carrier density of the magnetic TI sample, so the FM order can still exist when the magnetic TI sample is insulating.⁴² The realization of the QAH effect is just a result of this unique property in magnetic TI.^{12,13}

Figure 1b shows the cross-sectional scanning transmission electron microscopy (STEM) image of the 10 nm Te capped 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ /35 UC Cr_2O_3 heterostructure grown on a sapphire substrate and the corresponding energy dispersive spectroscopy (EDS) mappings of Al, Cr, Sb, and Te. Al and Cr reside in the sapphire substrate and Cr_2O_3 layers, respectively, while Sb and Te are found in the Cr-doped TI and Te capping layers. The absence of the Cr signal in Cr-doped Sb_2Te_3 is due to the low concentration of Cr in the TI layer. The trace Sb signal in the Te capping layer is a result of their neighboring peak positions in the X-ray spectrum. Additional TEM and EDS results on the other two heterostructures (7 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ /3 UC Cr_2O_3 and 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ /14 UC Cr_2O_3) are shown in Supporting Information (Figure S7).

The 10 nm Te-capped 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ / m UC Cr_2O_3 heterostructure samples were scratched into a Hall bar geometry using a computer-controlled probe station.¹⁰ We then carried out transport studies in a physical property measurement system (Quantum Design, 2 K, 9 T) with an external magnetic field perpendicular to the film. The excitation current is $1 \mu\text{A}$. All samples were field cooled at multiple different values of the external magnetic field $\mu_0 H_{\text{CF}}$. Cooling began from 320 K, which is above T_{N} of bulk Cr_2O_3 (~ 307 K),³³ and continued down to the target measurement temperatures. The field cooling process eliminates any possible spontaneous and random alignments of the AFM order in the Cr_2O_3 layer. Figure 2a shows the magnetic field ($\mu_0 H$) dependence of the Hall resistance (ρ_{yx}) at 2 K of the 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ /3 UC Cr_2O_3 heterostructure field cooled at $\mu_0 H_{\text{CF}} = 0.05, 0.3, 0.8, 2$, and 7 T. The blue (red) curves

correspond to ρ_{yx} measured while sweeping $\mu_0 H$ downward from 0.2 T to -0.2 T (upward from -0.2 to 0.2 T). The small $\mu_0 H$ sweep range is chosen to make sure we preserve the spin ordering of the AFM layers induced by the field cooling procedure. The nearly square ρ_{yx} hysteresis loops confirm a well-defined FM order with perpendicular magnetic anisotropy in Cr-doped Sb_2Te_3 films.^{12,42} The ρ_{yx} loop for the sample prepared at $\mu_0 H_{\text{CF}} = 0.05$ T shows a negative exchange bias, that is, the left coercive field $|\mu_0 H_{\text{CL}}|$ is larger than the right coercive field $\mu_0 H_{\text{CR}}$ thus indicating the presence of a unidirectional magnetic exchange anisotropy across the $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ / Cr_2O_3 interface. The $\mu_0 H_{\text{CF}} = 0.3$ T loop still shows the negative exchange bias, but the magnitude of the shift is reduced. The $\mu_0 H_{\text{CF}} = 0.8$ T loop is nearly symmetric with $|\mu_0 H_{\text{CL}}| = \mu_0 H_{\text{CR}}$. Upon further increase of $\mu_0 H_{\text{CF}}$ to 2 T, the negative exchange bias (i.e., $|\mu_0 H_{\text{CL}}| > \mu_0 H_{\text{CR}}$) is replaced by a positive exchange bias (i.e., $|\mu_0 H_{\text{CL}}| < \mu_0 H_{\text{CR}}$). The positive exchange bias becomes more pronounced if the sample is cooled under $\mu_0 H_{\text{CF}} = 7$ T. The observation of a crossover from negative to positive exchange bias indicates that opposite AFM domain states in the Cr_2O_3 layer were established under different $\mu_0 H_{\text{CF}}$.

Figure 2b,c shows the Hall traces of the 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ /2 UC Cr_2O_3 and 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ /1 UC Cr_2O_3 , respectively. These two samples also display a negative exchange bias under low $\mu_0 H_{\text{CF}}$ and positive exchange bias under high $\mu_0 H_{\text{CF}}$. The magnitude of the exchange bias field ($\mu_0 H_{\text{E}}$) is defined as $(\mu_0 H_{\text{CR}} - |\mu_0 H_{\text{CL}}|)/2$. In Figure 2d, we summarize the variation of the $\mu_0 H_{\text{E}}$ s as a function of $\mu_0 H_{\text{CF}}$ at $T = 2$ K for the 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ / m UC Cr_2O_3 heterostructures ($m = 1-4$). For $m \leq 3$ UC, the $\mu_0 H_{\text{E}}$ changes from a negative value through zero to a positive value when the $\mu_0 H_{\text{CF}}$ is increased from 0.05 to 7 T. The critical $\mu_0 H_{\text{CF}}$ with $\mu_0 H_{\text{E}} = 0$ is labeled as $\mu_0 H_{\text{CF}}^0$. The $\mu_0 H_{\text{CF}}^0$ are $0.8, 0.7$, and 0.3 T for 3 UC, 2 UC, and 1 UC Cr_2O_3 layers, respectively (inset of Figure 2d). However, for $m = 4$ UC, the ρ_{yx} loop is always symmetric, showing a negligible shift under

