Exchange-biasing topological charges by antiferromagnetism

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Geometric Hall effect is induced by the emergent gauge field experienced by the carriers adiabatically passing through certain real-space topological spin textures, which is a probe to non-trivial spin textures, such as magnetic skyrmions. We report experimental indications of spin-texture topological charges induced in heterostructures of a topological insulator (Bi, Sb)\textsubscript{2}Te\textsubscript{3} coupled to an antiferromagnet MnTe. Through a seeding effect, the pinned spins at the interface leads to a tunable modification of the averaged real-space topological charge. This effect experimentally manifests as a modification of the field-dependent geometric Hall effect when the system is field-cooled along different directions. This heterostructure represents a platform for manipulating magnetic topological transitions using antiferromagnetic order.
Spin-polarized carriers adiabatically moving through certain real-space topological spin textures can obtain a Berry’s phase as though they were in an applied magnetic field, resulting in a transverse carrier transport. Induced by this transverse transport, an extra Hall voltage can be observed, which is proportional to neither the applied external field nor the total magnetization. This spin-texture-induced extra Hall component is usually referred to as the topological or geometric Hall effect (GHE), which is a real-space counterpart of the $k$-space Berry phase in an intrinsic anomalous Hall effect (AHE). GHE is typically observed near magnetic reversal, within a window of the applied magnetic field and the temperature. Since the discovery of magnetic skyrmions in B20 compounds and heavy-metal multilayers, GHE has been considered as an experimental signature of topological spin textures (such as skyrmions), and along with other real-space detection methods, it enables mapping of the magnetic topological phase diagram. The topological charge of a real-space spin texture $S(r)$ is given by $N = \frac{1}{2\pi} \oint \mathbf{r} \times \mathbf{S}$.

In addition to the ferromagnet/TI interface, recent developments in topological antiferromagnetic (AFM) spintronics have demonstrated rich physics at the interface between a TI thin film and an AFM layer. At this interface, the surface Dirac fermions interact with the terminating magnetic atoms on the AFM surface, giving rise to interesting emergent phenomena, such as an interfacial ferromagnetic (FM) order. Such FM order breaks the time-reversal symmetry of the TI surface, inducing a finite Berry curvature and resulting in an AHE. With this FM layer stabilized by the AFM layer, the proximity-induced AHE survives at a much higher temperature than that in typical magnetically doped TIs. This scenario requires the interfacial FM layer to strongly couple with the AFM layer, and has been experimentally observed through exchange bias of the AHE and a staggered magnetic switching in a magnetic TI/AFM heterostructure.

Here, we report an experimental observation of the GHE modulated by uncompensated pinned spins in the AFM layer at the interface between an intrinsic TI thin film of $(\text{Bi,Sn})_{2}\text{Te}_{3}$ and an AFM layer of MnTe. This suggests that a topologically non-trivial chiral spin texture is induced in the TI through interactions with the spin-polarized Mn planes of the MnTe. We find that the magnetic topological charge can be manipulated by a seeding effect of pinned spins in the AFM layer. Systematic experimental results of the carrier magnetotransport, neutron scattering, and magnetic X-ray absorption (XA) spectroscopy support that the interfacial FM layer is induced in the TI through proximity interactions with the AFM layer.

