

Two-Channel Anomalous Hall Effect in Epitaxial MnBi₂Te₄/Mn-Doped Bi₂Te₃ Heterostructure

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Cite This: *Nano Lett.* 2026, 26, 10123–10131



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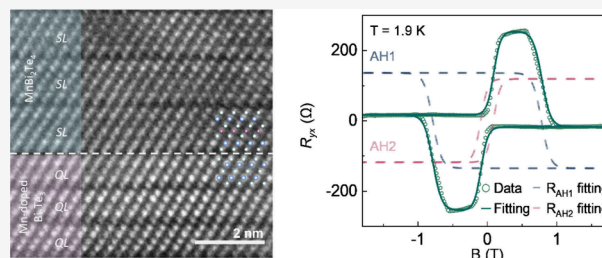
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Supporting Information

ABSTRACT: The coexistence of intrinsic magnetism and nontrivial band topology in MnBi₂Te₄ provides a fertile platform for exploring exotic quantum phenomena. Its tunable spin dynamics under external magnetic fields, combined with the van der Waals (vdW) layered nature, further enables the design of functional heterostructures with controllable magnetic and topological properties. Here, we report an observation of a two-channel anomalous Hall effect (AHE) in an epitaxial MnBi₂Te₄/Mn-doped Bi₂Te₃ vdW heterostructure. Notably, the Hall response exhibits unconventional nonsquare Hall hysteresis loops with pronounced hump-dip features, indicative of a two-channel AHE. Through temperature- and angle-dependent Hall measurements, we attribute this two-channel AHE to distinct magnetic critical behaviors of antiferromagnetic MnBi₂Te₄ and ferromagnetic Mn-doped Bi₂Te₃ layers and further reveal magnetic phase transitions under rotating magnetic fields. Our results establish a versatile platform for engineering multichannel AHE and provide new opportunities for exploring intertwined magnetic and topological phenomena in tailored vdW heterostructures.

KEYWORDS: Two-channel anomalous Hall effect, MnBi₂Te₄, Mn-doped Bi₂Te₃, vdW heterostructure, Chemical vapor deposition



MnBi₂Te₄, a van der Waals (vdW) antiferromagnet (AFM) hosting both intrinsic Néel order and nontrivial band topology, has emerged as an ideal platform for investigating novel quantum phenomena.^{1–5} In the two-dimensional limit, the A-type AFM configuration of MnBi₂Te₄ yields pronounced layer-number-dependent magnetism and topological states.^{1,6,7} Odd-layer MnBi₂Te₄ can potentially host the quantum anomalous Hall effect (QAHE), provided that the system maintains a topologically nontrivial band structure,^{1,4} whereas even-layer counterparts realize an axion insulator state.^{8–11} In addition, the rich spin-dynamics under magnetic field further enable a plethora of emergent phases, such as Weyl semimetal states^{2,12} and exotic magneto-electric responses.^{13–17} Moreover, the vdW nature and versatile magnetic structures of MnBi₂Te₄ make it an ideal building block for engineered heterostructures. For example, intercalation of nonmagnetic Bi₂Te₃ or ferromagnetic (FM) MnTe spacers allows effective tuning of the interlayer AFM superexchange coupling,^{18,19} while interfacing with superconductors can induce robust proximity superconductivity.^{20,21} Such structural engineering provides a powerful route to manipulate interlayer^{18,22,23} and interfacial coupling,^{24,25} enabling investigation of exotic properties and tunable transport behaviors.

Previous studies have revealed unusual hump-dip features in the Hall resistivity in MnBi₂Te₄ and its engineered heterostructures.^{18,26–30} Such signals are often attributed to

non-coplanar spin textures that generate real-space Berry curvature and modify the itinerant-electron dynamics near the magnetization-reversal field. Alternatively, a quantitatively equivalent interpretation arises from the superposition of multiple AHE channels, without involving chiral spin textures.^{18,27,31–34} Indeed, MnBi₂Te₄ systems may exhibit an unconventional Hall effect that deviates from the scaling relation of the AHE with magnetization in the presence of a non-collinear spin flip-flop state and field-induced canted magnetic structures.³⁵ However, extrinsic factors such as Mn-doped Bi₂Te₃ intergrowth,²⁷ Mn–Bi antisite defects,³⁶ and surface reconstruction^{26,37} can introduce additional AHE channels, complicating the interpretation. Therefore, the microscopic origin of the hump-dip featured AHE in MnBi₂Te₄-based systems still remains debated. Discriminating between these mechanisms is crucial for understanding the origin of unconventional Hall responses and realizing tunable multistate magnetotransport functionalities.