Figure 3. Temperature dependence of the positive exchange bias in Cr-doped $\text{Sb}_2\text{Te}_3/\text{Cr}_2\text{O}_3$ heterostructures field cooled at $\mu_0 H_{CF} = 7 \text{ T}$. (a–c) The Hall resistance ρ_{yx} at varying temperatures of 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ layer on 3 UC (a), 2 UC (b), 1 UC (c) Cr_2O_3 layers. (d) Temperature dependence of the exchange bias $\mu_0 H_E$ in 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ grown on 1–4 UC Cr_2O_3 heterostructures. The blocking temperature T_B at which $\mu_0 H_E = 0$ increases with increasing thickness of the Cr_2O_3 layer. The magnitude of the error bars reflects the standard deviations of the $\mu_0 H_{cL}$ and $\mu_0 H_{cR}$ used in calculating $\mu_0 H_E$. Inset shows T_B as a function of the thickness of the Cr_2O_3 layer in the heterostructures.

Figure 4. A schematic of the model of interfacial AFM coupling between Cr-doped Sb_2Te_3 and Cr_2O_3 layers. (a,b) The orientations of the magnetic moments in the Cr-doped Sb_2Te_3 and Cr_2O_3 layers field cooled under low (a) and high (b) $\mu_0 H_{CF}$. When the heterostructure is field cooled under a low $\mu_0 H_{CF}$ ($\mu_0 H_{CF} M_{AFM} < J_E S_{AFM} S_{FM}$), the spins of the Cr-doped TI and Cr_2O_3 layers are AFM aligned. For a high $\mu_0 H_{CF}$ ($\mu_0 H_{CF} M_{AFM} > J_E S_{AFM} S_{FM}$), the spins of the Cr-doped TI and Cr_2O_3 layers are forced to be FM aligned. (c) Negative exchange bias and the spin switching process of the Cr-doped TI layer prepared with a low $\mu_0 H_{CF}$. (d) Positive exchange bias and the spin switching process of the Cr-doped TI layer prepared with a high $\mu_0 H_{CF}$.

all $\mu_0 H_{CF}$. This indicates the disappearance of the exchange bias effect (Figure 2d and Figure S9).

Exchange bias, revealed by a shift in the magnetic hysteresis loop of a FM layer exchange coupled to an AFM layer, is a well-known phenomenon in systems with FM–AFM interfaces.⁴³

The AFM layer usually plays the role of a pinning layer, whose alignment direction determines the shift direction of the hysteresis loop. Most FM–AFM systems have the interfacial FM coupling, so the shift of the hysteresis loop is always along the direction opposite to $\mu_0 H_{CF}$ showing a negative exchange

bias, regardless of the magnitude of $\mu_0 H_{CF}$.⁴³ Our observations of a crossover between negative and positive exchange bias in heterostructures of Cr-doped Sb_2Te_3 and Cr_2O_3 heterostructures are similar to the behavior first observed in FeF_2 –Fe bilayers. In that system, the positive exchange bias under high $\mu_0 H_{CF}$ provided proof of an AFM coupling between the magnetic moments of a FM layer and the surface spins of an AFM layer.^{43–46} However, as we demonstrate later in this Letter, the interfacial exchange coupling in our system also enhances the FM ordering temperature (i.e., T_C) in the magnetic TI.

Next, we focus on the study of the evolution of positive $\mu_0 H_E$ of 4QL $Sb_{1.8}Cr_{0.2}Te_3$ on Cr_2O_3 of 3, 2 and 1 UC field-cooled with $\mu_0 H_{CF} = 7$ T. The ρ_{yx} hysteresis loops of these heterostructures at temperatures from 2 to 40 K are shown respectively in Figure 3a–c. These loops show that $\mu_0 H_{CR}$, $|\mu_0 H_{CL}|$, and the positive $\mu_0 H_E$ all monotonically decrease with increasing temperature. Figure 3d shows $\mu_0 H_E$ as a function of temperature for these three heterostructures. The blocking temperature T_B , that is, the temperature above which $\mu_0 H_E$ vanishes, increases with increasing m of the Cr_2O_3 layers. The T_B s for the heterostructures with 1 UC, 2 UC, and 3 UC Cr_2O_3 layers are 15, 25, and 35 K, respectively. The higher T_B in the heterostructures with thicker Cr_2O_3 layers suggests the T_N of thicker Cr_2O_3 layers is higher.

When the T_N of the Cr_2O_3 layer is higher than the T_C of the Cr-doped Sb_2Te_3 layer during the cooling process under an external magnetic field (H_{CF}), the temperature first arrives at $T = T_N$, and the AFM order in Cr_2O_3 layer is formed while the magnetic domains in Cr-doped Sb_2Te_3 layer are still random. The random magnetic domains in Cr-doped Sb_2Te_3 layer make the surface spins of the Cr_2O_3 layer also randomly aligned. With further cooling to $T = T_C$, the FM order is formed in Cr-doped Sb_2Te_3 layer but the coupling energy between the Cr_2O_3 and Cr-doped Sb_2Te_3 layers is not enough to realign the surface spins of the Cr_2O_3 layer when $T < T_N$. Therefore, to achieve the exchange bias effect in the systems with the FM–AFM interface, the T_N of the AFM layer must be lower than the T_C of the FM layer.