**Results**

**Topological insulator and antiferromagnet heterostructures.** The presence of an induced FM order at the TI/AFM interface is manifested as an AHE effect observed in the magnetotransport measurements. In this experiment, the TI thin film, $(\text{Bi,Sn})_{2}\text{Te}_{3}$, was epitaxially grown on the top of a lattice-matched AFM, NiAs-phase MnTe, using molecular beam epitaxy (see Methods). The crystal quality is evidenced by the streaky reflection high-energy electron diffraction (RHEED) patterns, whereas the quality of the interface can be seen in the high-resolution transmission electron microscopy image, as shown in Fig. 1a, b, respectively. An atomic-resolution energy-dispersive X-ray spectroscopy (EDX) line-scan was measured across the $(\text{Bi,Sn})_{2}\text{Te}_{3}$/MnTe interface (Fig. 1c), demonstrating the sharpness of the elemental profiles. Bulk MnTe is known to be a semiconductor A-type antiferromagnet (Néel temperature $T_N \approx 307$ K), with parallel Mn spins in each basal plane. The spins between adjacent planes are anti-parallel, as shown in Fig. 1d inset. To confirm this magnetic order in the MnTe thin film, neutron diffraction experiments were carried out and the results are summarized in Fig. 1d, e. A (0001) peak (Fig. 1d) was observed as the signature of this magnetic order, which vanishes above $T_N \approx 300$ K (Fig. 1e), similar to the $T_N$ of the bulk. A possible interfacial spin structure is shown in Fig. 1f, and is discussed in more detail below. The AHE is plotted in Fig. 1g, where a hysteretic AHE is captured by scanning the applied perpendicular magnetic field. Likewise, a control sample of a pure MnTe thin film is shown in Fig. 1h. Note that the linear component corresponding to the ordinary Hall effect is not subtracted from the measured data. When the applied field is large (e.g., above $\pm 1$ T), the $R_{xy}$ curve saturates to a line with a finite intercept at $B = 0$, indicating the sign and the magnitude of the AHE component. Below $\sim 12$ K, an unexpected transition occurs: the AHE component of $R_{xy}$ is negative for the positive field, and positive for the negative field. This polarity is opposite to those above $12$ K, where the AHE has the same sign as the magnetization. To clearly demonstrate this trend, the magnitude of AHE ($R_{xy}$) is extracted from Fig. 1g as shown by the red curve in Fig. 1i. In addition to the change of sign, the magnitude of the Hall resistance surprisingly overshoots the saturated value, which is attributed to a GHE, as discussed below.

**Origins of anomalous Hall effects.** The presence of two AHE components with opposite signs at different temperature ranges (Fig. 1g) suggests a significant change in charge transport scenario. To demonstrate this, the longitudinal resistance as a function of temperature [$R_{xx}(T)$] was measured using control samples: a pure MnTe thin film, a pure TI thin film, and the combined heterostructure, as plotted in Fig. 1. In this figure, the pure TI thin film (blue) exhibits metallic behavior below $\sim 20$ K, since the thermal broadening is narrow enough to exclude the scattering given by the bulk states so that the transport is dominated by the TI surface. On the other hand, MnTe (red) behaves as a semiconductor up to $\sim T_N$, and the resistance drops faster with increasing temperature compared to the TI layer. Therefore, it is likely that the TI surface dominates the low-temperature ($<20$ K) transport, whereas the MnTe layer dominates in high temperature.

The AHE contributions of the TI and MnTe layers are different, likely originating in interfacial proximity interactions and MnTe bulk defect spins, respectively. As illustrated schematically in Fig. 1f, since the spins of the front-most Mn atoms cannot be fully compensated, they are easy to tilt out-of-plane in an applied perpendicular field [highlighted near the interface in Region (I) of Fig. 1f]. These tilted surface spins may induce magnetic moments with z components on the TI side through exchange coupling (white arrows with beige spheres). In this way, a proximity-induced FM order on a TI surface will break the time-reversal symmetry and induce an intrinsic AHE. The MnTe contribution likely arises from defects in the bulk MnTe that yield uncompensated spins [Region (II) in Fig. 1f], which can function as spin-dependent scattering centers, resulting in another AHE component. This is confirmed by referring to the data of the control sample prepared in the same
condition, in which the AHE in a single MnTe layer does not change polarity (Fig. 1b). Note that the illustration in Fig. 1f suggests that both spin-sublattices can induce magnetization in the TI layer, and therefore the proximity-induced transport signatures can survive even with surface roughness and terraces. When the temperature is below 12 K, no AHE was observed from the control sample, suggesting that the negative AHE at $T < 12$ K observed in the TI/AFM heterostructure (Fig. 1g) was indeed associated with the TI layer.