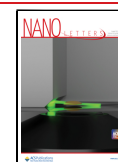
Here, we report a two-channel AHE in an epitaxial MnBi₂Te₄/Mn-doped Bi₂Te₃ vdW heterostructure, synthesized

Received: April 1, 2026

Revised: July 29, 2026

Accepted: July 29, 2026

Published: August 3, 2026



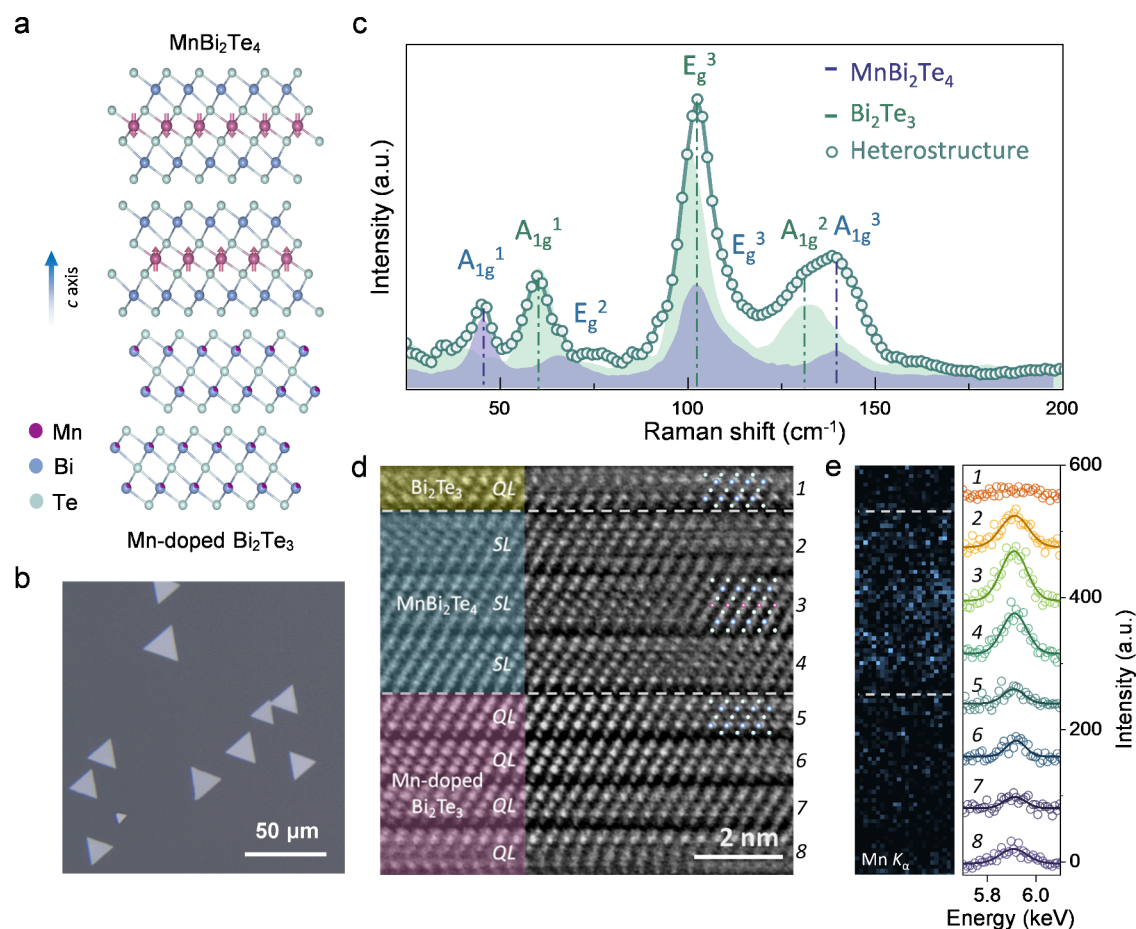


Figure 1. Structural characterizations of the $\text{MnBi}_2\text{Te}_4/\text{Mn-doped Bi}_2\text{Te}_3$ vdW heterostructure. (a) Schematic illustration of the $\text{MnBi}_2\text{Te}_4/\text{Mn-doped Bi}_2\text{Te}_3$ heterostructure. The arrows denote the spin configuration of Mn^{2+} ions in MnBi_2Te_4 . (b) A representative optical microscope image for the as-grown heterostructures on a mica substrate, showing well-defined triangular domains. (c) Raman spectra of the heterostructure, with contributions from MnBi_2Te_4 and Bi_2Te_3 highlighted by blue and green shaded regions. (d) A large-area cross-sectional HAADF-STEM image of the $\text{MnBi}_2\text{Te}_4/\text{Mn-doped Bi}_2\text{Te}_3$ heterostructure, revealing the distinct stacking of SLs and QLs. The heterostructure consists of the 1-QL topmost Bi_2Te_3 , 3-SL MnBi_2Te_4 , and 4-QL bottom $\text{Mn-doped Bi}_2\text{Te}_3$ layers. (e) Mn-K_α elemental mapping of the entire heterostructure acquired under the same STEM-EDS conditions and the corresponding Mn-K_α EDS spectra extracted from each layer according to the layer numbering defined in (d), showing that the Mn signal is predominantly localized in the MnBi_2Te_4 and the bottom $\text{Mn-doped Bi}_2\text{Te}_3$ layers. The spectra are displayed on the same intensity scale with vertical offsets for clarity.

via a two-step growth approach. Raman spectroscopy, atomic-resolution scanning transmission electron microscopy (STEM), and elemental analysis confirm the coexistence of MnBi_2Te_4 septuple layers (SLs) and $\text{Mn-doped Bi}_2\text{Te}_3$ quintuple layers (QLs). Magneto-transport measurements reveal the pronounced hump-dip Hall features that can be quantitatively decomposed into two distinct AHE channels: one originating from the antiferromagnetic MnBi_2Te_4 layers with spin-flop transitions and the other from ferromagnetic $\text{Mn-doped Bi}_2\text{Te}_3$ layers. The distinct critical behaviors observed in temperature- and angle-dependent measurements uncover complex magnetic phase transitions under rotating magnetic fields, further substantiating the two-channel scenario. Our results show that unconventional, nonsquare AHE can arise from multichannel Hall contributions without requiring chiral spin textures, offering a viable route to engineer magneto-transport in vdW heterostructures.

