Ambi-chiral anomalous Hall effect in magnetically doped topological insulators

Chang Liu¹,²†, YunYi Zang¹,²†, Yan Gong¹, Ke He¹,²,³*, XuCun Ma¹,³, QiKun Xue¹,²,³, and YaYu Wang¹,³*

¹ State Key Laboratory of Low Dimensional Quantum Physics, Department of Physics, Tsinghua University, Beijing 100084, China; ² Beijing Academy of Quantum Information Sciences, Beijing 100193, China; ³ Frontier Science Center for Quantum Information, Beijing 100084, China

Received December 14, 2021; accepted March 1, 2022; published online April 29, 2022

The chirality associated with broken time-reversal symmetry in magnetically doped topological insulators has important implications for the quantum transport phenomena. Here we report anomalous Hall effect studies in Mn- and Cr-doped Bi₂Te₃ topological insulators with varied thicknesses and doping contents. By tracing the magnitude of the anomalous Hall resistivity, we find that the Mn-type anomalous Hall effect characterized with clockwise chirality is strengthened by the reduction of film thickness, which is opposite to that of the Cr-type anomalous Hall effect with counterclockwise chirality. We provide a phenomenological physical picture to explain the evolution of the magnetic order and the anomalous Hall chirality in magnetically doped topological insulators.

topological insulator, anomalous Hall effect, chirality, magnetic order

PACS number(s): 73.20.At, 73.25.+i, 73.50.Jt, 85.70.Kh

Citation: C. Liu, Y. Y. Zang, Y. Gong, K. He, X. C. Ma, Q. K. Xue, and Y. Y. Wang, Ambi-chiral anomalous Hall effect in magnetically doped topological insulators, Sci. China-Phys. Mech. Astron. 65, 266812 (2022), https://doi.org/10.1007/s11433-021-1883-x

1 Introduction

Chirality plays crucial roles in a wide range of systems, ranging from fundamental particles in high energy physics to grand-scale objects like spiral galaxies in astrophysics. Generally, an object is called chiral when it cannot be distinguished from itself under reflection or inversion operation [1]. Representative examples include human hands in biology, chiral molecules in chemistry, spin systems in physics, etc. In condensed matter physics, chiral phenomena not only manifest themselves in material structure but also in emergent electromagnetic responses, such as skyrmions in chiral magnets, and chiral anomaly phenomena in Dirac-Weyl semimetals [1-6]. Magnetic topological insulators (TIs) represent versatile platforms for exploring the exotic topological quantum phenomena with chiral nature. For example, the ferromagnetic (FM) order breaks the time-reversal symmetry (TRS) of TIs and endows the Dirac fermions with finite mass through the exchange coupling [7-14]. The helicity of the topological surface states is lifted and the resulting nonzero Berry curvature leads to a pronounced anomalous Hall effect (AHE) that exhibits hysteresis with a specific chirality. In the two-dimensional (2D) limit when the Fermi level is tuned to the band gap, the one-dimensional (1D) helical channels of TIs are split into chiral edge states that contribute to the quantized AHE [15-17]. The realization

*Corresponding authors (Ke He, email: kehe@tsinghua.edu.cn; YaYu Wang, email: yayuwang@tsinghua.edu.cn)
†These authors contributed equally to this work.
of the quantized AHE undoubtedly demonstrates the critical role of Berry curvature in the electronic properties of TIs and provides a new building block for realizing other exotic topological quantum phenomena such as magnetic monopoles and chiral Majorana fermions [18-26].

Despite considerable efforts on magnetic TIs, particularly in pursuing the quantized AHE, there are still intriguing open questions. In previous studies, the AHE behaviors in magnetic TIs can be divided into two different groups. In the first group, exemplified by the Cr- and V-doped Bi$_2$Se$_3$ family TI films, the magnetic-field-dependent Hall resistivity $\rho_{yx}$ traces display hysteresis loops with counterclockwise chirality [15-17,27-29], as schematically displayed on the left of Figure 1(a). Here, $\rho_{yx}^+$ and $\rho_{yx}^-$ denote the $\rho_{yx}$ profiles when the magnetic field is swept back to zero from positive and negative polarizations, respectively. The violet arrows mark the magnetization at the corresponding magnetic field. At positive magnetization, the AH resistivity $\rho_{yx}^0$ (defined as $\rho_{yx}^+$ at zero magnetic field) has a positive value. For the quantized AHE such as in the Cr-doped (Bi,Sb)$_2$Te$_3$ TI system, $\rho_{yx}^0$ reaches $h/e^2$, where $h$ denotes the Planck constant and $e$ is the electron charge, corresponding to the Chern number $C = +1$ [15-17]. In the second group, represented by Mn-doped Bi$_2$Te$_3$, the Hall resistivity traces display a hysteresis loop with clockwise chirality and the sign of $\rho_{yx}^0$ is negative [30-32]. Thus, for its quantized AHE version, the Chern number is $C = -1$ and $\rho_{yx}^0$ is quantized at $-h/e^2$, which has recently been realized in MnBi$_2$Te$_4$ intrinsic antiferromagnetic TI with an odd number of layers [33]. The schematic picture of the AHE with clockwise chirality is displayed on the right of Figure 1(a).