As mentioned before, in the TI/AFM heterostructure, when $T < 20$ K, an extra GHE component appears right before the saturation of $R_{xy}$ during each magnetic reversal, suggesting the presence of topological spin textures such as skyrmions. For example, at 1.9 K (Fig. 1g), the field scan was initiated from $+2$ T along $+z$, where $R_{xy}$ is negative. As the field is swept to $−z$ direction, $R_{xy}$ reverses at about $−0.2$ T and becomes positive. As the field along $−z$ is increased further, $R_{xy}$ continues to climb and peaks at about $−0.7$ T, after which it drops to the value coinciding with that of a saturated magnetization at a large field, e.g., $−2$ T. Such a non-monotonic field dependence suggests the presence of a GHE. In Fig. 1i, the magnitude of this GHE is extracted from Fig. 1g at different temperatures, shown by the black curve. Since both the low-temperature AHE and the GHE signal occur only in the heterostructure, the GHE is likely induced at the TI/AFM interface as discussed before. Thus, the total Hall resistance can be considered to have three components: $R_{xy} = R_{xy}^{G} + R_{xy}^{AHE} + R_{xy}^{G}$, where $R_{0}$ denotes the ordinary linear Hall resistance, $R_{xy}^{AHE}$ is the anomalous Hall resistance, and $R_{xy}^{G}$ is the geometric Hall component induced by topological spin textures. Here, $R_{xy}^{G}$ is proportional to the ensemble average of the topological charge density $\langle N \rangle$, which is determined by the topological order of the real-space spin texture.

**Exchange-biased geometric Hall effects.** More importantly, the observed magnitude of $R_{xy}^{G}$ is modulated depending on the field-cooling (FC) direction, i.e., $R_{xy}^{G} = R_{xy}^{G}(+z) \pm \delta R_{xy}^{G}$, where $R_{xy}^{G}(+z)$ is the GHE component obtained after zero FC (ZFC), $\pm$ is for the positive FC, whereas $−$ is for negative FC. This suggests that $\langle N \rangle$ can be exchange-biased by the pinning spins frozen after the FC: $\langle N \rangle = \langle N \rangle_0 \pm \delta \langle N \rangle$. As exhibited in Fig. 2a, this effect was captured by heating the heterostructure to 350 K and then cooled to 1.9 K in an applied field of 5 T along $+z$ (positive FC) or $−z$ (negative FC). Although the Néel vector of MnTe lies within the $x$-$y$ easy plane, some AFM surface spins may still be canted and

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Fig. 1 Emergent geometric Hall effect in a TI and antiferromagnet bilayer. a Reflection high-energy electron diffraction patterns of TI $(Bi,Sb)_2Te_3$ and antiferromagnet MnTe. b A high-resolution transmission electron microscopy image of the bilayer cross-section. The scale bar represents 2 nm. c An atomic-resolution energy-dispersive X-ray spectroscopy (EDX) line-scan across the bilayer interface, demonstrating the sharpness of the elemental profiles at this interface. d The neutron diffraction measurement of the MnTe (0001) peak at 100 and 300 K. e The fitted temperature-dependent peak height suggests a $T_0$ of ~300 K, consistent with the one shown in (d). f Schematics illustrating the spins in the TI bottom surface, the interfacial FM layer, and the AFM spins in the bilayer under a perpendicular magnetic field. The polarized spins in the TI bottom surface is induced by the TI-AFM proximity (Region I), while some small magnetizations in the AFM layer may come from the defects in the bulk (Region II).
obtained by subtracting the normalized magnetization (pink) from the applied perpendicular exchange bias experienced by the topological charges after a negative FC process. Such an asymmetric phase diagram contrasts with the symmetric one obtained after a ZFC process, demonstrating the negative ones (green circles). The anchoring spins in the AFM layer assist the nucleation of positive topological charges (red circles) while prohibiting the nucleation of negative topological charges during the above four scenarios. The microscopic picture is schematically shown by the pink arrows. While the spin-pinned regions, but with a lower density (green circles), these positive topological charges are annihilated. When the field sweeps to I, positive topological charges (red circles) move from the pinned spins. After the saturation along a negative field II, negative topological charges are prohibited by the AFM layer, which is symmetric with respect to the $B = 0$ line. The red $R_{xy}$ component grows while the blue one shrinks after a negative FC process. Such an asymmetric phase diagram contrasts with the symmetric one obtained after a ZFC process, demonstrating the exchange bias experienced by the topological charges.