As MnBi_2Te_4 multilayers can be synthesized via the chemical vapor deposition (CVD) approach,³⁸ we employ a two-step growth strategy with precisely controlled deposition temperatures to realize a $\text{MnBi}_2\text{Te}_4/\text{Mn-doped Bi}_2\text{Te}_3$ vdW heterostructure (see Methods and Supplementary Figure S1).

Figure 1a schematically illustrates the vdW heterostructure. In the MnBi_2Te_4 layer, the intercalated MnTe bilayer into a Bi_2Te_3 quintuple layer (QL) forms a natural vdW tetradymite structure with a septuple-layer (SL) stacking sequence ($\text{Mn}-\text{Te}-\text{Bi}-\text{Te}-\text{Bi}-\text{Te}-\text{Mn}$) along the c -axis. The interlayer exchange coupling leads to antiferromagnetic alignment between adjacent layers, whereas intralayer coupling stabilizes ferromagnetic ordering with out-of-plane anisotropy, resulting in an A-type AFM structure (Figure 1a). In contrast, $\text{Mn-doped Bi}_2\text{Te}_3$ exhibits a perpendicular ferromagnetic order.^{39–41} Optical microscope image (Figure 1b) reveals that the as-grown heterostructures on a mica substrate form regular triangular shapes with lateral sizes of a dozen micrometers.

The composition of the heterostructure is first examined by Raman spectroscopy. As shown in Figure 1c, the spectra of MnBi_2Te_4 multilayers, Bi_2Te_3 multilayers, and the as-grown heterostructure are compared. The heterostructure exhibits characteristic Raman modes from both MnBi_2Te_4 and Bi_2Te_3 , indicating phase coexistence rather than the formation of a single superlattice phase such as MnBi_4Te_7 (see Figure S2 for comparison). Specifically, the peak at 45.5 cm^{-1} is assigned to

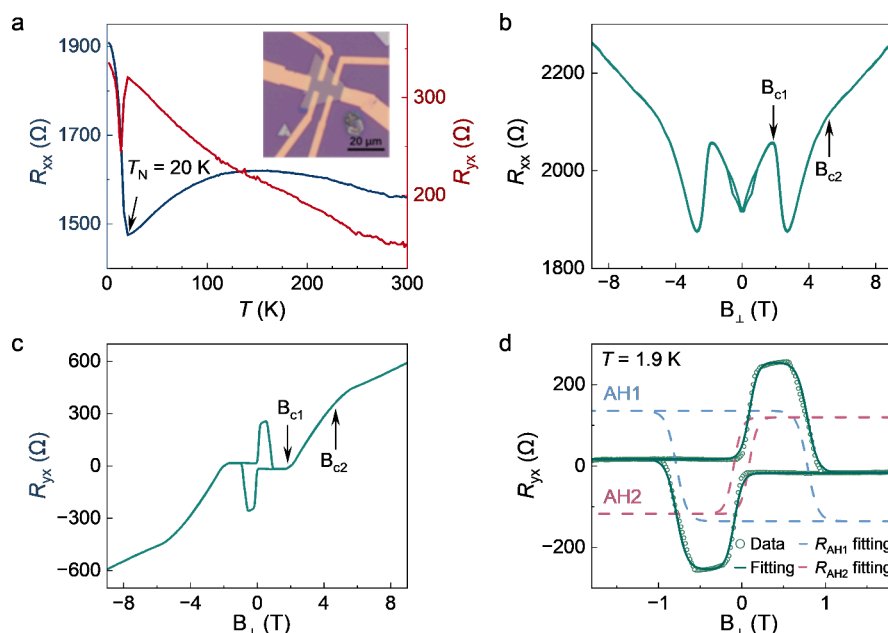


Figure 2. Observation of two-channel AHE in the $\text{MnBi}_2\text{Te}_4/\text{Mn-doped Bi}_2\text{Te}_3$ vdW heterostructure. (a) Temperature-dependent R_{xx} and R_{yx} for the heterostructure, showing an antiferromagnetic transition at 20 K. Inset shows an optical microscope image of a Hall bar device based on the heterostructure. (b and c) Magnetic-field-dependent R_{xx} and R_{yx} for the heterostructure, respectively. The data were collected at 1.9 K under an out-of-plane magnetic field. The critical fields corresponding to the canted AFM and field-driven FM transitions are labeled B_{c1} and B_{c2} with black arrows. (d) The two-channel anomalous Hall effect fitting of the R_{yx} . The two dashed lines are the curves decomposed from the original experimental data. The green solid line is the fitting result described in the main text.

the A_{1g}^1 mode of MnBi_2Te_4 , distinct from the corresponding mode of MnBi_4Te_7 at 40.8 cm^{-1} . The feature at 59 cm^{-1} originates from the A_{1g}^1 mode of Bi_2Te_3 ,^{42,43} which differs markedly from the E_g^2 mode of MnBi_2Te_4 . The overlapping E_g^3 modes of both MnBi_2Te_4 and Bi_2Te_3 locate at 102.2 cm^{-1} , dominating the Raman spectrum of the heterostructure. In addition, a broad peak at 139 cm^{-1} is observed, which is originated from the A_{1g}^2 and A_{1g}^3 mode of Bi_2Te_3 and MnBi_2Te_4 , respectively.