Accompanied by the rise of chiral spintronics in topological matters [1], the chirality issue of the AHE has attracted more attention in recent years. A variety of exotic topological quantum phenomena are found to be associated with chirality [20,30,34-38]. For example, theoretical calculations propose that the opposite chiralities of the AHE in Mn- and Cr-doped Bi$_2$Te$_3$ can be utilized to realize the long-sought quantized topological magnetoelectric effect when they are constructed into heterostructures [20]. Very recently, transport studies in the pulsed magnetic field demonstrated that the Chern insulator phase in Mn-based magnetic TI with clockwise chirality and $C = -1$ evolves into a helical phase with $C = 0$ [33,39,40]. In contrast, the Chern insulator phase in Cr-based magnetic TI with counterclockwise chirality and $C = +1$ becomes more stable in the high field limit [15]. These results provide more motivations for understanding and controlling the chirality of AHE in magnetic TIs.

Although there have been several experiments studying the AHE in Mn- and Cr-doped TIs [15-17,27-31,41,42], for a certain magnetic TI thin film, the chirality of AHE is usually fixed. It is difficult to switch between opposite chiralities in one sample. Moreover, the different sample fabrication methods [43,44], selections of dopants and parent TI compounds [44-50], effects of interface [34,35], and even dimensionalities [30,31,41,42], make it elusive to determine how the AHE with opposite chiralities evolves under different parameters. Therefore, it is highly desired to have accurate control of parameters in one system to study the chirality of the AHE.

In this work, we report systematic transport studies of Cr- and Mn-doped (Bi$_{1-2x}$Mn$_x$Cr$_{1-x}$)$_3$Te$_3$ magnetic TIs with various thicknesses $d$ and doping contents $x$ at different temperatures $T$ and gate voltages $V_g$. By tracing the magnitude of $\rho_{yx}^0$, we find that the Mn-type AHE with clockwise chirality is strengthened as $d$ is reduced from eight quintuple-layers (QLs) to six QLs, whereas the Cr-type AHE characterized with counterclockwise chirality exhibits an opposite behavior. The different response of AHE to film thickness is universally present in the $x$, $T$, and $V_g$-dependent measurements, consistent with the distinct mechanisms for FM order found in Mn- and Cr-doped magnetic TIs. We propose a phenomenological physical picture to explain the observed ambi-chiral AHE in magnetically doped TIs.

2 Methods

The ten magnetic TI samples studied in this work were grown by molecular beam epitaxy (MBE) on insulating...
SrTiO$_3$ (111) substrates following the similar method reported previously [15,30,51]. Mn and Cr are uniformly deposited during sample growth. The SrTiO$_3$ substrate also acts as a back gate due to its large dielectric constant at low $T$. Figure 1(b) shows the crystal structure and the schematic cross-section of a (Bi$_{0.9}$Mn$_x$Cr$_{0.1-x}$)$_2$Te$_3$ TI. The black and red arrows mark the Mn and Cr magnetic moments, respectively. Two film thicknesses (eight and six QLs) are chosen because they are small enough to ensure sufficient gating ability and large enough to avoid the effect of surface hybridization on the magnetic properties [30,32,52].

For transport measurement, the standard four-probe AC lock-in method is carried out after the samples are manually scratched into Hall bar structures. Indium pieces are pressed directly on the surface of the samples as electrical contacts. The magnetic field is applied perpendicular to the sample plane, and the Hall signals are antisymmetrized with respect to the magnetic field to correct the errors caused by geometric misalignments. An excitation current of 0.2 μA is applied by a Keithley 6221 current source, and a $V_g$ is applied by a Keithley 2400 source meter. For the convenient comparison between the current results and the previous studies of the AHE in magnetic TIs [28,30,43,53], throughout this work, we use $\rho_{yx}^0$ to characterize the AHE.