**Theoretical model of the seeding effect.** This modulation of $\delta R_{xy}^G$ may stem from a seeding effect induced by the anchoring spins in the AFM layer frozen after the FC process. Experimentally, it has been shown that special domain nucleation patterns can be induced by the spins in an adjacent AFM layer due to interfacial exchange coupling. Here we show that this exchange coupling can result in a seeding effect for the spin-texture topology. The microscopic picture is schematically shown by the four scenarios illustrated in Fig. 2e. After positive FC, some pinned spins can be frozen in the AFM layer along the FC direction (black arrows) due to thermoremanent effects. When the applied field sweeps to I, positive topological charges [red circles with up central spins in I] are created through interactions with the pinned spins. After the saturation along a negative field II, these positive topological charges are annihilated. When the field scans to III, negative topological charges are prohibited by the pinning spins and therefore are more likely to nucleate outside the spin-pinned regions, but with a lower density (green circles with down central spins in III). These negative charges again vanish after magnetic saturation IV. The modification of the topological charge density during the above four scenarios can be described as $\langle N \rangle_x = \langle N \rangle_0 \pm \delta(N)$. This results in a shift of the Hall signal: $R_{xy}^{G} = R_{xy}^{G(0)} + \delta R_{xy}^{G}$. A similar modulation of $-\delta R_{xy}^{G}$ and $-\delta(N)$ occurs after a negative FC. To further understand this...
Effect, a micromagnetic simulation based on a toy model was carried out, mimicking the thin FM layer at the TI/MnTe interface. Anchoring spins were randomly generated in the simulation plane, with their spins $\{S_{pin}\}$ fixed along either $+z$ to mimic a positive FC or $-z$ for a negative one. These anchoring spins interact with the spin texture $\{S_{i}\}$ through the exchange Hamiltonian $h_{pin} = -J_0 \sum_{i} S_{pin} \cdot S_{i}$. Dynamical behavior of the spin texture was obtained by solving Landau–Lifshitz–Gilbert equation, with a stochastic field describing the effect of the thermal fluctuation (see Methods). Although the magnetic topological charge can arise from different spin textures, in this model we use magnetic skyrmions for simplicity. The average total topological charge, $\langle N \rangle$, was tracked by averaging over 300 reversals of the external field, as shown in Fig. 3a. Snapshots during one of the 300 simulations are shown in Fig. 3b–d. When the anchoring spins are fixed along $+z$, the spin texture first presents a phase of helical domains (Fig. 3b), and then the helical domains break and shrink into individual positive skyrmions (Fig. 3c). When this occurs, $\langle N \rangle$ is maximized. As the field is increased along $-z$, Zeeman coupling dominates, annihilating all topological charges and resulting in a saturated FM order along $-z$. As the field is scanned back from negative to positive, the spin texture evolves in an analogous way, with the sign of $\langle N \rangle$ reversed (Fig. 3d). A movie recording this dynamical process among one of the 300 simulations can be found in Supplementary Movie 1. Strikingly, the modulation of the topological charge density is numerically captured, namely, $\langle N \rangle_+ = \langle N \rangle_0 + \delta \langle N \rangle$ for positive FC, whereas $\langle N \rangle_- = \langle N \rangle_0 - \delta \langle N \rangle$ for negative FC, suggesting that the topological charge density is determined by the direction of the pinning spins with respect to the sign of the created topological charge. This is in agreement with our experimental results. To further demonstrate this point, using the $+z$ pinning as an example, the averaged topological charge areal density, $\langle \rho(\mathbf{r}) \rangle = \frac{1}{bc} \sum_i \langle \partial_i S \times \partial_i S \rangle$, is plotted in Fig. 3e, f, for the cases of II and III, respectively. The positions of the anchoring spins are shown by the solid black dots, whereas the color illustrates the map of $\langle \rho(\mathbf{r}) \rangle$. As shown by the topological charge density, $+z$ anchoring spins assist in the nucleation of positive charges but prohibit negative ones. Induced by the exchange coupling $h_{ach} = -J_0 \sum_{i} S_{i} \cdot S_{ach}$, positive $\langle N \rangle$ are energetically preferred to coincide with $+z$ anchoring spins, since the spin at each skyrmion core is parallel with the anchoring spin (Fig. 3g). Negative skyrmions, on the other hand, have their central spins anti-parallel with the anchoring spins, and therefore are more difficult to stabilize (Fig. 3h). Note that the creation and annihilation of a topological charge only involve several local spins, therefore the seeding effect of topological charge creation in this simulation should also be expected in other more complex topological spin textures.