The vertical stacking structure is further confirmed by the aberration-corrected high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM). The cross-sectional image of the heterostructure unambiguously demonstrates the separated atomic structures of MnBi_2Te_4 SLs and Bi_2Te_3 QLs with well-defined vdW gaps (Figure 1d). As displayed in Figure 1d, the heterostructure with a thickness of 9.8 nm consists of the topmost 1-QL Bi_2Te_3 , 3-SL MnBi_2Te_4 , and bottom 4-QL Bi_2Te_3 layers, which is used to fabricate the device for transport measurements (Figure S3). We note that the STEM imaging reveals the presence of an additional 1-QL Bi_2Te_3 layer on top of the MnBi_2Te_4 layers. This layer is likely formed during the rapid cooling process, during which Bi_2O_3 continues to evaporate while the evaporation of the MnCl_2 precursor is substantially suppressed. Figure 1e displays the electron energy-dispersive X-ray spectroscopy (EDS) elemental mapping of the entire heterostructure acquired under identical experimental conditions and the corresponding layer-resolved Mn- K_α EDS spectra integrated from each layer. The Mn- K_α elemental mapping reveals that the Mn signal is predominantly localized in the MnBi_2Te_4 SLs and the lower Mn-doped Bi_2Te_3 QLs. Consistently, the Mn- K_α spectra exhibit pronounced characteristic Mn peaks in these two regions, whereas no discernible Mn characteristic peak is detected from the topmost Bi_2Te_3 layer. To further verify the

Mn distribution, electron energy loss spectroscopy (EELS) line profiles acquired from the corresponding regions are presented in Figure S4b. The EELS spectra likewise show no detectable Mn-L edge signal and only a featureless linear background for the topmost Bi_2Te_3 layer, confirming that it contains negligible Mn within the detection limit of the measurements. Combined with the Raman analysis, atomic-resolution STEM imaging, and elemental mapping, we unambiguously demonstrate that MnBi_2Te_4 and Mn-doped Bi_2Te_3 layers retain distinct structures and form a vertically stacked epitaxial heterostructure.

To investigate the transport properties, the as-grown heterostructures are transferred from the mica to SiO_2/Si substrates (see Methods), followed by fabrication of Hall bar devices using standard electron beam lithography. The inset of Figure 2a displays an optical image of a Hall bar device. The temperature-dependent longitudinal resistance ($R_{xx}-T$) measured from 300 to 1.9 K (Figure 2a) exhibits a transition around 20 K, consistent with the AFM transition in MnBi_2Te_4 . To clarify the distinct temperature dependence of R_{yx} and R_{xx} below T_N , we note that the heterostructure contains two magnetically active components with different ordering temperatures. Upon cooling below T_N , the MnBi_2Te_4 layer enters an A-type AFM state, which is accompanied by the upturn in R_{xx} and a large anomalous Hall response that causes the initial decrease in R_{yx} . Near 15 K, the Mn-doped Bi_2Te_3 layer likely undergoes a paramagnetic-to-ferromagnetic transition. While this transition yields only a weak anomaly in the R_{xx} , it introduces an additional Hall component that partially compensates the AFM contribution, resulting in the observed upturn in R_{yx} (Figure S5). Figure 2b and 2c present the magnetic-field-dependent R_{xx} and R_{yx} curves measured at 1.9 K under an out-of-plane magnetic field; both exhibit hysteresis at low magnetic fields with two transitions. Notably, the

