Crossover of Quantum Anomalous Hall to Topological Hall Effect in Magnetic Topological Insulator Sandwich Heterostructures

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The intrinsic anomalous Hall (AH) effect is a result of a non-zero Berry curvature in momentum-space. The quantized version of the AH effect, known as the quantum anomalous Hall (QAH) effect, harbors dissipation-free chiral edge states in the absence of an external magnetic field. On the other hand, the topological Hall (TH) effect, a transport hallmark of the nontrivial spin textures, is a consequence of real-space Berry curvature. While both the QAH and TH effect have been reported separately, their coexistence, as a manifestation of entangled momentum-space and real-space topology, in a single sample has not been reported. Here, by inserting a TI layer between two magnetic TI layers to form a sandwich heterostructure, we observed the crossover from the QAH to TH effect through tuning the chemical potential. This observation can be understood through the Dzyaloshinskii-Moriya interaction of the topological surface and bulk quantum well states.
The unification of topological quantum phenomena in momentum-space and real-space in a single device provides a unique platform for dissipationless spin-textured spintronic applications.

Electronic band structures of nontrivial topology in momentum-space and magnetic chiral spin textures in real-space have attracted enormous attention in the past decade since they harbor elegant Berry curvature physics. The intrinsic anomalous Hall (AH) effect is such an example: it is induced by the Berry curvature in momentum-space in ferromagnetic (FM) materials and can even be quantized under certain circumstances, leading to the quantum anomalous Hall (QAH) effect. The QAH effect has been theoretically proposed and experimentally realized in magnetically doped topological insulator (TI) films. On the other hand, chiral magnetic textures (e.g. skyrmions or chiral domain walls) provide another example of non-trivial topology, but in real-space. It has been shown that chiral spin textures can also induce a Hall current: this is known as the topological Hall (TH) effect and is generally regarded as the transport signature of non-zero spin chirality. The TH effect has been experimentally observed in many magnetic systems, such as MnSi, MnGe, FeGe, and SrIrO3/SrRuO3 interface as well as magnetically doped TI films and heterostructures. Although both the QAH and TH effect have been separately observed in magnetically doped TI materials, the sample conditions for realizing them are quite different and to our knowledge, there is no existing report that reveals both phenomena in a single sample. Therefore, it is an interesting question to ask if both effects can coexist in a single sample by fine-tuning the carrier density through electric gating. The observation of both phenomena in a single sample will provide a new platform to understand the interplay of momentum-space and real-space Berry curvatures in magneto-transport phenomena.
To unify the QAH and TH effects in a TI-based device, the sample should accommodate the following three conditions: (i) The time-reversal (TR) symmetry is broken, which is the common prerequisite for both the QAH and TH effect; (ii) The chemical potentials of the top and bottom surfaces can be simultaneously tuned into the magnetic exchange gaps, which is essential for the QAH effect; (iii) A significant Dzyaloshinskii-Moriya (DM) interaction, \( H = D_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) \), can be created, which is required for the TH effect. Recently, two papers reported the observation of the TH effect in TI materials. Yasuda et al.\textsuperscript{20} fabricated the Cr-(Bi,Sb)\textsubscript{2}Te\textsubscript{3}/(Bi,Sb)\textsubscript{2}Te\textsubscript{3} bilayer heterostructures, where only one surface is gapped owing to the magnetic exchange interaction. The consequential spatial asymmetry thus favors the formation of the DM interaction and gives rise to the TH effect in this bilayer heterostructure when it is tuned into the \( p \)-type regime. However, the QAH effect is not possible in such a sample since the other surface is non-magnetic and gapless. Liu et al.\textsuperscript{21} grew 4 quintuple layers (QL) Mn-doped Bi\textsubscript{2}Te\textsubscript{3} films with hybridized surface states (SSs) on a SrTiO\textsubscript{3}(111) substrate, revealing the TH effect in both \( p \)- and \( n \)-type regimes. Since the Dirac point is usually buried under the bulk valence bands in Bi\textsubscript{2}Te\textsubscript{3} films\textsuperscript{22,23}, the QAH state is also unlikely in these samples. In uniformly doped QAH samples, the inversion symmetry is respected in the bulk\textsuperscript{9,10}, so that the DM interaction is induced only on the surface\textsuperscript{24,25}. However, the DM interactions from the two opposite SSs have opposite signs. Once two surfaces are strongly coupled, no chiral structure can be formed. Effectively, the overall DM interaction felt by the magnetization is greatly reduced and it is difficult to realize the TH effect in such uniformly doped QAH samples. Therefore, to realize the crossover from the QAH to TH effects in one system, the two surfaces of a QAH sample should be separated and inversion symmetry must be broken by an external knob (e.g. gating), in order
to engender finite DM interactions. In this Article, we report the realization of all the necessary conditions needed for observing both the QAH and TH effect in a single sample.

We fabricated a TI-based sandwich structure with an undoped TI layer (5 QL (Bi, Sb)₂Te₃ layers) inserted between two magnetic TI layers (two 3QL Cr-doped (Bi, Sb)₂Te₃ layers with the same Cr concentration) (Figs. 1a and 1b). Such a 3-5-3 sandwich heterostructure has the following advantages: (i) The nonmagnetic TI layer can serve as a spacer to separate the magnetic exchange interaction between the two magnetic TI layers. As a result, the influence of the DM interaction can be maximized since the magnetic moments in each magnetic TI layer interacts only with their own SS. (ii) Both the top and bottom SSs are separately gapped by the magnetization in the two magnetic TI layers, thus making the QAH effect possible. We also grew a variety of other sample configurations as well as a second 3-5-3 sample to substantiate the arguments that will be presented in this Article (see Supplementary Information).