Mechanisms of the exchange-biased topological charges. An important question in the observed exchange-biased GHE is the origin of the induced FM layer at the TI/AFM interface. This layer could be induced either by the proposed magnetic proximity effect, or by the migration of enough Mn from the AFM layer into the TI layer. The latter would result in a magnetically doped TI,
Mn\textsubscript{6}(Bi,Sb)\textsubscript{2}Te\textsubscript{3}, where the observed 20 K onset temperature of the GHE would require that δ is significantly in excess of 0.15 in order to achieve the necessary Curie temperature\textsuperscript{29,30}. Such doping-induced ferromagnetism would be accompanied by an increase in the Mn valence from 2+ toward 3+ and a significant enhancement of the X-ray magnetic circular dichroism (XMCD) magnitude\textsuperscript{31,32}. We therefore probed Mn valence and magnetic ordering using total electron yield (TEY) soft XA spectroscopy and XMCD, shown in Fig. 4a, b. When using the TEY mode, the probing depth is approximately 5 nm\textsuperscript{33}, increasing sensitivity to the nuclear and magnetic scattering length densities (SLD) used to obtain the ferromagnetic alignment of moments. The error bars are one standard deviation.

On one hand, the detected Mn L\textsubscript{3,2} edge XMCD of a capped sample (black) (15 K) probing the in-plane and out-of-plane ferromagnetic alignment of moments. The error bars are one standard deviation. c Antiferromagnetic order probed by XLD. Data give a comparison of a Mn L\textsubscript{3} edge XLD for both capped and uncapped samples. The XMCD and XLD data suggest that the AFM order of the MnTe is not altered by the TI capping layer. d Fitted polarized neutron reflectometry of the TI/MnTe heterostructure measured at 7.5 K in a 700 mT in-plane applied field. e Spin asymmetry of the measurement with the fit shown in (d). Error bars represent one standard deviation. f Structural and magnetic depth profile are represented by the nuclear and magnetic scattering length densities (SLD) used to obtain the fit shown in (d) and (e). These polarized neutron reflectometry results (d, e, and f) suggest an interfacial ferromagnetic layer was induced in the Ti/AFM heterostructure.
densities (SLD) nuclear profile is attributed to the terraces and long-range surface roughness typically show up in epitaxial Ti growth, and does not indicate intermixing. The magnitude of the induced moment at the interface is comparable to other examples of interfacial proximity interactions reported in the literature. Note that alternative models (Supplementary Figs. 3, 4), which do not incorporate an interfacial magnetization in the Ti, fail to describe the data. For more information on the PNR fitting process and alternative models, see Supplementary Note 2.

In summary, we have experimentally demonstrated the seeding of topological charges at a Ti/AFM interface induced by the pinning spins. The exchange-biased topological charges are experimentally observed as a modulation of the GHE magnitude captured in a transport measurement under different FC conditions. This suggests that the creation of topological charges in a FM spin texture can be strongly affected by an adjacent antiferromagnet. Further X-ray and neutron experiments suggest that this FM order is likely induced by the proximity effect, rather than unintentional Mn migration.