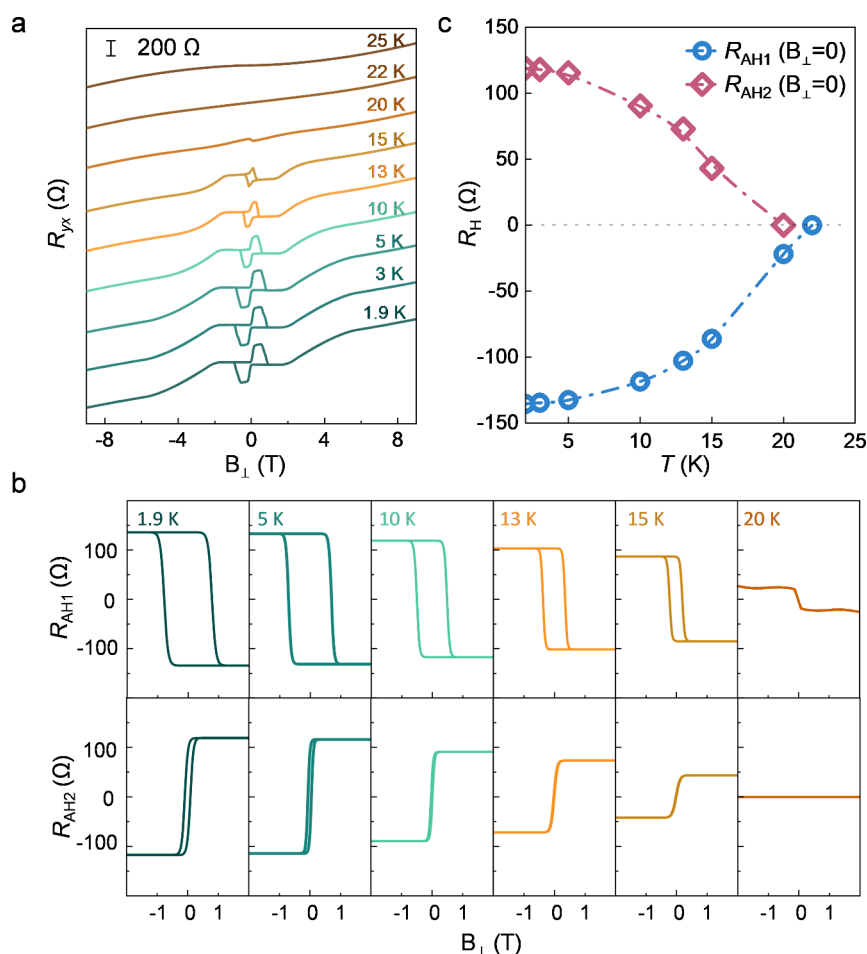


Figure 3. Temperature dependence of the two-channel anomalous Hall resistance. (a) Magnetic-field-dependent R_{yx} at various temperatures for the heterostructure, showing the evolution of hump-dip-featured loops with increasing temperature. The loops shrink gradually with temperature reaching 20 K. (b) The extracted R_{AH1} and R_{AH2} for the two AHE components at various temperatures. The two channels exhibit opposite polarities and critical behaviors. The AH1 channel originates from the 3-SL MnBi_2Te_4 , and the AH2 channel originates from Mn-doped Bi_2Te_3 QLs. (c) Temperature-dependent zero magnetic field R_{AH1} and R_{AH2} , showing zero-field resistances drop monotonically with increasing temperature. R_{AH1} diminishes to zero at 20 K, whereas R_{AH2} persists until a slightly higher temperature of 22 K before vanishing completely.

transitions fields in R_{xx} coincide precisely with those observed in R_{yx} (Figure S6), which indicates that the different field-driven spin reconfigurations of Mn-doped Bi_2Te_3 and MnBi_2Te_4 modify the spin-dependent scattering. Intriguingly, in contrast to pristine MnBi_2Te_4 , the heterostructure exhibits a pronounced centrosymmetric hump-dip hysteresis in the Hall resistance at low fields. Such features are often regarded as signatures of chiral spin textures (e.g., magnetic skyrmions) and can be described either as the superposition of anomalous and topological Hall contributions or, equivalently, as arising from two parallel AHE channels.³²

However, several considerations rule out a skyrmionic origin in the present system. First, both MnBi_2Te_4 and Bi_2Te_3 crystallize in centrosymmetric space groups, which suppresses the Dzyaloshinskii–Moriya interaction (DMI) required to stabilize skyrmions. Moreover, the heterointerface is not expected to generate appreciable DMI. Although MnBi_2Te_4 can host noncollinear magnetic states, such as the surface-spin-flop (SSF) state and the canted AFM phase, these phases occur at magnetic fields far exceeding the regime where the hump-dip anomaly emerges.^{6,35} In this heterostructure, the Mn^{2+} – Mn^{2+} exchange interaction drives the intrinsic A-type AFM of MnBi_2Te_4 , whereas Ruderman–Kittel–Kasuya–Yosida

(RKKY) interaction mediates perpendicular ferromagnetism in the Mn-doped Bi_2Te_3 . As discussed above, no discernible Mn signal is detected in the topmost Bi_2Te_3 layer within the detection limit of our measurements. Consequently, this layer contributes only to the ordinary Hall background and does not introduce an additional AHE channel. Consequently, the observed hysteresis is more naturally understood as the superposition of two independent anomalous Hall contributions, from MnBi_2Te_4 and Mn-doped Bi_2Te_3 , rather than as a manifestation of a topological Hall effect associated with chiral spin textures.

To further elucidate the underlying mechanism, the Hall resistance is decomposed into two anomalous Hall components with an ordinary square hysteresis loops.^{28,32} The multichannel Hall resistance can be well described by

$$R_{\text{AHE}}(H) = R_{\text{AH1}} \tanh\left(\frac{B \pm B_{c1}}{B_{01}}\right) + R_{\text{AH2}} \tanh\left(\frac{B \pm B_{c2}}{B_{02}}\right)$$

where R_{AH1} and R_{AH2} denote the amplitudes of the two AHE components, B_{c1} and B_{c2} are their respective coercive fields, and B_{01} and B_{02} are constants. As shown in Figure 2d, the experimental data can be quantitatively decomposed into two AHE channels with opposite polarities and distinct coercive