We now explain why the observation of the crossover from the negative to positive exchange bias demonstrates that the magnetic moments of the Cr-doped Sb_2Te_3 and Cr_2O_3 layers are AFM coupled. On the basis of the above analysis, we know for $m \leq 3$ UC that the T_N of Cr_2O_3 layer must be lower than the $T_C \sim 38$ K of 4 QL $Sb_{1.8}Cr_{0.2}Te_3$ (Figure S14). Since $T_C > T_N$ during the cooling of the samples under a positive $\mu_0 H_{CF}$, the temperature first arrives at $T = T_C$, and the magnetic moments of the 4 QL $Sb_{1.8}Cr_{0.2}Te_3$ layer are consequently aligned upward as shown in Figure 4a,b. With further cooling to $T = T_N$, the AFM order is formed in Cr_2O_3 layers. If the AFM and FM layers favor the interfacial AFM coupling, the spins of the aligned $Sb_{1.8}Cr_{0.2}Te_3$ layer exert an effective field to force the surface spins of the Cr_2O_3 layer to be oppositely aligned, that is, pointing downward. The AFM coupling energy between the AFM and FM layers is $J_E S_{AFM} S_{FM}$, where J_E is the exchange coupling strength between the surface spins of Cr_2O_3 (S_{AFM}) and $Sb_{1.8}Cr_{0.2}Te_3$ (S_{MTI}) layers. In addition, the Zeeman energy in the AFM layer under the external magnetic field $\mu_0 H_{CF}$ is $\mu_0 H_{CF} M_{AFM}$, where M_{AFM} is the surface magnetization of the AFM Cr_2O_3 layer. This Zeeman energy forces the surface spins of the Cr_2O_3 layer to be aligned along the direction of the external magnetic field.^{44,46} The alignment between AFM and FM layers is determined by the competition between the AFM

coupling energy $J_E S_{AFM} S_{FM}$ and the Zeeman energy in the AFM layer $\mu_0 H_{CF} M_{AFM}$.

When the sample is field-cooled under a low $\mu_0 H_{CF}$, $\mu_0 H_{CF} M_{AFM} < J_E S_{AFM} S_{FM}$ (Figure 4a), the AFM coupling energy $J_E S_{AFM} S_{FM}$ is dominant and, as a consequence, the positive magnetization of the Cr-doped Sb_2Te_3 will force the top surface spins of the Cr_2O_3 layer to point downward. With a high $\mu_0 H_{CF}$, $\mu_0 H_{CF} M_{AFM} > J_E S_{AFM} S_{FM}$ (Figure 4b), the Zeeman energy in Cr_2O_3 layer $\mu_0 H_{CF} M_{AFM}$ is dominant and makes the top surface spins of the Cr_2O_3 layer to point upward. To compensate for the AFM coupling, the orientation of the surface spins in the Cr_2O_3 layer will be slightly tilted. Previous studies have demonstrated the spin-flop transition in Cr_2O_3 only occurs at the higher magnetic field (a few Teslas).^{40,47} Therefore, when the magnetic field sweep range is ± 0.2 T, the adjacent magnetic layers in the Cr_2O_3 layer remain AFM ordered (Figure 4b). With a critical value of $\mu_0 H_{CF}$ (i.e., $\mu_0 H_{CF}^0$), $\mu_0 H_{CF}^0 M_{AFM} = J_E S_{AFM} S_{FM}$, the Cr_2O_3 layer is in a randomly distributed multidomain state, and thus $\mu_0 H_E = 0$ and gives rise to the symmetric hysteresis loop. $\mu_0 H_0$ CF increases with m as a result of the J_E enhancement, which is induced by the higher T_N in thicker Cr_2O_3 layers.⁴⁶