Methods

Growth of magnetic heterostructures. All heterostructures in this work were fabricated on epi-ready semi-insulating GaAs(111)B substrates in an ultrahigh vacuum molecular beam epitaxy system. Prior to sample growth, oxide-desorption processes were carried out at 850 °C for 30 min. A thin insulating NiAs-type CrSe layer serves as a buffer for the growth of the MnTe layer. The substrate temperature was then maintained at 380 °C during the growth of MnTe and 200 °C for (Bi,Sb)2Te3 layers, whereas Se, Sb, and Te were evaporated by standard thermal cracker cells. Real-time RHEED (as shown in Fig.1a) was used to monitor all the growth cycles. The growth conditions of CrSe, MnTe, and (Bi,Sb)2Te3 layers were optimized such that the growth rate, the Heisenberg exchange coupling between nearest neighbors, $D$ denotes the DMI magnitude, $H$ is the applied field, and $L$ is the exchange coupling between the pinning spins ($S^{\parallel}$) and the spin texture, $\langle S^{\parallel} \rangle_{L} = 1$ and $\langle S^{\perp} \rangle = 0.8$ are applied as an example. The spin dynamical process was obtained by solving the Landau-Lifschitz-Gilbert equation $\dot{S} = -\gamma S \times H_{\text{eff}} - \alpha S \times \dot{S}$, where $\gamma$ is the gyromagnetic ratio, and $\alpha = 0.1$ is the damping coefficient. The effective field is $H_{\text{eff}} = \frac{H}{1 + L}$, where $L$ is the stochastic field satisfying dissipation–fluctuation relation $\langle \dot{S}(t) \rangle L \langle \dot{S}^{\parallel}(t') \rangle = \delta(t - t') \delta_{\mu} \delta_{\nu} \delta_{\alpha} \delta_{\beta} (\mu, \nu = x, y, z)$, and $\xi = \frac{4\pi}{2\alpha}$. Here, $\frac{H}{1 + L}$ is 0.3 is used as an example to mimic the thermal fluctuation given by the finite temperature.

X-ray absorption spectroscopy and dichroisms. X-ray spectroscopy at the Mn L3,2 absorption edge (635–660 eV) was performed using TEY as the detection mode at beamline 4.0.2 of the Advanced Light Source at Lawrence Berkeley National Lab. All XA spectra were normalized to the intensity of the non-resonant pre-edge region; i.e., at energies below the L3,2 absorption edge. XMC and XLD spectra were further normalized to allow direct comparisons, by shifting the pre-edge intensity to 0 and then scaling to set the polarization-averaged height of the most intense peak to 1.

Induced FM moments were probed by XMC, in which photon energy scans were measured with 90% circularly polarized X-rays in applied magnetic fields of $\pm 0.4 T$. The XMC was obtained as the difference between scans with opposite magnetic fields, and confirmed by also reversing the circular polarization. The X-ray beam was oriented along either a 30° grazing incidence or at (90°) normal incidence to probe in-plane or out-of-plane FM moments, respectively.

XLD was utilized to probe AFM order by measuring linear dichroism at 15 and 300 K. Without any structural phase transition in that range, the crystal field effects are presumed to be constant vs. temperature, so that the difference in dichroism can be attributed mainly to the magnetic contribution. XLD was measured in the 30° grazing incidence configuration by rotating X-rays, and by rotating the linear polarization of X-rays between two orthogonal settings: fully in-plane (vertical polarization) and 30° from the film normal (horizontal polarization).

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Author contributions
Q.L.H., G.Y., and K.L.W. conceived and designed the experiments. K.L.W. supervised the work. Q.L.H. and L.P. grew the samples. X.C. fabricated the devices. G.Y., Y.L. and R.K.L. performed the theoretical calculations and simulations. B.Z. and X.H. performed the neutron diffraction. B.J.K. performed the neutron reflectometry and analysis. All the authors contributed to the measurements and analyses. Q.L.H., G.Y., A.J.G. and K.L.W. wrote the manuscript with contributions from all the authors.

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