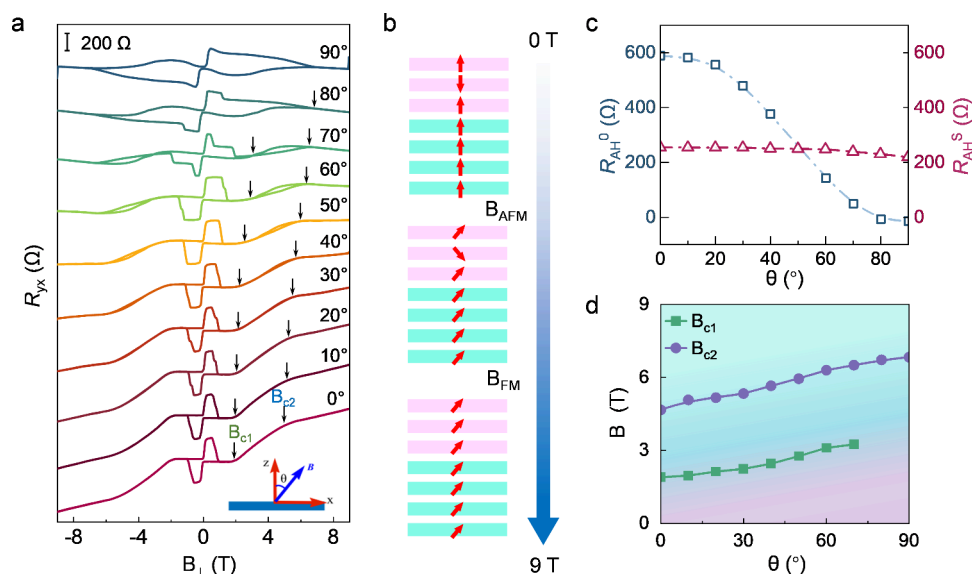


Figure 4. Angular dependence of the two-channel AHE under rotating magnetic fields. (a) Magnetic-field-dependent R_{yx} measured at various rotating angles. (b) Schematic illustration of the magnetic structure evolution under different rotated magnetic fields, showing that MnBi_2Te_4 remains in the AFM state for $B < B_{\text{AFM}}$, transitions to a canted AFM state when $B_{\text{AFM}} < B < B_{\text{FM}}$, and finally evolves into an FM state for $B > B_{\text{FM}}$. (c) Angle-dependent zero magnetic field R_{AH}^0 and Hall resistance at saturation field, R_{AH}^s . (d) B – θ phase diagram of the heterostructure.

fields. The two AHE components are distinguished and assigned based on their distinct coercive fields and the sign of ρ_{yx}^{AHE} . The AH1 component with a larger coercive field is attributed to 3 SL MnBi_2Te_4 with uncompensated magnetization, while the AH2 component with a smaller coercive field originates from the Mn-doped Bi_2Te_3 owing to dilute Mn doping. The signs of the two AHE channel are determined by the Berry curvature of their different occupied bands near the Fermi level, which show opposite polarities as positive in Mn-doped Bi_2Te_3 and negative in MnBi_2Te_4 .^{1,4,44} Moreover, the rapid drop and a following pronounced upturn below T_{N} in the R_{yx} – T curve further indicate the opposite ρ_{yx}^{AHE} signs of the two channels. The sum of these two contributions reproduces the measured Hall response remarkably well. The corresponding transitions in R_{yx} thus can be assigned to distinct magnetic processes: the lower-field feature reflects the spin-flop process (B_{c1}), whereas the higher-field one corresponds to a complete spin-flip transition of the 3-SL MnBi_2Te_4 (B_{c2}).

The distinct characteristics of the two AHE channels suggest different magnetic origins. To clarify this, we perform temperature-dependent magneto-transport measurements and track the evolution of both the Hall hysteresis and the decomposed AHE components. The temperature-dependent R_{yx} and R_{xx} are displayed in Figure 3a and Figure S7, respectively. As temperature increases, the hump-dip Hall hysteresis gradually diminishes and disappears near 20 K (Figure 3a), consistent with the transition of both MnBi_2Te_4 and Mn-doped Bi_2Te_3 toward the paramagnetic state. The centrosymmetric nonsquare Hall resistances can be consistently fitted using the two-channel AHE model with opposite polarities (Figure S8). Figure 3b summarizes the temperature dependence of the extracted R_{AH1} and R_{AH2} . Moreover, we find that both the coercive fields and zero-field resistances decrease with increasing temperature. Notably, the distinct critical temperatures of the two components further reveal their different magnetic origins: the AH2 component vanishes above ~ 20 K, consistent with the Curie temperature of slightly Mn-doped Bi_2Te_3 , while the AH1 component persists up to the

Néel temperature of the 3-SL MnBi_2Te_4 . The extract zero-field Hall resistance as a function of temperature further confirms that the hump-dip AHE stems from the coexistence of two distinct magnetic contributions in the $\text{MnBi}_2\text{Te}_4/\text{Mn-doped Bi}_2\text{Te}_3$ heterostructure (Figure 3c).