The magnetic/nonmagnetic/magnetic TI sandwich heterostructures were grown on 0.5mm thick heat-treated SrTiO₃ (111) substrate in a molecular beam epitaxy (MBE) chamber with a base vacuum ~ 2×10⁻¹⁰ mbar. The Bi/Sb ratio in each layer is optimized to tune the chemical potentials near the charge neutral point. The transport studies were carried out in a dilution refrigerator (Leiden Cryogenics, 10 mK, 9 T) and a Physical Property Measurement System (Quantum Design, 2 K, 9 T) with the magnetic field applied perpendicular to the film plane. Six-terminal Hall bars with bottom-gate electrodes (Fig. 1a) were used for the transport studies.

We now focus on measurements of the 3-5-3 heterostructure. When the bottom gate V₉ = 0V, the FM order at low-temperatures gaps out the top and bottom SSs, and the chemical potential is located inside the magnetic exchange gaps of both surfaces (Fig. 1b). This is evidenced by the observation of the perfect quantized Hall resistance (ρₓᵧ) of h/e² and the vanishing longitudinal
resistance ($\rho_{xx}$) at $T = 30$ mK (Figs. 1c and 1d). With increasing temperature, the sample deviates from the QAH state and shows transport properties of a conventional FM material: hysteretic $\rho_{yx}$ loops and butterfly-shaped $\rho_{xx}$. The Curie temperature ($T_C$) of the 3-5-3 sandwich heterostructure sample is determined to be ~ 19 K using Arrott plots (see Supplementary Information).

**Figure 2** shows the Hall traces of the TI sandwich heterostructure at different bottom gate voltages $V_g$. When $V_g = V_g^0 = +20$ V, the sample displays a perfect QAH state: at zero magnetic field, $\rho_{xx}(0) = \pm h/e^2$ and $\rho_{yx}(0) < 1$ $\Omega$. The Hall curves under upward and downward magnetic field ($\mu_0 H$) sweeps completely overlap when $\mu_0 H > \mu_0 H_c$ ($\mu_0 H_c$ is the coercive field) (Fig. 2d). When $V_g$ is tuned to -80 V, a number of hole carriers are injected into the sample and dissipative channels are introduced. $\rho_{xx}(0)$ thus deviates from $h/e^2$. Intriguingly, a “hump” feature within a range of a fraction of a Tesla above $\mu_0 H_c$ appears in $\rho_{yx}$ curves (green shadow area), i.e., the Hall curve under downward $\mu_0 H$ sweep does not overlap with that under upward $\mu_0 H$ sweep when $\mu_0 H > \mu_0 H_c$ (Fig. 2c). The “hump” feature observed here is usually interpreted as a signature of the TH effect and considered as strong evidence for the existence of chiral magnetic textures in the real-space $^3, 14, 15, 16, 17, 18, 19, 20, 21$. Therefore, by utilizing the bottom electrostatic gating effect on such magnetic/nonmagnetic/magnetic TI sandwich heterostructures, our experiment demonstrated a clear crossover between the QAH and TH effect. This “hump” feature of TH effect becomes more pronounced as the chemical potential is approaching the bulk valence bands (Figs. 2a and 2b).

When $V_g > V_g^0$, the electron carriers are injected into the sandwich sample, allowing the chemical potential to approach the bulk conduction band. A trace of the “hump” feature also appears (Fig. 2e), and then fades away when more electrons are introduced (Fig. 2f). The
asymmetric behavior of the TH effect in $n$- and $p$-type regions is possibly a result of the non-symmetric electronic band structure of TI materials $^{23, 29}$, as discussed below. We note that the slope of the Hall traces at high magnetic fields ($0.5 \, \text{T} < \mu_0 H < 1.5 \, \text{T}$) is always negative in both $V_g < V_g^0$ and $V_g > V_g^0$ regions, which suggests that the standard Hall coefficient $R_N$ cannot be used to estimate carrier density near the QAH regime (see Fig. S8 and the relevant discussion in Supplementary Information).

In a FM material, the total Hall resistance $\rho_{yx}$ is a result of three contributions: the normal Hall (NH) resistance $\rho_{yx}^{\text{NH}}$, the AH resistance $\rho_{yx}^{\text{AH}}$, and the TH resistance $\rho_{yx}^{\text{TH}}$,

$$\rho_{yx} = \rho_{yx}^{\text{NH}} + \rho_{yx}^{\text{AH}} + \rho_{yx}^{\text{TH}}$$