The surface spin configuration of the Cr_2O_3 layer at low temperature is locked by its AFM magnetic structure during the field cooling process. As noted above, when the external $\mu_0 H$ is swept in the ± 0.2 T range the FM spins of the 4 QL $Sb_{1.8}Cr_{0.2}Te_3$ layer with a $\mu_0 H_c$ of ~ 0.1 T can be switched by the external magnetic field while the AFM spins of the Cr_2O_3 layer are unchanged.^{40,47} The existence of the AFM coupling energy $J_E S_{AFM} S_{FM}$ across Cr-doped Sb_2Te_3 and Cr_2O_3 interface will cause the shift of the FM hysteresis loop of the 4 QL $Sb_{1.8}Cr_{0.2}Te_3$ layer. For samples field-cooled with a low $\mu_0 H_{CF}$, the top surface spins of the Cr_2O_3 layer are locked downward when $\mu_0 H$ is swept at $T = 2$ K and the magnetic moments of the 4 QL $Sb_{1.8}Cr_{0.2}Te_3$ layer are pointing upward for $\mu_0 H = 0.2$ T (Step I in Figure 4c). When $\mu_0 H$ is swept downward near the left coercive field $\mu_0 H_{CL}$, the upward magnetic domains of the 4 QL $Sb_{1.8}Cr_{0.2}Te_3$ layer is reversed. Because of the interfacial AFM coupling, the spins of $Sb_{1.8}Cr_{0.2}Te_3$ are energetically more favorable to point upward to retain the antiparallel alignment. Therefore, to overcome the interfacial AFM coupling energy, a magnetic field $\mu_0 H$ larger than $\mu_0 H_c$ is needed to reverse the magnetic domains from upward to downward, that is, $|\mu_0 H_{CL}| > \mu_0 H_c$ (Step II in Figure 4c). When $\mu_0 H$ is swept back from -0.2 T, the presence of the interfacial AFM coupling helps the magnetization reversal of 4 QL $Sb_{1.8}Cr_{0.2}Te_3$ layer from parallel to antiparallel alignments, so the right coercive field $\mu_0 H_{CR} < \mu_0 H_c$ (Step IV in Figure 4c). This explains why a negative exchange bias, i.e., $|\mu_0 H_{CL}| > \mu_0 H_{CR}$, is observed under a low $\mu_0 H_{CF}$.

When the sample is field-cooled under a high $\mu_0 H_{CF}$, the magnetic moments at $\mu_0 H = 0.2$ T of the Cr-doped Sb_2Te_3 are still pointing upward but the top surface spins of the Cr_2O_3 layer are also pointing upward but slightly tilted (Step I in Figure 4d). When $\mu_0 H$ is swept downward, the existence of the AFM coupling will favor the magnetization reversal of 4 QL $Sb_{1.8}Cr_{0.2}Te_3$ layer from upward to downward, so $|\mu_0 H_{CL}| < \mu_0 H_c$ (Step II in Figure 4d). When $\mu_0 H$ is swept backward from -0.2 T, the interfacial AFM coupling will impede the magnetization reversal of 4 QL $Sb_{1.8}Cr_{0.2}Te_3$ layer from downward to upward, so $\mu_0 H_{CR} > \mu_0 H_c$. This corresponds to the positive exchange bias, that is, $|\mu_0 H_{CL}| < \mu_0 H_{CR}$, observed for the samples field cooled with a high $\mu_0 H_{CF}$.

We have also systematically studied the T_C of the 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ layer for $m \geq 4$. The T_C of 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ layer in all heterostructures was determined using the Arrott-plots (Figure S11).⁴⁸ The m dependence of T_C is summarized in Figure S12. For $m \leq 4$ UC, T_C is ~ 39 K, consistent with the T_C of 4 QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ film grown on the nonmagnetic $\text{SrTiO}_3(111)$ substrate (Figure S13). For $m \geq 5$ UC, the T_C starts to increase and saturates near 50 K for $m \geq 14$ UC. The T_C enhancement of 4QL $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ grown on the thicker Cr_2O_3 films further demonstrate the existence of the interfacial exchange coupling.

To summarize, we demonstrated the tuning of the exchange bias in a given magnetic TI on AFM insulator heterostructure from negative to positive values by field cooling with different $\mu_0 H_{\text{CF}}$. This is made possible by the interfacial AFM coupling between the FM aligned spins of the magnetic TI layer and the surface spins of the AFM insulator. This efficient tuning of the exchange bias provides a new route to effectively manipulate the magnetic spins of the TI layer. Our findings, when combined with the linear magnetoelectric effect of Cr_2O_3 ,^{34–36} could facilitate the development of proof-of-concept electric field-controlled TI-based energy-efficient spintronic devices.

EXPERIMENTAL METHODS

PLD Growth of the Cr_2O_3 Films. The Cr_2O_3 films were grown on sapphire (0001) substrates in a pulsed laser deposition (PLD) chamber with a base pressure $\sim 2 \times 10^{-8}$ mbar. Before the growth of Cr_2O_3 layers, the sapphire substrates were first cleaned in a liquid mixture of $\text{NH}_4\text{OH}/\text{H}_2\text{O}_2/\text{H}_2\text{O} \sim 1:1:50$ and rinsed with isopropyl alcohol and deionized water. The sapphire substrates were then annealed in a tube furnace at 1000 °C for 2 h with flowing oxygen. The surface of the heat-treated sapphire substrates shows atomically flat terraces. The Cr_2O_3 film was deposited on the heat-treated sapphire in the PLD chamber at 500 °C and postannealed at 600 °C for 30 min under a pressure of ~ 0.03 mbar O_2 . The power and frequency of the laser are 8.0 ± 0.2 mJ and 8 Hz, respectively.