### 3 Experimental results

We first studied the magnetic-field-dependent $\rho_{yx}$ at different $T$’s for five (Bi$_{0.9}$Mn$_x$Cr$_{0.1-x}$)$_2$Te$_3$ samples with eight QLs, as shown in Figure 2(a). With the decrease of $T$, $\rho_{yx}$ exhibits a hysteresis loop as the magnetic field is swept back and forth, indicating the formation of the FM order. The blue and red arrows denote the opposite chiralities of the Hall traces. In the purely Cr-doped Bi$_2$Te$_3$ sample ($x = 0$), $\rho_{yx}^0$ reaches $\sim$1.5 kΩ at $T = 1.6$ K. With the increase of $x$, $\rho_{yx}^0$ progressively decreases as more Cr dopants are substituted by Mn. At $x = 0.08$, $\rho_{yx}^0$ is suppressed to a vanishingly small value. As $x$ is further increased to 0.10, the sample becomes purely Mn-doped, in which $\rho_{yx}^0$ turns negative and the chirality of the hysteresis loop becomes clockwise.

Next, we studied $\rho_{yx}$ in another set of samples but with six QLs, where the contribution from the bulk state is reduced. The overall behaviors are similar to that of the eight-QL samples, but there is an obvious difference in the $x$ value when the $\rho_{yx}^0$ sign change occurs. As indicated by the data set enclosed by the red frames, it is reduced from $x = 0.08$ in the eight-QL samples to $x = 0.07$ in the six-QL samples. To better visualize the thickness-dependent chirality change, we plot the difference between $\rho_{yx}^+$ and $\rho_{yx}^-$ (defined as $\Delta \rho_{yx} = \rho_{yx}^+ - \rho_{yx}^-$) as a function of magnetic field for different $x$ and $d$, as shown in Figure 2(c) and (d). The peak and valley in $\Delta \rho_{yx}$ near zero magnetic field directly reflect the chirality of the AHE. Clearly, in the thinner samples with six QLs, less Mn is required to tune the chirality from counterclockwise to clockwise. The effect of film thickness on the AHE is also reflected by the value of $\rho_{yx}^0$. In the purely Cr-doped sample, with the reduction of $d$, $|\rho_{yx}^0|$ decreases from $\sim$0.85 to $\sim$0.35 kΩ at $T = 1.6$ K. In contrast, $|\rho_{yx}^0|$ increases from $\sim$0.15 to $\sim$0.25 kΩ in the purely Mn-doped sample. The above results suggest that for fixed doping content, reducing the film thickness strengthens the Mn-type AHE with clockwise...
chirality, whereas weakens the Cr-type AHE with counterclockwise chirality. Because the AHE is sensitive to Fermi level, which may be affected by film thickness, we explore the magnetic-field-dependent $\rho_{yx}$ at varied $V_g$ in these samples. Figure 3(a) and (b) display the main results for the two critically doped samples ($x = 0.08$ for eight-QL and $x = 0.07$ for six-QL) that are close to the boundary of the chirality change. Interestingly, the chirality in the two samples exhibits quite different evolutions with the variation of $V_g$. For the eight-QL sample, the chirality remains counterclockwise for the entire $V_g$ range, suggesting that the Cr-related AHE dominates the transport. In contrast, for the six-QL sample, increasing $V_g$ dramatically changes the shape of the $\rho_{yx}$ loop and even causes a change in chirality. At $V_g = -100$ V, the hysteresis is clockwise, and the sign of $\rho_{yx}$ is negative. As $V_g$ increases to $-25$ V, the two $\rho_{yx}$ curves obtained from opposite field sweeps touch together at zero field, forming a hysteresis with ill-defined chirality. As $V_g$ is further increased to 100 V, the two $\rho_{yx}$ curves form a hysteresis loop again but now with counterclockwise chirality and positive $\rho_{yx}$. In the insets of Figure 3(b), we plot the zoomed-in data in the field range of 0.3 T, which clearly shows the change of chirality with $V_g$.

To visualize the $V_g$-dependent ambipolar AHE more clearly, we display the magnetic-field-dependent $\Delta \rho_{yx}$ for varied $V_g$'s, as shown in Figure 4(a) and (b). In the eight-QL sample, a positive peak near zero field is universally present throughout the $V_g$ range, in accord with the positive sign of $\rho_{yx}$. However, in the six-QL sample, the crossing of $\rho_{yx}^+$ and $\rho_{yx}^-$ curves give rise to a more complex evolution in $\Delta \rho_{yx}$, as displayed in Figure 4(b). With the decrease of $V_g$, the peak value at zero field shifts downward, accompanied by the sign change from positive to negative. Eventually, $\Delta \rho_{yx}$ becomes an overall negative curve characterized by two negative dips near zero field. The above $V_g$-dependent chirality change is highly reminiscent of the doping-dependent chirality change as shown in Figure 2(c) and (d).