In order to single out the TH component $\rho_{yx}^{\text{TH}}$, we need to subtract $\rho_{yx}^{\text{NH}}$ and $\rho_{yx}^{\text{AH}}$ from $\rho_{yx}$. Here we interpret the offset resistance under upward and downward $\mu_0 H$ sweeps (green shadow area in Fig. 2) as the $\rho_{yx}^{\text{TH}}$ for the following reasons. Let us consider the positive $\mu_0 H$ regime: During the downward $\mu_0 H$ sweep (red curves in Fig. 2), the system should be in a FM state without any spin texture and thus the $\rho_{yx}$ should include $\rho_{yx}^{\text{NH}}$ and $\rho_{yx}^{\text{AH}}$. For the upward $\mu_0 H$ sweep (blue curves in Fig. 2), the system undergoes a magnetic transition around the $\mu_0 H_c$ and chiral spin textures, particularly chiral magnetic domain walls (see the relevant discussion on Figs. 4a and 4b below), can be formed. In this situation, all three Hall contributions can exist and $\rho_{yx}^{\text{NH}} + \rho_{yx}^{\text{AH}}$ keeps the same value during the downward $\mu_0 H$ sweep. Thus, the $\rho_{yx}^{\text{TH}}$ can be extracted by the difference between the red and blue curves. We note that the standard expressions for the $\rho_{yx}^{\text{NH}}$ and $\rho_{yx}^{\text{AH}}$ ($\rho_{yx}^{\text{NH}} = \mu_0 R_N H$ and $\rho_{yx}^{\text{AH}} = R_A M$ with the AH coefficient $R_A$ and the sample magnetization $M$ $^{9, 30, 31}$) are only applicable to metallic systems and thus invalid in our system close to the QAH insulating regime. When $V_g = -200 \, \text{V}$, the maximum of $\rho_{yx}^{\text{TH}}$ is $\sim 1.65 \, \text{k}\Omega$, 
which is much larger than the TH resistances observed in all previous studies on metallic systems\textsuperscript{3, 14, 15, 16, 17, 18, 19, 20, 21}. A larger $\rho_{yx}^{TH}$ indicates the higher density or the smaller size of the chiral magnetic textures in our sandwich heterostructures\textsuperscript{4, 19, 21}.

To further study the crossover of the QAH to TH effect, we summarize the $V_g$-dependence of $\rho_{yx}^{TH}$ at $T = 30$ mK in Fig. 3a. When $-10 < V_g < +60$ V, since $\rho_{yx}$ is fully quantized, $\rho_{yx}^{TH}$ is constantly 0. When $V_g$ is tuned from $-10$ V to $-200$ V, $\rho_{yx}^{TH}$ increases monotonically. When $V_g$ is increased from $+60$ V to $+200$ V, the $\rho_{yx}^{TH}$ still remains finite but has much smaller values. The value of $\rho_{yx}^{TH}$ shows a peak at $V_g = +90$ V and goes down with further increases in $V_g$. We attribute the much smaller $\rho_{yx}^{TH}$ when $V_g > V_g^0$ to the non-participation of the bulk conduction bands; consequently, the TH feature at $V_g > V_g^0$ is solely induced by the chemical potential difference between the top and bottom surfaces crossing the helical SSs\textsuperscript{32, 33}, as discussed below.

Figure 3b shows the $\mu_0 H$ dependence of $\rho_{yx}^{TH}$ with the maximum peak at different temperatures under the $V_g = V_g^{TH, max}$. With $T$ increasing from $30$ mK to $5$ K, the $\rho_{yx}^{TH}$ decreases gradually and the $\rho_{yx}^{TH}$ feature completely disappears at $T = 5$ K. The peak position of $\rho_{yx}^{TH}$ and the magnetic field range of the “hump” feature also monotonically decreases with increasing $T$. We summarize the $\rho_{yx}^{TH}$ as a function of ($V_g - V_g^0$) at different temperatures in Fig. 3c. The $\rho_{yx}^{TH}$ curve at each $T$ is asymmetric between $V_g < V_g^0$ and $V_g > V_g^0$. When $60$ mK $\leq T \leq 1$ K, the $\rho_{yx}^{TH}$ shows a peak as denoted by the arrows in Fig. 3c when $V_g < V_g^0$. This observation indicates the DM interaction strength is maximized when the chemical potential crosses the bulk valence bands.

In order to understand the experimental observations, we propose a physical picture based on the emergence of chiral spin textures around the $\mu_0 H_c$ regime. The observed “hump” structure in the $\rho_{yx}$ has been observed in a variety of noncollinear magnetic systems, particularly magnetic skyrmion systems\textsuperscript{14, 15, 16, 17}, and is regarded as the key signature for the chirality of skyrmions.
However, our sample has a robust FM ground state for the occurrence of the QAH effect at low $\mu_0H$, thus stable skyrmions are unlikely to be formed 34. Our theoretical calculation (see Supplementary Information) indeed confirms that the magnetic TI sample is dominated by FM states (i.e. Heisenberg magnetic coupling). Since the TH effect only occurs near the $\mu_0H_c$ regime, this fact motivates us to consider the possible spin textures during magnetization reversal. During a FM transition, magnetic domains with opposite polarization are nucleated and chiral walls can be formed at the domain boundaries, due to the presence of the strong DM interaction (Figs. 4a and 4b). Net scalar chirality $\chi = \sum S_1 \cdot (S_2 \times S_3)$ is thus non-zero and leads to the TH effect. This is similar to the emergent chirality reported in Refs. 35, 36, 37, where magnetization reversal is driven by thermal fluctuations. Chirality is caused by canting of neighboring spins, so that the DM interaction is essential. Furthermore, as the scalar chirality $\chi$ respects full rotational symmetry while the DM interaction strength $D$ breaks inversion symmetry, $\chi \sim D^2$. The TH effect observed above can thus be understood qualitatively by investigating the DM interaction in magnetic/nonmagnetic/magnetic TI sandwich systems.