To further elucidate the magneto-transport behavior, angle-dependent measurements were performed at 1.9 K under varying magnetic field orientations. As illustrated in the inset of Figure 4a, the tilt angle θ is defined with $\theta = 0^\circ$ corresponding to an out-of-plane magnetic field, and the field is rotated toward the in-plane direction. Figures 4a and S9 display the Hall resistivity R_{yx} and longitudinal resistivity R_{xx} as functions of magnetic field at different θ values. The extracted coercive fields for both the MnBi_2Te_4 (AH1) and Mn-doped Bi_2Te_3 (AH2) contributions scale approximately linearly with $1/\cos(\theta)$ (Figure S10), indicating that each layer individually exhibits the perpendicular magnetic anisotropy. The corresponding magnetic structure evolution of the MnBi_2Te_4 SLs and Mn-doped Bi_2Te_3 QLs is schematically illustrated in Figure 4b. With increasing polar angle θ , the loop at intermediate fields, initially barely discernible, gradually emerges. The additional step-like features at large tilt angles (e.g., 70°) may arise from sequential flips of individual ferromagnetic SLs in MnBi_2Te_4 . Once θ surpasses $\sim 80^\circ$, the loop becomes significantly broadened and persists over nearly the entire field range. This evolution suggests that the out-of-plane magnetic field strongly influences the domain dynamics and the coercive field of the heterostructure. At small tilt angles ($\theta < 40^\circ$), the dominant out-of-plane field aligns the magnetization of both MnBi_2Te_4 and Mn-doped Bi_2Te_3 along the vertical direction. As θ increases ($\theta > 40^\circ$), the reduced perpendicular component leads to incomplete magnetic reversal, and the persistence of hysteresis at high fields indicates strong in-plane pinning. This prevents full recovery of the antiferromagnetic alignment in MnBi_2Te_4 upon field removal.⁴⁵ In MnBi_2Te_4 , the competition among the magnetic anisotropy, AFM exchange, and Zeeman energy leads to two metastable states at intermediate tilt angles (40 – 70°):

canted AFM and more strongly field-induced FM states. During magnetic field sweeping, the system can become trapped in different local minima depending on the sweep direction, leading to the observed hysteresis between $B_{c1}(\theta)$ and $B_{c2}(\theta)$. The coincident angular and field ranges of hysteresis in R_{xx} and R_{yx} indicate that the observed transport features directly reflect the underlying magnetic dynamics.

Overall, Mn-doped Bi_2Te_3 exhibits a lower perpendicular magnetic anisotropy than MnBi_2Te_4 , rendering its magnetization more susceptible to tilted magnetic fields. With increasing in-plane field component, MnBi_2Te_4 evolves into a canted AFM state, in contrast to the field-polarized FM configuration.⁴⁵ In the intermediate field range ($B_{\text{AFM}} < B < B_{\text{FM}}$), MnBi_2Te_4 maintains canted AFM order due to its large magnetic anisotropy energy, while Mn-doped Bi_2Te_3 tends to align in-plane. At high magnetic fields, the anomalous Hall resistance R_{AH}^S decreases approximately following a cosine dependence as θ increases from 0° to 90° (Figure 4c), indicating that the magnetization of both MnBi_2Te_4 and Mn-doped Bi_2Te_3 aligns with the external field in the forced ferromagnetic regime. In contrast, near zero field, the intrinsic AFM order of MnBi_2Te_4 and FM state in Mn-doped Bi_2Te_3 are restored. Therefore, the hump-dip Hall hysteresis is preserved, and the zero-field anomalous-Hall resistance at R_{AH}^0 remains nearly constant.

Despite the evolution of the composite hysteresis with varying θ , the changes of the two transition fields, B_{c1} and B_{c2} , are also observed. Their angular dependence enables us to establish a B - θ phase diagram (Figure 4d), which reveals a sequence of magnetic transitions from AFM to canted AFM and ultimately to FM in the MnBi_2Te_4 SLs. The phase diagram shows two obvious phase boundaries $B_{c1}(\theta)$ and $B_{c2}(\theta)$ indicated by two dashed lines in Figure 4d. The 3-SL MnBi_2Te_4 adopts an AFM state below $B_{c1}(\theta)$, a fully polarized FM state above $B_{c2}(\theta)$, and a canted AFM state in between. Notably, the B_{c1} boundary vanishes when $\theta > 80^\circ$, indicating that the AFM state is gradually polarized into the FM state without passing through a metastable canted AFM phase when the in-plane field component dominates. The reduced out-of-plane field cannot lead to a spin-flop transition. Instead, the moments tilt continuously toward the in-plane field direction, reflecting a gradual polarization of the AFM state into a field-induced ferromagnetic state.

In summary, we realize a $\text{MnBi}_2\text{Te}_4/\text{Mn-doped Bi}_2\text{Te}_3$ vdW heterostructure through controlled two-step CVD growth, which exhibits hump-dip-featured anomalous Hall responses with a clear two-channel origin. Quantitative decomposition of the Hall resistance reveals two distinct AHE components arising from the AFM MnBi_2Te_4 and FM Mn-doped Bi_2Te_3 , respectively, which are further corroborated by temperature-dependent measurements. Furthermore, angle-dependent magnetotransport measurements uncover the evolution of magnetic states under rotating fields, reflecting the distinct magnetic anisotropies of the two components. Our results demonstrate that the superposition of multiple AHE channels can produce Hall responses resembling a topological Hall effect, providing a natural explanation for the debated origin of the hump-dip AHE in MnBi_2Te_4 . This work establishes a viable strategy for engineering multichannel AHE and offers a versatile platform for exploring emergent magneto-transport phenomena in tailored vdW heterostructures.