To further illustrate the distinct thickness dependences of Mn- and Cr-type AHE, in Figure 4(c) and (d), we display the color maps of $\Delta \rho_{yx}$ as functions of magnetic field and $V_g$ for samples with different $x$ values. In each figure, $d$ is fixed and $x$ increases from panels (i) to (iii). The doped data represent the position where $\rho_{yx}^+$ and $\rho_{yx}^-$ cross, and the red and blue colors represent the area with $\Delta \rho_{yx} > 0$ and $\Delta \rho_{yx} < 0$. In the purely Mn-doped Bi$_2$Te$_3$, $\rho_{yx}^+$ is always smaller than $\rho_{yx}^-$ in the hysteresis regime, thus $\Delta \rho_{yx}$ is negative near zero field. Whereas in the purely Cr-doped sample, $\Delta \rho_{yx}$ has a positive value. Therefore, we can use the sign of $\Delta \rho_{yx}$ to characterize the Mn- and Cr-type AHE. Clearly, in the eight-QL samples, the Mn-type AHE is not favored until $x$ is increased to 0.08 and when negative $V_g$ is applied. However, in the six-QL samples, a smaller Mn doping level ($x = 0.05$) is sufficient to induce a similar trend in $\Delta \rho_{yx}$, as also shown by the growing area with $\Delta \rho_{yx} > 0$ (red). Again, it suggests that the Mn-type AHE is strengthened in thinner samples, opposite to that of the Cr-type AHE.

4 Discussion and conclusions

Previous theories have proposed that the topologically pro-

Figure 3  (Color online) Magnetic field dependent $\rho_{yx}$ for an eight-QL sample with $x = 0.08$ (a), and a six-QL sample with $x = 0.07$ (b) for varied $V_g$'s. The blue and red colors represent the data obtained from different field sweep directions, as labeled by $\rho_{yx}^+$ and $\rho_{yx}^-$, respectively. The insets present the zoomed-in data at a low magnetic field range of 0.3 T. All the data are acquired at $T = 1.6$ K.
tected surface states in TIs could be more susceptible to magnetic order than the insulating bulk states, thus is anticipated to give rise to a distinct magnetic ordering phenomenon in Mn-doped Bi$_2$Se$_3$ TIs [54]. There have been transport and spectroscopy experiments suggesting that the FM order in Mn-doped TIs is mediated by the surface states [32,45,47-49], while it is the bulk that mainly contributes to the FM order in Cr-doped systems [28,43,44,53]. The opposite thickness dependences of the Mn- and Cr-related AHE observed here suggest a scenario of dopant-selective surface and bulk magnetic order in Mn- and Cr-doped Bi$_2$Te$_3$, consistent with previous reports. In Figure 5(a) and (b), we summarize the $T$ dependent $\rho_{yx}$ for all ten samples, and the two insets show the zoomed-in data for the two critical samples. Clearly, not only the value of $|\rho_{yx}|$ but also the onset temperature ($T_{AH}$) responds oppositely to thickness in Mn- and Cr-doped Bi$_2$Te$_3$. As $d$ is reduced from eight to six QLs, $T_{AH}$ of the Mn-Bi$_2$Te$_3$ sample remains almost unchanged (red arrows). However, it decreases from 26 to 14 K (black arrows) in the Cr-Bi$_2$Te$_3$ sample. Such effect is more clearly visualized in the color plot of $\Delta\rho_{yx}$ in Figure 5(c) and (d). These results demonstrate that reducing $d$ from eight to six QLs has a more dramatic effect on the Cr-type AHE than the Mn-type one. According to previous measurements [52], the penetration depth of the topological surface state is about two QLs; thus reducing $d$ from eight to six QLs mainly suppresses the bulk volume while keeping the surface states less affected. These results strongly indicate that the thickness dependence of the chirality change in our samples is related to the different surface and bulk FM order in Mn- and Cr-doped TIs [28,30,32,43-45,47-49,53].