MBE Growth of Magnetic TI Films. The magnetic topological insulator (TI) film growth was carried out in a molecular beam epitaxy (MBE) system with a vacuum $\sim 2 \times 10^{-10}$ mbar. The Cr_2O_3 films on sapphire (0001) substrates were outgassed at ~ 500 °C for 1 h before the growth of the TI films. High-purity Sb (6N), Cr (5N) and Te (6N) were evaporated from Knudsen effusion cells. During growth of the magnetic TI film, the substrate was maintained at ~ 240 °C. The flux ratio of $\text{Te}/(\text{Sb} + \text{Cr})$ was set to be greater than ($>$) 10 to prevent Te deficiency in the samples. The growth rate of Cr-doped Sb_2Te_3 film was ~ 0.25 quintuple (QL)/min. Following the growth, the magnetic TI films were annealed at ~ 240 °C for 30 min to improve the crystal quality before being cooled down to room temperature. Finally, a 10 nm thick Te layer was deposited at room temperature on top of the magnetic TI films prior to the removal from the MBE chamber for ex situ transport and other characterization measurements.

Hall Bar Device Fabrications. The magnetic TI film on Cr_2O_3 heterostructure samples grown on 5 mm $\times$ 5 mm sapphire substrate was scratched into a Hall bar geometry using a computer-controlled probe station. The effective area of the Hall bar device is ~ 1 mm $\times$ 0.5 mm. The electrical Ohmic-contacts for transport measurements were made by pressing indium dots on the Hall bar.

Electrical Transport Measurements. Electrical transport measurements were conducted using a Physical Property

Measurement System (Quantum Design, 2 K, 9 T) with the magnetic field applied perpendicular to the film plane. The excitation current in the dc measurements was 1 μA . All the magneto-longitudinal and Hall resistances shown in the main text and Supporting Information, unless pointed out otherwise, were antisymmetrized as a function of the magnetic field. Each sample was cooled from 320 K, above the Néel temperature (T_N) of Cr_2O_3 (~ 307 K),³³ to the target measuring temperatures under a fixed cooling magnetic field $\mu_0 H_{\text{CF}}$ ranging from 0.05 to 7 T.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.9b00027.

Characterizations of Cr_2O_3 films on sapphire; characterizations of $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3/\text{Cr}_2\text{O}_3$ heterostructures; transport results of $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3/\text{Cr}_2\text{O}_3$ heterostructures; determining the Curie temperatures of $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3/\text{Cr}_2\text{O}_3$ heterostructures; transport results of the $\text{Sb}_{1.8}\text{Cr}_{0.2}\text{Te}_3$ film on $\text{SrTiO}_3(111)$; determining the Blocking temperatures of Cr_2O_3 films (PDF)

AUTHOR INFORMATION

Corresponding Author

*E-mail: cxc955@psu.edu (C.Z.C.).

ORCID

Wei Han: 0000-0002-1757-4479

Cui-Zu Chang: 0000-0003-3515-2955

Author Contributions

F.W., D.X. and W.Y. contributed equally. C.Z.C. conceived and designed the research. W.Y. and Y.Y. grew the antiferromagnetic insulator films on sapphire substrates using PLD with the help of J.S. and W.H. D.X. grew the magnetic TI films on antiferromagnetic insulator substrates with the help of N.S. and C.Z.C. F.W. performed characterizations of the samples with the help of W.L., Z.Z. and C.Z.C. F.W., J.J., Y.F.Z., and L.Z. carried out the PPMS measurements with the help of M.H., W.C., and C.Z.C. C.X.L. provided theoretical support. F.W. and C.Z.C. analyzed the data and wrote the manuscript with contributions from all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

The authors would like to thank B. H. Yan, B. J. Dong, and X. D. Xu for the helpful discussions. F.W., Y.Z., and C.Z.C. acknowledge the support from ARO Young Investigator Program Award (W911NF1810198) and the Alfred P. Sloan Research Fellowship. D.X. and N.S. acknowledge the support from the Penn State Two-Dimensional Crystal Consortium-Materials Innovation Platform (2DCC-MIP) under NSF Grant DMR-1539916 and the Office of Naval Research (N00014-15-1-2370). C.X.L. acknowledges the support from the Office of Naval Research (N00014-15-1-2675). F.W., W.L., and Z.D.Z. acknowledge the support from the State Key Program of Research and Development of China (2017YFA0206302), the National Natural Science Foundation of China (NSFC) (51590883, 51331006, and 51771198) and the Key Research

Program of Chinese Academy of Sciences (KJZD-EW-M05-3). J.S. acknowledges the support from DOE Grant (DE-FG02-07ER46351). W.H. acknowledges the support from the National Basic Research Programs of China (2015CB921104) and the National Natural Science Foundation of China (NSFC) (11574006). Support for the electrical transport measurements and data analysis is provided by the DOE Grant (DE-SC0019064).