The DM interaction here is attributed to the strong spin-orbit coupling in our system. It can be computed via the spin susceptibility of a simplified model for TI films 38. The Hamiltonian consists of two parts: the SS Hamiltonian $H_{ss}$ and the bulk state Hamiltonian $H_{QW}$ that describes the quantum well (QW) states due to the quantum confinement effect, details of which are shown in the Methods. The energy dispersions of the SS and QW bands are shown in Fig. 4c. For simplicity, only one set of QW bands are considered, but the inclusion of more QW bands is straightforward and does not affect our qualitative interpretation. The Dirac cones of SS bands are close to the valence band top of QW states, and this is consistent with the early theoretical and experimental studies 32, 33, 39. Electrons couple to the magnetization $M$ via $H_{Zeeman} = -M \cdot$
\[ \Gamma, \text{ where } \Gamma \text{ are proper } 4 \times 4 \text{ matrices for electron spin operators. The spin susceptibility } \chi_{\alpha\beta} (\alpha, \beta = x, y, z) \text{ is evaluated for the model Hamiltonian based on linear response theory} \]
\[ \chi_{\alpha\beta}(q) = \frac{T}{2v} \text{Tr}[G_0(q + k, \omega_m) \Gamma_{\alpha} G_0(k, i\omega_m) \Gamma_{\beta}]. \]

The DM interaction is given by \( D_\alpha(q) = \epsilon_{\alpha\beta\gamma} \chi_{\beta\gamma}(q) \) where \( \epsilon \) is the Levi-Civita symbol. As the system breaks mirror symmetry with respect to xy-plane, \( \chi_{xy} = 0 \), we thus focus on the off-diagonal components \( \chi_{xz} \) and \( \chi_{yz} \). As expected from the Moriya rule \(^{40}\), \( \chi_{xz} (\chi_{yz}) \) is linearly proportional to the momentum \( q_x (q_y) \) (see Supplementary Information). Fixing a nonzero \( q_x \) value, Figures 4d and 4e show \( \chi_{xz} \) as a function of energy for the QW states and SSs with different asymmetric potentials \( U \). A nonzero \( U \) is indeed required to break the inversion symmetry and induce finite \( \chi_{xz} \) and \( \chi_{yz} \), as well as the related DM interactions. The SSs contribution to \( \chi_{xz} \) shows a double peak structure around the charge neutral point (i.e. \( E_F = 0 \)). For the QW states, \( \chi_{xz} \) reveals a peak when the Fermi energy (\( E_F \)) lies between two spin-split (valence) bands and then drops and even changes its sign when the \( E_F \) crosses both spin bands. The bulk conduction band is well above the energy range of interest (-30~40 meV) and thus has no contribution. Figure 4f shows the total \( \chi_{xz} \), which behaves similarly as the \( \rho_{yx}^{\text{TH}} \) in our experiment shown in Fig. 3a. We first notice a large asymmetry between the electron and hole doping sides. This is because the SS is close to the bulk valence band but well below the bulk conduction band. At \( V_g < V_g^0 \), a large contribution to \( \chi_{xz} \) from the bulk valence band top of QW states significantly enhances the DM interaction and net spin chirality or \( \rho_{yx}^{\text{TH}} \) in consequence. We note that including more QW states in the model can further enhance the DM interaction in the hole doping regime. On the other hand, the SS contribution prevails in the \( V_g > V_g^0 \) regime. When the \( E_F \) is high above the charge neutral point, \( \chi_{xz} \) vanishes, which is consistent with the \( \rho_{yx}^{\text{TH}} \) in the electron doping regime. Note that \( \chi_{xz} \) is non-zero in the charge neutral regime, but experimental \( \rho_{yx}^{\text{TH}} \) does vanish, particularly at low
temperatures. This is because there are no bulk carriers and the current flows only along chiral edge state in the QAH regime. Therefore, even though spin chirality is present, no charge transport can be supported. This scenario is demonstrated through a numerical simulation of the TH effect for a single chiral domain wall (see Supplementary Information). At higher temperatures (0.4 K ~ 3 K), bulk carriers can be thermally excited and the $\rho_{yx}^{TH}$ gradually decreases in Fig. 3c. At an even higher temperature (>5 K), the vanishing of $\rho_{yx}^{TH}$ is the result of the fact that the thermal fluctuation is much larger than the energy scale of DM interaction and thus destroy the chirality of magnetic domain walls.