Finally, we discuss the possible factors that lead to the opposite chiralities in Mn- and Cr-doped TIs. Theoretically, the AHE in magnetic TIs is induced through the exchange coupling $J M_z S_z$ between local moments $M_z$ and electron spins $S_z$, where $J$ denotes the coupling constant [7,20,32]. For the fixed direction of $M_z$, opposite $J$ could endow Dirac fermions with opposite masses, thus leading to the opposite chiralities of the AHE. The opposite signs of the quantized AHE in MnBi$_2$Te$_4$ and Cr-(Bi,Sb)$_2$Te$_3$ unambiguously demonstrates the signs of Dirac mass are opposite in Mn- and Cr-based TIs [15,33], which implies the opposite chiralities in the current unquantized cases may arise from the same mechanism [20]. However, the microscopic mechanism of the sign of $J$ is still unclear. Here, we propose a phenomenological physical picture to address the chirality issue in terms of the $d$-orbital configurations of Mn and Cr dopants. According to previous studies, the spin states of Mn and Cr dopants in TIs are $S_z = 5/2$ and $3/2$, respectively [55,56]. Therefore, in Mn-based TIs, each of the five $3d$-orbits of Mn$^{2+}$ is singly occupied. When the itinerant electron spins couple with Mn$^{2+}$, only electrons with opposite spin are allowed to virtually hop to the unoccupied orbits. However, in Cr-based TIs, each Cr$^{3+}$ has three singly occupied and two
empty orbits. In this case, it is energetically more favorable for electrons with the same spin to hop to the empty orbits. Consequently, the exchange couplings and the resulting Dirac fermion masses in the two systems are expected to have opposite signs, which in turn leads to the opposite chiralities in the AHE. The above pictures are schematically displayed in Figure 5(e) and (f), respectively. To be noted, the Dirac mass sign is not the sole explanation for the sign change in magnetic TIs. Recent theories show that the spin-charge correlated disorder may also give rise to the AHE sign change even without considering flipping the Dirac mass [57]. More future experiments are required to pin down the underlying microscopic origins.

In summary, we performed systematic studies of the AHE in Mn- and Cr-codoped Bi$_2$Te$_3$ TI thin films. By varying the Cr/Mn doping level and film thickness, we can actively control the chirality of the AHE. These results can be interpreted by the different mechanisms for FM order and AHE chiralities in Mn- and Cr-doped TIs. Our experiments shed new light on manipulating the interplay between FM order and band topology in magnetic TIs.

This work was supported by the Basic Science Center Project of National Science Foundation of China (Grant No. 51788104) and the National Key R&D Program of China (Grant Nos. 2018YFA0307100, and 2018YFA0305603). This work was supported in part by the Beijing Advanced Innovation Center for Future Chip (ICFC). Chang Liu is grateful to Peizhe Tang from Beihang University for useful discussions.

---

**Figure 5** (Color online) (a), (b) Temperature-dependent $\rho_{xy}$ for ten samples with different thicknesses and doping levels. The insets display the zoomed-in data for the two critical samples with $x = 0.08$ for the eight-QL sample and $x = 0.07$ for the six-QL sample. (c), (d) Color plots of $\rho_{xy}$ as functions of doping level and temperature. The red and blue colors represent the regimes for $\rho_{xy} > 0$ and $\rho_{xy} < 0$, respectively. The magenta dots denote the onset temperatures of AHE in purely Mn- and Cr-doped Bi$_2$Te$_3$ samples. (e), (f) Schematic pictures of the Dirac gap opening process in Mn- and Cr-based TI systems. The green and red arrows represent the spin states for localized d-electrons and itinerant electrons, respectively.

1. S. H. Yang, R. Naaman, Y. Paltiel, and S. S. P. Parkin, *Nat. Rev. Phys.* 3, 328 (2021).
2. X. Z. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. H. Han, Y. Matsu, N. Nagaosa, and Y. Tokura, *Nature* **465**, 901 (2010).
3. S. Mulhauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neu-bauer, R. Georgii, and P. Boni, *Science* **323**, 915 (2009), arXiv: 0902.1968.
4. C. Train, R. Gheorghe, V. Krsic, L. M. Chamoreau, N. S. Ovanesyan, G. L. J. A. Rikken, M. Gruselle, and M. Verdaguer, *Nat. Mater.* **7**, 729 (2008).
5. N. Nagaosa, and Y. Tokura, *Nat. Nanotech.* **8**, 899 (2013).
6. N. P. Ong, and S. Liang, *Nat. Rev. Phys.* **3**, 394 (2021), arXiv: 2010.08564.
7. Y. Tokura, K. Yasuda, and A. Tsukazaki, *Nat. Rev. Phys.* **1**, 126 (2019).
8. X. L. Qi, and S. C. Zhang, *Rev. Mod. Phys.* **83**, 1057 (2011), arXiv: 1008.2026.
9. M. Z. Hasan, and C. L. Kane, *Rev. Mod. Phys.* **82**, 3045 (2010), arXiv: 1002.3895.
10. X. L. Qi, T. L. Hughes, and S. C. Zhang, *Phys. Rev. B* **78**, 195424 (2008), arXiv: 0802.3537.
11. R. Yu, W. Zhang, H. J. Zhang, S. C. Zhang, X. Dui, and Z. Fang,