REFERENCES

- (1) Hasan, M. Z.; Kane, C. L. Colloquium: Topological Insulators. *Rev. Mod. Phys.* **2010**, *82* (4), 3045–3067.
- (2) Qi, X. L.; Zhang, S. C. Topological Insulators and Superconductors. *Rev. Mod. Phys.* **2011**, *83* (4), 1057–1110.
- (3) Bernevig, B. A.; Hughes, T. L.; Zhang, S. C. Quantum Spin Hall Effect and Topological Phase Transition in HgTe Quantum Wells. *Science* **2006**, *314* (5806), 1757–1761.
- (4) König, M.; Wiedmann, S.; Brune, C.; Roth, A.; Buhmann, H.; Molenkamp, L. W.; Qi, X. L.; Zhang, S. C. Quantum Spin Hall Insulator State in HgTe Quantum Wells. *Science* **2007**, *318* (5851), 766–770.
- (5) Zhang, H. J.; Liu, C. X.; Qi, X. L.; Dai, X.; Fang, Z.; Zhang, S. C. Topological Insulators in Bi₂Se₃, Bi₂Te₃ and Sb₂Te₃ with a Single Dirac Cone on the Surface. *Nat. Phys.* **2009**, *5* (6), 438–442.
- (6) Hsieh, D.; Xia, Y.; Qian, D.; Wray, L.; Meier, F.; Dil, J. H.; Osterwalder, J.; Patthey, L.; Fedorov, A. V.; Lin, H.; Bansil, A.; Grauer, D.; Hor, Y. S.; Cava, R. J.; Hasan, M. Z. Observation of Time-Reversal-Protected Single-Dirac-Cone Topological-Insulator States in Bi₂Te₃ and Sb₂Te₃. *Phys. Rev. Lett.* **2009**, *103* (14), 146401.
- (7) Haldane, F. D. M. Model for a Quantum Hall-Effect without Landau Levels: Condensed-Matter Realization of the "Parity Anomaly". *Phys. Rev. Lett.* **1988**, *61* (18), 2015–2018.
- (8) Qi, X. L.; Hughes, T. L.; Zhang, S. C. Topological Field Theory of Time-Reversal Invariant Insulators. *Phys. Rev. B: Condens. Matter Mater. Phys.* **2008**, *78* (19), 195424.
- (9) Yu, R.; Zhang, W.; Zhang, H. J.; Zhang, S. C.; Dai, X.; Fang, Z. Quantized Anomalous Hall Effect in Magnetic Topological Insulators. *Science* **2010**, *329* (5987), 61–64.
- (10) Xiao, D.; Jiang, J.; Shin, J. H.; Wang, W. B.; Wang, F.; Zhao, Y. F.; Liu, C. X.; Wu, W. D.; Chan, M. H. W.; Samarth, N.; Chang, C. Z. Realization of the Axion Insulator State in Quantum Anomalous Hall Sandwich Heterostructures. *Phys. Rev. Lett.* **2018**, *120* (5), 056801.
- (11) Qi, X. L.; Li, R. D.; Zang, J. D.; Zhang, S. C. Inducing a Magnetic Monopole with Topological Surface States. *Science* **2009**, *323* (5918), 1184–1187.
- (12) Chang, C. Z.; Zhang, J. S.; Feng, X.; Shen, J.; Zhang, Z. C.; Guo, M. H.; Li, K.; Ou, Y. B.; Wei, P.; Wang, L. L.; Ji, Z. Q.; Feng, Y.; Ji, S. H.; Chen, X.; Jia, J. F.; Dai, X.; Fang, Z.; Zhang, S. C.; He, K.; Wang, Y. Y.; Lu, L.; Ma, X. C.; Xue, Q. K. Experimental Observation of the Quantum Anomalous Hall Effect in a Magnetic Topological Insulator. *Science* **2013**, *340* (6129), 167–170.
- (13) Chang, C. Z.; Zhao, W. W.; Kim, D. Y.; Zhang, H. J.; Assaf, B. A.; Heiman, D.; Zhang, S. C.; Liu, C. X.; Chan, M. H. W.; Moodera, J. S. High-Precision Realization of Robust Quantum Anomalous Hall State in a Hard Ferromagnetic Topological Insulator. *Nat. Mater.* **2015**, *14* (5), 473–477.
- (14) Mogi, M.; Yoshimi, R.; Tsukazaki, A.; Yasuda, K.; Kozuka, Y.; Takahashi, K. S.; Kawasaki, M.; Tokura, Y. Magnetic Modulation Doping in Topological Insulators toward Higher-Temperature Quantum Anomalous Hall Effect. *Appl. Phys. Lett.* **2015**, *107* (18), 182401.
- (15) Ou, Y.; Liu, C.; Jiang, G.; Feng, Y.; Zhao, D.; Wu, W.; Wang, X. X.; Li, W.; Song, C.; Wang, L. L.; Wang, W.; Wu, W.; Wang, Y.; He, K.; Ma, X. C.; Xue, Q. K. Enhancing the Quantum Anomalous Hall Effect by Magnetic Codoping in a Topological Insulator. *Adv. Mater.* **2018**, *30*, 1703062.
- (16) Zhang, J. S.; Chang, C. Z.; Tang, P. Z.; Zhang, Z. C.; Feng, X.; Li, K.; Wang, L. L.; Chen, X.; Liu, C. X.; Duan, W. H.; He, K.; Xue, Q. K.; Ma, X. C.; Wang, Y. Y. Topology-Driven Magnetic Quantum Phase Transition in Topological Insulators. *Science* **2013**, *339* (6127), 1582–1586.
- (17) Chang, C. Z.; Wei, P.; Moodera, J. S. Breaking Time Reversal Symmetry in Topological Insulators. *MRS Bull.* **2014**, *39* (10), 867–872.