EXPERIMENTAL METHODS

Synthesis of $\text{MnBi}_2\text{Te}_4/\text{Mn-Doped-Bi}_2\text{Te}_3$ vdW Heterostructures

The $\text{MnBi}_2\text{Te}_4/\text{Mn-doped}$ heterostructures were synthesized via a two-step CVD method using a two-zone tube furnace connected to a glovebox. Tellurium lumps, MnCl_2 powders, and Bi_2O_3 powders were used as precursors, and freshly cleaved mica was employed as the growth substrates. A quartz boat containing tellurium lumps was placed in the upstream zone, while a second quartz boat loaded with MnCl_2 and Bi_2O_3 powders was placed downstream. The mica substrate was oriented face-down directly above the precursors, ensuring efficient exposure to the evaporated species. Prior to growth, the tube was purged with a mixture of H_2/Ar (1:9, v/v) gas for 10 min. The furnace was then ramped up to $500/550^\circ\text{C}$ in 15 min under a H_2/Ar carrier gas at a flow rate of 100 sccm (standard cubic centimeters per minute). During the initial low-temperature stage, the limited evaporation of MnCl_2 precursors favors the formation of Mn-doped Bi_2Te_3 . Subsequently, elevated temperatures enabled the epitaxial growth of MnBi_2Te_4 on top of the Mn-doped Bi_2Te_3 layers. This temperature-dependent sequential growth exploits the distinct vapor pressures of the precursors, leading to the formation of a well-defined van der Waals heterostructure.

Sample Characterizations

The morphology of the synthesized heterostructures was characterized using an Olympus BX51-SC30 optical microscope. Raman spectra were obtained on a WITec ALPHA300R system. The atomic force microscopy images were collected by OXFORD Cypher S AFM. Specimens for STEM were prepared using the focused ion beam (FIB). STEM characterization was performed on an aberration-corrected JEOL GRANDARM2 operated at 200 kV. The beam convergence semiangle was 32 mrad, while the collection semiangles were 68–280 mrad for high-angle annular dark-field (HAADF) imaging and 17 mrad for annular bright-field (ABF) imaging. The HAADF images were processed using Wiener filtering. For energy-dispersive X-ray spectroscopy (EDS) analysis, the convergence semiangle was also set to 32 mrad, and the probe current was approximately 81 pA. For the EELS analysis, the convergence semiangle was set to 32 mrad, and the collection semiangle was approximately 21 mrad.

Transfer of the Heterostructure

The $\text{MnBi}_2\text{Te}_4/\text{Mn-doped-Bi}_2\text{Te}_3$ heterostructures were transferred from the mica to the SiO_2/Si substrate by using hydrophobic polystyrene (PS) polymer. First, 2 g PS particles were dissolved in 20 mL of toluene, and then the PS solution was spin-coated (3000 rpm for 60 s) on the mica substrates. Subsequently, it was baked for 20 min at 60°C , thus forming a solid polymer layer covering the samples after evaporation of the solvent. Then, the polymer film which includes the samples is wedged in water and separated quickly with mica because of surface tension forces. The SiO_2/Si substrate served as the receiver substrate to pick up the polymer. After the PS/heterostructures film is successfully transferred to the SiO_2/Si substrate, they were baked for 40 min at 80°C and the PS was dissolved in toluene.

Device Fabrication and Transport Measurements

The Hall bar devices based on heterostructures were fabricated by standard electron beam lithography (Raith 150), followed by the deposition of Cr/Au (5/50 nm) metal as electrodes through thermal evaporation. After the lift-off process, the standard Hall electrodes were obtained. Electrical transport measurements were carried out on a Quantum Design physical property measurement system (PPMS).

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.6c01588>.

Schematic of the CVD setup for synthesizing the Bi_2Te_3 heterostructures, Raman spectra of the heterostructure and MnBi_4Te_7 , atomic force microscopy image, additional structural and elemental characterizations, a zoom-in comparison of $R_{xx}-T$ and $R_{yx}-T$, comparison of $R_{xx}-B$ and $R_{yx}-B$, magnetic-field-dependent R_{xx} at various temperatures, fitting of the R_{yx} magnetic-field-dependent R_{xx} at various angles, coercive fields of AH1 and AH2 as a function of $1/\cos(\theta)$ (PDF)

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Author Contributions

#H.-J.G. and H.T.Y. supervised and coordinated the research project. H.G. and C.Y.B. synthesized the materials. C.Y.B., H.G., Z.Q.W., J.A.S., X.L., and W.Z. performed the structural characterizations. C.Y.B., H.G., K.Z., S.H.L., R.W.W., Y.C.H., G.J.H., and L.H.B. performed the device fabrication and transport measurements. All of the authors participated in analyzing the experimental data, plotting figures, and writing the manuscript. C.B. and Z.W. contributed equally to this work.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the Ministry of Science and Technology of China (2022YFA1204100), the National Natural Science Foundation of China (62488201, 52572188), the Chinese Academy of Sciences (YSBR-003, YSBR-053), and the Innovation Program of Quantum Science and Technology (2021ZD0302700).

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