There is a recent experiment suggesting an alternative interpretation of the “hump” feature in $\rho_{yx}$ of the SrIrO$_3$/SrRuO$_3$/SrTiO$_3$ sandwich structures $^{41}$, in which the SrIrO$_3$/SrRuO$_3$ and SrRuO$_3$/SrTiO$_3$ interfaces have the opposite AH signs and different $\mu_0H_c$. In their sandwich structure sample, the antiferromagnetic alignment configuration between the two magnetic interfaces will induce a larger $\rho_{yx}$ than the FM alignment. This can lead to the TH effect-like hump structure. However, this scenario is unlikely to occur in our magnetic/nonmagnetic/magnetic TI sandwich heterostructure for the following reasons: (i) The $\rho_{yx}$ with a negative sign together with a significantly enhanced $\mu_0H_c$ has never been observed in Cr-doped (Bi,Sb)$_2$Te$_3$ samples. (ii) As a control experiment, we grew 5QL V-doped TI (Bi,Sb)$_2$Te$_3$ on top of the 3-5-3 sandwich sample and used the exchange coupling to increase the $\mu_0H_c$ of the top Cr-doped TI layer. This structure configuration favors the formation of the antiferromagnetic alignment between the top and bottom Cr-doped TI layers. The “hump” feature, however, disappears rather than being enhanced (see Supplementary Information).

To summarize, we fabricated magnetic/nonmagnetic/magnetic TI sandwich heterostructures and observed the crossover between the QAH to TH effect by applying an
electrostatic gating voltage. The observation of the crossover from the QAH effect to the TH effect in a single sample reveals the transformation of Berry curvature from the momentum-space to the real-space. Our theoretical calculations demonstrate that the two surfaces of the sandwich heterostructure with asymmetric potentials can form a finite DM interaction, which corresponds to the emergence of the TH effect. The chiral magnetic domain walls relevant to the TH effect can be utilized to record the spin information \(^{34}\), while the dissipation-free chiral edge states in the QAH effect can be used to transfer this information with low-energy cost. The marriage of the TH and QAH effect opens the door for further explorations of magnetic TI-based multilayer heterostructures for proof-of-concept next generation energy-efficient spintronic and electronic applications.

**Methods**

**MBE growth of TI sandwich heterostructure.**

The magnetic/nonmagnetic/magnetic TI sandwich heterostructure growth was carried out using a Veeco/Applied EPI MBE system with a vacuum \(\sim 2\times10^{-10}\) mbar. The heat-treated insulating SrTiO\(_3\) (111) substrates were outgassed at \(\sim 530^\circ C\) for 1 hour before the growth of the TI sandwich heterostructures. High-purity Bi (5N), Sb (6N), Cr (5N) and Te (6N) were evaporated from Knudsen effusion cells. During growth of the TI, the substrate was maintained at \(\sim 240^\circ C\). The flux ratio of Te per (Bi + Sb) was set to be \(>10\) to prevent Te deficiency in the samples. The magnetic or nonmagnetic TI growth rate was at \(\sim 0.25\) QL/min. Each layer of the sandwich heterostructure was grown with the different Bi/Sb ratio by adjusting their K-cell temperatures to tune the chemical potential close to its charge neutral point. Following the growth, the TI films were annealed at \(\sim 240^\circ C\) for 30 minutes to improve the crystal quality before being cooled down to room temperature. Finally, to avoid possible contamination, a 10
nm thick Te layer is deposited at room temperature on top of the sandwich heterostructures prior to their removal from the MBE chamber for *ex-situ* transport and other characterization measurements.

**Hall-bar device fabrications.**

The TI sandwich heterostructures grown on 2 mm × 10 mm heated-treated insulating SrTiO$_3$ (111) were scratched into a Hall bar geometry using a computer-controlled probe station. The effective area of the Hall bar device is ~ 1 mm × 0.5 mm. The electrical Ohmic-contacts for transport measurements were made by pressing indium spheres on the Hall bar. The bottom gate electrode was prepared through an indium foil on the back side of the SrTiO$_3$ substrate.

**Transport measurements.**

Transport measurements were conducted using both a Quantum Design Physical Property Measurement System (PPMS; 2 K, 9 T) and a Leiden Cryogenics dilution refrigerator (10 mK, 9 T) with the magnetic field applied perpendicular to the film plane. The bottom gate voltage was applied using a Keithley 6430. The excitation currents in the DC PPMS measurements (≥ 2 K) is 1 µA. We used a PicoWatt AVS-47 AC resistance bridge to conduct the dilution refrigerator measurements (< 2 K) with a low excitation current (1 nA) to suppress heating effects. The results reported here have been reproduced on 2 samples measured in the dilution refrigerator and more than 10 samples measured in PPMS. All the transport results shown here were anti-symmetrized as a function of the magnetic field. More transport results are found in the Supplementary Information.

**Theoretical calculations.**
The QW Hamiltonian is $H_{QW} = \varepsilon_0(k) + N(k)\tau_z + A(k_y\sigma_x - k_x\sigma_y)\tau_x + U\tau_x$, where Pauli matrices $\sigma$’s stand for spins and $\tau$’s stand for two orbitals. $\varepsilon = C_0 + C_1k^2$ and $N = N_0 + N_2k^2$. Different sets of QW states differ by different $C_0$ and $N_0$ values. Dispersion in Fig. 4c takes values $C_0 = 0.145\text{eV}$, $C_2 = 10.0\text{eV} \cdot \AA^2$, $N_0 = -0.18\text{eV}$, $N_2 = 15.0\text{eV} \cdot \AA^2$, $A = 3.0\text{eV} \cdot \AA$, and $U = 0.02\text{eV}$. Coupling of the QW electrons to magnetization $M$ is simply $H_{Zeeman}^{QW} = -M \cdot \sigma$.