- (18) Wei, P.; Katmis, F.; Assaf, B. A.; Steinberg, H.; Jarillo-Herrero, P.; Heiman, D.; Moodera, J. S. Exchange-Coupling-Induced Symmetry Breaking in Topological Insulators. *Phys. Rev. Lett.* **2013**, *110* (18), 186807.
- (19) Katmis, F.; Lauter, V.; Nogueira, F. S.; Assaf, B. A.; Jamer, M. E.; Wei, P.; Satpati, B.; Freeland, J. W.; Eremin, I.; Heiman, D.; Jarillo-Herrero, P.; Moodera, J. S. A High-Temperature Ferromagnetic Topological Insulating Phase by Proximity Coupling. *Nature* **2016**, *533* (7604), 513–516.
- (20) Kandala, A.; Richardella, A.; Rench, D. W.; Zhang, D. M.; Flanagan, T. C.; Samarth, N. Growth and Characterization of Hybrid Insulating Ferromagnet-Topological Insulator Heterostructure Devices. *Appl. Phys. Lett.* **2013**, *103* (20), 202409.
- (21) Yang, W. M.; Yang, S. O.; Zhang, Q. H.; Xu, Y.; Shen, S. P.; Liao, J.; Teng, J.; Nan, C. W.; Gu, L.; Sun, Y.; Wu, K. H.; Li, Y. Q. Proximity Effect between a Topological Insulator and a Magnetic Insulator with Large Perpendicular Anisotropy. *Appl. Phys. Lett.* **2014**, *105* (9), 092411.
- (22) Alegria, L. D.; Ji, H.; Yao, N.; Clarke, J. J.; Cava, R. J.; Petta, J. R. Large Anomalous Hall Effect in Ferromagnetic Insulator-Topological Insulator Heterostructures. *Appl. Phys. Lett.* **2014**, *105* (5), 053512.
- (23) Wang, H. L.; Kally, J.; Lee, J. S.; Liu, T.; Chang, H. C.; Hickey, D. R.; Mkhoyan, K. A.; Wu, M. Z.; Richardella, A.; Samarth, N. Surface-State-Dominated Spin-Charge Current Conversion in Topological-Insulator-Ferromagnetic-Insulator Heterostructures. *Phys. Rev. Lett.* **2016**, *117* (7), 076601.
- (24) Lang, M. R.; Montazeri, M.; Onbasli, M. C.; Kou, X. F.; Fan, Y. B.; Upadhyaya, P.; Yao, K. Y.; Liu, F.; Jiang, Y.; Jiang, W. J.; Wong, K. L.; Yu, G. Q.; Tang, J. S.; Nie, T. X.; He, L.; Schwartz, R. N.; Wang, Y.; Ross, C. A.; Wang, K. L. Proximity Induced High-Temperature Magnetic Order in Topological Insulator - Ferrimagnetic Insulator Heterostructure. *Nano Lett.* **2014**, *14* (6), 3459–3465.
- (25) Jiang, Z. L.; Chang, C. Z.; Tang, C.; Wei, P.; Moodera, J. S.; Shi, J. Independent Tuning of Electronic Properties and Induced Ferromagnetism in Topological Insulators with Heterostructure Approach. *Nano Lett.* **2015**, *15* (9), 5835–5840.
- (26) Tang, C.; Chang, C. Z.; Zhao, G.; Liu, Y.; Jiang, Z.; Liu, C.-x.; McCartney, M. R.; Smith, D. J.; Chen, T.; Moodera, J. S.; Shi, J. Above 400K robust Perpendicular Ferromagnetic Phase in a Topological Insulator. *Sci. Adv.* **2017**, *3*, No. e1700307.
- (27) Lee, J. S.; Richardella, A.; Fraleigh, R. D.; Liu, C. X.; Zhao, W. W.; Samarth, N. Engineering the Breaking of Time-Reversal Symmetry in Gate-Tunable Hybrid Ferromagnet/Topological Insulator Heterostructures. *npj Quantum Mater.* **2018**, *3*, 51.
- (28) Luo, W. D.; Qi, X. L. Massive Dirac Surface States in Topological Insulator/Magnetic Insulator Heterostructures. *Phys. Rev. B: Condens. Matter Mater. Phys.* **2013**, *87* (8), 085431.
- (29) Baltz, V.; Manchon, A.; Tsoi, M.; Moriyama, T.; Ono, T.; Tserkovnyak, Y. Antiferromagnetic Spintronics. *Rev. Mod. Phys.* **2018**, *90* (1), 015005.
- (30) Smejkal, L.; Mokrousov, Y.; Yan, B. H.; MacDonald, A. H. Topological Antiferromagnetic Spintronics. *Nat. Phys.* **2018**, *14* (3), 242–251.
- (31) Jungwirth, T.; Marti, X.; Wadley, P.; Wunderlich, J. Antiferromagnetic Spintronics. *Nat. Nanotechnol.* **2016**, *11* (3), 231–241.
- (32) He, Q. L.; Kou, X. F.; Grutter, A. J.; Yin, G.; Pan, L.; Che, X. Y.; Liu, Y. X.; Nie, T. X.; Zhang, B.; Disseler, S. M.; Kirby, B. J.; Ratcliff, W.; Shao, Q. M.; Murata, K.; Zhu, X. D.; Yu, G. Q.; Fan, Y. B.; Montazeri, M.; Han, X. D.; Borchers, J. A.; Wang, K. L. Tailoring Exchange Couplings in Magnetic Topological-Insulator/Antiferromagnet Heterostructures. *Nat. Mater.* **2017**, *16* (1), 94–100.
- (33) McGuire, T. R.; Scott, E. J.; Grannis, F. H. Antiferromagnetism in a Cr₂O₃ Crystal. *Phys. Rev.* **1956**, *102* (4), 1000–1003.