On the other hand, the SSs have the Hamiltonian $H_{SS} = v_F(k_y\sigma_x - k_x\sigma_y)\tau_z + U\tau_z + m_0\sigma_x$, where $\sigma$’s again stand for spins but $\tau$’s stand for two surfaces instead. $U$ is the asymmetric potential applied to two surfaces. Its coupling to magnetizations $M^t$ and $M^b$ on top and bottom surfaces, respectively, is $H_{Zeeman}^{SS} = M^t \cdot \sigma(1 + \tau_z)/2 + M^b \cdot \sigma(1 - \tau_z)/2$. In Fig. 4c, we use $m_0 = 0.005\text{eV}$, $v_F = 3.0\text{eV} \cdot \AA$, and $U = 0.02\text{eV}$.

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**Author contributions**
N. S., M. H. W. C., and C. Z. C. conceived and designed the experiment. D. X. grew the sandwich heterostructure samples with the help of N. S. and C. Z. C. F. W., K. W., and J. J. performed characterizations of the samples with the help of C. Z. C. J. J., F. W., J. H. S., R. X., and M. K. performed the dilution refrigerator measurements with the help of M. H. W. C. and C. Z. C. J. J., F. W., Y. Z. and L. Z. carried out the PPMS transport measurement with the help M. H. W. C. and C. Z. C. D. A., J. X. Z., J. D. Z., and C. L. provided theoretical support and did all theoretical calculations. J. J., J. D. Z., C. L., N. S., M. H. W. C., and C. Z. C. analyzed the data and wrote the manuscript with contributions from all authors.

Additional information

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Competing financial interests

The authors declare no competing financial interests.
Figures and figure captions

**Figure 1** | **QAH effect in TI sandwich heterostructures at** \( V_g = 0 \, \text{V} \). (a) Schematic of the field effect transistor device with a bottom gate \((V_g)\) used in transport measurements. The electrical contacts on the Hall bar and the back-gate contact are made by pressed indium dots. The 0.5 mm SrTiO\(_3\) (111) substrate is used as the dielectric layer for the bottom gate. (b) Schematic of the magnetic/nonmagnetic/magnetic TI sandwich heterostructure. The total thickness of the sample \((3\text{QL (Bi,Sb)}_{1.85}\text{Cr}_{0.15}\text{Te}_3/5\text{QL (Bi,Sb)}_2\text{Te}_3/3\text{QL (Bi,Sb)}_{1.85}\text{Cr}_{0.15}\text{Te}_3)\) is 11QL. When \( T < T_C \), an exchange gap opens at the Dirac points of top and bottom surfaces. (c, d) Magnetic field \( \mu_0H \) dependence of the longitudinal resistance \( \rho_{xx} \) (c) and the Hall resistance \( \rho_{yx} \) (d) at \( V_g = 0 \, \text{V} \). At \( T = 30 \, \text{mK} \) and \( V_g = 0 \, \text{V} \), the quantized \( \rho_{yx} \) and the vanished \( \rho_{xx} \) suggest this sandwich sample is in the QAH state.
Figure 2 | Crossover of QAH to TH effect at $T = 30$ mK. (a-f) Magnetic field $\mu_0H$ dependence of the Hall resistance $\rho_{yx}$ at different $V_g$s. The sample harbors a perfect QAH state when $V_g = V_g^0 = +20$ V. When $V_g$ is tuned away from $V_g^0$, $\rho_{yx}$ deviates from the quantized value (i.e., $h/e^2$) and a “hump” feature shaded in green which is known as the TH effect appears. Insets of (a-f) show the TH resistance $\rho_{yx}^{\text{TH}}$, which is subtracted using the following method: the offset resistance of $\rho_{yx}$ when the external $\mu_0H$ is swept upward and downward. Blue (red) curve represents the process for increasing (decreasing) $\mu_0H$. 
Figure 3 | TH component $\rho_{yx}^{\text{TH}}$ at different $V_g$s and $T$s. (a) $V_g$ dependence of $\rho_{yx}^{\text{TH}}$ at $T = 30$ mK. $\rho_{yx}^{\text{TH}}$ shows asymmetric behavior between $V_g < V_g^0$ and $V_g > V_g^0$. $\rho_{yx}^{\text{TH}}$ for $V_g < V_g^0$ is much larger than $\rho_{yx}^{\text{TH}}$ for $V_g > V_g^0$. (b) $\mu_0H$ dependence of $\rho_{yx}^{\text{TH}}$ for different $T$ at $V_g = V_g^{\text{TH}, \text{max}}$. $\rho_{yx}^{\text{TH}}$ decreases with increasing temperature. $\rho_{yx}^{\text{TH}}$ is 1.65 KΩ at $T = 30$ mK and disappears at $T = 5$K. (c) The maximum $\rho_{yx}^{\text{TH}}$ as a function of $(V_g - V_g^0)$ at different $T$. $\rho_{yx}^{\text{TH}}$ shows a peak denoted by the arrows for $V_g < V_g^0$. 
Figure 4 | Chiral magnetic domain walls and theoretical interpretations of the crossover between the QAH to TH effects. (a) The formation of the chiral magnetic domain walls during magnetization reversals. (b) A magnified view depicting the spin distribution of the chiral magnetic domain wall. (c) The energy dispersions of the SS and QW bands in the magnetic TI sandwich heterostructures. (d-e) $\chi_{xz}$ as a function of energy for the QW and SS states, respectively, under different asymmetric potentials $U$. (f) The QW contribution to $\chi_{xz}$, SS contribution to $\chi_{xz}$, and total $\chi_{xz}$ in the magnetic TI sandwich heterostructures when $U=0.02$. $q_x = 0.005\AA^{-1}$ and $q_y = 0$ in figures (d-f).
References