- (34) He, X.; Wang, Y.; Wu, N.; Caruso, A. N.; Vescovo, E.; Belashchenko, K. D.; Dowben, P. A.; Binek, C. Robust Isothermal Electric Control of Exchange Bias at Room Temperature. *Nat. Mater.* **2010**, *9* (7), 579–585.
- (35) Borisov, P.; Hochstrat, A.; Chen, X.; Kleemann, W.; Binek, C. Magnetoelectric Switching of Exchange Bias. *Phys. Rev. Lett.* **2005**, *94* (11), 117203.
- (36) Fiebig, M. Revival of the Magnetoelectric Effect. *J. Phys. D: Appl. Phys.* **2005**, *38* (8), R123–R152.
- (37) Lenz, K.; Zander, S.; Kuch, W. Magnetic proximity Effects in Antiferromagnet/Ferromagnet Bilayers: the Impact on the Neel Temperature. *Phys. Rev. Lett.* **2007**, *98* (23), 237201.
- (38) Ambrose, T.; Chien, C. L. Finite-Size Effects and Uncompensated Magnetization in Thin Antiferromagnetic Co Layers. *Phys. Rev. Lett.* **1996**, *76* (10), 1743–1746.
- (39) Yuan, W.; Wang, T. Y.; Su, T.; Song, Q.; Xing, W. Y.; Chen, Y. Y.; Han, W. Positive Exchange Bias between Permalloy and Twined (10–10)-Cr₂O₃ Films. *J. Magn. Magn. Mater.* **2017**, *422*, 397–401.
- (40) Yuan, W.; Zhu, Q.; Su, T.; Yao, Y. Y.; Xing, W. Y.; Chen, Y. Y.; Ma, Y.; Lin, X.; Shi, J.; Shindou, R.; Xie, X. C.; Han, W. Experimental Signatures of Spin Superfluid Ground State in Canted Antiferromagnet Cr₂O₃ via Nonlocal Spin Transport. *Sci. Adv.* **2018**, *4* (4), No. eaat1098.
- (41) Ohno, H.; Chiba, D.; Matsukura, F.; Omiya, T.; Abe, E.; Dietl, T.; Ohno, Y.; Ohtani, K. Electric-Field Control of Ferromagnetism. *Nature* **2000**, *408* (6815), 944–946.
- (42) Chang, C. Z.; Zhang, J. S.; Liu, M. H.; Zhang, Z. C.; Feng, X.; Li, K.; Wang, L. L.; Chen, X.; Dai, X.; Fang, Z.; Qi, X. L.; Zhang, S. C.; Wang, Y. Y.; He, K.; Ma, X. C.; Xue, Q. K. Thin Films of Magnetically Doped Topological Insulator with Carrier-Independent Long-Range Ferromagnetic Order. *Adv. Mater.* **2013**, *25* (7), 1065–1070.
- (43) Nogues, J.; Schuller, I. K. Exchange Bias. *J. Magn. Magn. Mater.* **1999**, *192* (2), 203–232.
- (44) Nogues, J.; Lederman, D.; Moran, T. J.; Schuller, I. K. Positive Exchange Bias in FeF₂-Fe Bilayers. *Phys. Rev. Lett.* **1996**, *76* (24), 4624–4627.
- (45) Nogues, J.; Leighton, C.; Schuller, I. K. Correlation between Antiferromagnetic Interface Coupling and Positive Exchange Bias. *Phys. Rev. B: Condens. Matter Mater. Phys.* **2000**, *61* (2), 1315–1317.
- (46) Yang, D. Z.; Du, J.; Sun, L.; Wu, X. S.; Zhang, X. X.; Zhou, S. M. Positive Exchange Biasing In GdFe/NiCoO Bilayers with Antiferromagnetic Coupling. *Phys. Rev. B: Condens. Matter Mater. Phys.* **2005**, *71* (14), 144417.
- (47) Seki, S.; Ideue, T.; Kubota, M.; Kozuka, Y.; Takagi, R.; Nakamura, M.; Kaneko, Y.; Kawasaki, M.; Tokura, Y. Thermal Generation of Spin Current in an Antiferromagnet. *Phys. Rev. Lett.* **2015**, *115* (26), 266601.
- (48) Arrott, A. Criterion for Ferromagnetism from Observations of Magnetic Isotherms. *Phys. Rev.* **1957**, *108* (6), 1394–1396.