1. Hasan M. Z., Kane C. L. Colloquium: Topological Insulators. *Rev. Mod. Phys.* **82**, 3045-3067 (2010).

2. Qi X. L., Zhang S. C. Topological Insulators and Superconductors. *Rev. Mod. Phys.* **83**, 1057-1110 (2011).

3. Nagaosa N., Tokura Y. Topological Properties and Dynamics of Magnetic Skyrmions. *Nat. Nanotechnol.* **8**, 899-911 (2013).

4. Nagaosa N., Sinova J., Onoda S., MacDonald A. H., Ong N. P. Anomalous Hall effect. *Rev. Mod. Phys.* **82**, 1539-1592 (2010).

5. Haldane F. D. M. Model for a Quantum Hall-Effect without Landau Levels: Condensed-Matter Realization of the "Parity Anomaly". *Phys. Rev. Lett.* **61**, 2015-2018 (1988).

6. Liu C. X., Qi X. L., Dai X., Fang Z., Zhang S. C. Quantum anomalous Hall effect in Hg_{1-y}Mn_yTe quantum wells. *Phys. Rev. Lett.* **101**, 146802 (2008).

7. Qi X. L., Hughes T. L., Zhang S. C. Topological Field Theory of Time-Reversal Invariant Insulators. *Phys. Rev. B* **78**, 195424 (2008).

8. Yu R., Zhang W., Zhang H. J., Zhang S. C., Dai X., Fang Z. Quantized Anomalous Hall Effect in Magnetic Topological Insulators. *Science* **329**, 61-64 (2010).

9. 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* **340**, 167-170 (2013).

10. 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.* **14**, 473-477 (2015).

11. Checkelsky J. G., Yoshimi R., Tsukazaki A., Takahashi K. S., Kozuka Y., Falson J., Kawasaki M., Tokura Y. Trajectory of the Anomalous Hall Effect towards the Quantized State in a Ferromagnetic Topological Insulator. *Nat. Phys.* **10**, 731-736 (2014).

12. Kou X. F., Guo S. T., Fan Y. B., Pan L., Lang M. R., Jiang Y., Shao Q. M., Nie T. X., Murata K., Tang J. S., Wang Y., He L., Lee T. K., Lee W. L., Wang K. L. Scale-Invariant Quantum Anomalous Hall Effect in Magnetic Topological Insulators beyond the Two-Dimensional Limit. *Phys. Rev. Lett.* **113**, 137201 (2014).
13. 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.* **107**, 182401 (2015).

14. Neubauer A., Pfleiderer C., Binz B., Rosch A., Ritz R., Niklowitz P. G., Boni P. Topological Hall Effect in the A Phase of MnSi. *Phys. Rev. Lett.* **102**, 186602 (2009).

15. Lee M., Kang W., Onose Y., Tokura Y., Ong N. P. Unusual Hall Effect Anomaly in MnSi under Pressure. *Phys. Rev. Lett.* **102**, 186601 (2009).

16. Kanazawa N., Onose Y., Arima T., Okuyama D., Ohoyama K., Wakimoto S., Kakurai K., Ishiwata S., Tokura Y. Large Topological Hall Effect in a Short-Period Helimagnet MnGe. *Phys. Rev. Lett.* **106**, 156603 (2011).

17. Huang S. X., Chien C. L. Extended Skyrmion Phase in Epitaxial FeGe(111) Thin Films. *Phys. Rev. Lett.* **108**, 267201 (2012).

18. Matsuno J., Ogawa N., Yasuda K., Kagawa F., Koshiba W., Nagaosa N., Tokura Y., Kawasaki M. Interface-Driven Topological Hall Effect in SrRuO$_3$-SrIrO$_3$ Bilayer. *Sci. Adv.* **2**, e1600304 (2016).

19. Ohuchi Y., Matsuno J., Ogawa N., Kozuka Y., Uchida M., Tokura Y., Kawasaki M. Electric-Field Control of Anomalous and Topological Hall Effects in Oxide Bilayer Thin Films. *Nat. Commun.* **9**, 213 (2018).

20. Yasuda K., Wakatsuki R., Morimoto T., Yoshimi R., Tsukazaki A., Takahashi K. S., Ezawa M., Kawasaki M., Nagaosa N., Tokura Y. Geometric Hall Effects in Topological Insulator Heterostructures. *Nat. Phys.* **12**, 555-559 (2016).

21. Liu C., Zang Y. Y., Ruan W., Gong Y., He K., Ma X. C., Xue Q. K., Wang Y. Y. Dimensional Crossover-Induced Topological Hall Effect in a Magnetic Topological Insulator. *Phys. Rev. Lett.* **119**, 176809 (2017).

22. Chen Y. L., Analytis J. G., Chu J. H., Liu Z. K., Mo S. K., Qi X. L., Zhang H. J., Lu D. H., Dai X., Fang Z., Zhang S. C., Fisher I. R., Hussain Z., Shen Z. X. Experimental Realization of a Three-Dimensional Topological Insulator, Bi$_2$Te$_3$. *Science* **325**, 178-181 (2009).

23. Zhang J. S., Chang C. Z., Zhang Z. C., Wen J., Feng X., Li K., Liu M. H., He K., Wang L. L., Chen X., Xue Q. K., Ma X. C., Wang Y. Y. Band Structure Engineering in (Bi$_{1-x}$Sb$_x$)$_2$Te$_3$ Ternary Topological Insulators. *Nat. Commun.* **2**, 574 (2011).

24. Zhu J. J., Yao D. X., Zhang S. C., Chang K. Electrically Controllable Surface Magnetism on the Surface of Topological Insulators. *Phys. Rev. Lett.* **106**, 097201 (2011).
25. Ye F., Ding G. H., Zhai H., Su Z. B. Spin Helix of Magnetic Impurities in Two-Dimensional Helical Metal. *Euro. Phys. Lett.* **90**, 47001 (2010).

26. 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.* **120**, 056801 (2018).

27. Mogi M., Kawamura M., Yoshimi R., Tsukazaki A., Kozuka Y., Shirakawa N., Takahashi K. S., Kawasaki M., Tokura Y. A Magnetic Heterostructure of Topological Insulators as a Candidate for an Axion Insulator. *Nat. Mater.* **16**, 516-521 (2017).

28. Mogi M., Kawamura M., Tsukazaki A., Yoshimi R., Takahashi K. S., Kawasaki M., Tokura Y. Tailoring Tricolor Structure of Magnetic Topological Insulator for Robust Axion Insulator. *Sci. Adv.* **3**, eaao1669 (2017).

29. Zhang Y., He K., Chang C. Z., Song C. L., Wang L. L., Chen X., Jia J. F., Fang Z., Dai X., Shan W. Y., Shen S. Q., Niu Q., Qi X. L., Zhang S. C., Ma X. C., Xue Q. K. Crossover of the Three-Dimensional Topological Insulator Bi$_2$Se$_3$ to the Two-Dimensional Limit. *Nat. Phys.* **6**, 584-588 (2010).

30. 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.* **25**, 1065-1070 (2013).

31. Zhang Z. C., Feng X., Guo M. H., Li K., Zhang J. S., Ou Y. B., Feng Y., Wang L. L., Chen X., He K., Ma X. C., Xue Q. K., Wang Y. Y. Electrically Tuned Magnetic Order and Magnetoresistance in a Topological Insulator. *Nat. Commun.* **5**, 4915 (2014).

32. Chang C. Z., Zhao W. W., Kim D. Y., Wei P., Jain J. K., Liu C. X., Chan M. H. W., Moodera J. S. Zero-Field Dissipationless Chiral Edge Transport and the Nature of Dissipation in the Quantum Anomalous Hall State. *Phys. Rev. Lett.* **115**, 057206 (2015).

33. Wang W. B., Ou Y. B., Liu C., Wang Y. Y., He K., Xue Q. K., Wu W. D. Direct Evidence of Ferromagnetism in a Quantum Anomalous Hall System. *Nat. Phys.* **14**, 791-795 (2018).

34. Jiang W. J., Chen G., Liu K., Zang J. D., te Velthuis S. G. E., Hoffmann A. Skyrmions in Magnetic Multilayers. *Phys. Rep.* **704**, 1-49 (2017).

35. Hou W. T., Yu J. X., Daly M., Zang J. D. Thermally Driven Topology in Chiral Magnets. *Phys. Rev. B* **96** (2017).

36. Bottcher M., Heinze S., Egorov S., Sinova J., Dupe B. B-T Phase Diagram of Pd/Fe/Ir(111) Computed with Parallel Tempering Monte Carlo. *New J. Phys.* **20**, 103014 (2018).
37. Wang W., Daniels M. W., Liao Z., Zhao Y., Wang J., Koster G., Rijnders G., Chang C.-Z., Xiao D., Wu W. Universal Spin Chirality Fluctuation in Two-Dimensional Ising Ferromagnets. *arXiv:1812.07005* (2018).

38. Liu C. X., Zhang H., Yan B. H., Qi X. L., Frauenheim T., Dai X., Fang Z., Zhang S. C. Oscillatory Crossover from Two-Dimensional to Three-Dimensional Topological Insulators. *Phys. Rev. B* **81**, 041307 (2010).

39. Li W., Claassen M., Chang C. Z., Moritz B., Jia T., Zhang C., Rebec S., Lee J. J., Hashimoto M., Lu D. H., Moore R. G., Moodera J. S., Devereaux T. P., Shen Z. X. Origin of the Low Critical Observing Temperature of the Quantum Anomalous Hall Effect in V-Doped (Bi, Sb)$_2$Te$_3$ Film. *Sci. Rep.* **6**, 32732 (2016).

40. Moriya T. Anisotropic Superexchange Interaction and Weak Ferromagnetism. *Phys. Rev.* **120**, 91-98 (1960).

41. Groenendijk D. J., Autieri C., Thiel T. C. v., Brzezicki W., Gauquelin N., Barone P., van den Bos K. H. W., van Aert S., Verbeeck J., Filippetti A., Picozzi S., Cuoco M., Caviglia A. D. Berry Phase Engineering at Oxide Interfaces. *arXiv:1810.05619* (2018).