Interplay between quantum anomalous Hall effect and magnetic skyrmions

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Quantum anomalous Hall effect (QAHE) and magnetic skyrmion (SK) represent two typical topological states in momentum ($\mathbf{K}$) and real ($\mathbf{R}$) spaces, respectively. However, little is known about the interplay between these two states. Here, we propose that the coexistence of QAHE and SK may generate a previously unknown SK state, named the $\mathbf{R}\mathbf{K}$ joint topological skyrmion (SK), which is characterized by the SK surrounded by nontrivial chiral boundary states (CBSs). Interestingly, beyond the traditional SK state that can solely be used via creation or annihilation, the number and chirality of CBS reflect the different chirality. Beyond the conventional SK, which may be tunable under external fields as demonstrated in Janus monolayer (ML) MnBi$_2$X$_2$Te$_2$ ($X = S, Se$), creating additional degrees of freedom for SK-state manipulations. Moreover, it is also found that external fields can induce a continuous topology phase transition from $\mathbf{K}$-space QAHE to $\mathbf{R}$-space SK in ML MnBi$_2$X$_2$Te$_2$, providing an ideal platform to understand the cross-over phenomena of multiple-space topologies.

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In this article, combining density functional theory (DFT) calculations and Monte Carlo (MC) simulations (Materials and Methods), we propose that applying both external biaxial strain ($\varepsilon$) and magnetic field ($B_z$) can realize a reversible topological phase transition between $K$-space QAHE and $R$-space SK/SkX states in monolayer (ML) MnBi$_2$X$_2$Te$_2$ ($X$ = S, Se), in which our proposed RK-SK/SkX states with tunable $N_{CBS}$ can appear in a certain region of the $\varepsilon$–$B_z$ phase diagram. With the increase of $\varepsilon$, the $K$-space topology of ML MnBi$_2$X$_2$Te$_2$ will transform from high-$C$ insulator ($C = +2$) to low-$C$ insulator ($C = -1$) to trivial insulator ($C = 0$) due to the $\varepsilon$-controlled different types of band inversions; simultaneously, the $R$-space topology can transform from trivial ferromagnetic (FM) phase to nontrivial SK phase to nontrivial SkX phase as the result of the dramatically different $\varepsilon$-dependent behaviors for collinear magnetic interaction and DM interaction. Therefore, the ML MnBi$_2$X$_2$Te$_2$ can serve as an ideal platform to understand the interplay and cross-over of multiple-space topological phenomena.

Results and Discussion

Band Topology of ML MnBi$_2$X$_2$Te$_2$. As shown in Fig. 2A, the ML MnBi$_2$X$_2$Te$_2$ is a Janus variant (36–39) created by replacing Te with $X$ in the bottom two layers of ML MnBi$_2$Te$_4$ (40), forming an inversion-asymmetric structure. This septuple structure has the $P3m1$ symmetry, which contains three mirror operations and a threefold rotation with respect to the $z$ axis (Fig. 2B). The Mn atoms locate at the center of the edge-sharing distorted octahedra, arranging in a triangle lattice and contributing to the long-range magnetic order. The same as the ML MnBi$_2$Te$_4$ (40), the FM configuration is the magnetic ground state of ML MnBi$_2$X$_2$Te$_2$ (SI Appendix, Fig. S1 and Table S1). The magnetic anisotropy energy (MAE) calculation shows that the easy axis of ML MnBi$_2$X$_2$Te$_2$ is along the $z$ direction. The dynamical and thermodynamic stabilities of ML MnBi$_2$X$_2$Te$_2$ are confirmed by the phonon dispersion calculations (SI Appendix, Fig. S2) and ab initio molecular dynamics simulations (SI Appendix, Fig. S3).

Fig. 2. Topological phase transition of ML MnBi$_2$S$_2$Te$_2$ in $K$ space. (A) Side view of crystal structure of ML MnBi$_2$X$_2$Te$_2$. (B) Top view of the middle Te–Mn–X layer in ML MnBi$_2$X$_2$Te$_2$. (C) Orbital-projected band structure of ML MnBi$_2$S$_2$Te$_2$ in FM configuration with the SOC effect. Valence band maximum is set to zero. (D) Topological phase diagram of ML MnBi$_2$S$_2$Te$_2$ under tensile biaxial strain $\varepsilon$. Insets are the band structures near the two phase transition points.
respectively. It should be noted that Janus materials usually have higher energy compared with the disordered counterparts (SI Appendix, Figs. S4 and S5 and Tables S2 and S3). Given the successful growth of the Janus ML MoSe2 and antismirically decorated graphene (36–38), it is reasonable to expect that other Janus materials, including ML MnBi2X2Te2, are also experimentally accessible. In the following, we will focus on the discussion of ML MnBi2X2Te2 and only briefly mention the results of ML MnBi2Se2Te2.

Fig. 2C shows the band structure of ML MnBi2X2Te2 in the FM configuration. Similar to ML MnBi2Te4 (40), the electronic states around the Fermi level (E_F) are mainly contributed by Bi-3p and Te-3p orbitals; the Mn-3d states that are extremely localized far away from E_F (SI Appendix, Fig. S6) exhibit much larger exchange splitting (~7 eV) than crystal-field splitting (~1 eV), leading to a high spin configuration of SIA (1). Different from trivial insulator ML MnBi2Te4, the anticrossing between Bi-p_z and Te-p_x,y states around the Γ-point at E_F in ML MnBi2X2Te2 (SI Appendix, Fig. S7) can open a sizeable bandgap of ~66 meV when the SOC effect is included, indicating a topological nontrivial nature. Indeed, the calculated Chern number is C = +2, indicating that ML MnBi2X2Te2 is an intrinsic high-C QAH insulator (41). Interestingly, although the band structures of ML MnBi2Se2Te2 and ML MnBi2S2Te2 are similar, the ML MnBi2X2Te2 is an intrinsic low-C QAH insulator (C = −1) with a bandgap of ~28 meV (SI Appendix, Figs. S6 and S8) (41).

Since the absolute volume deformation potentials of Bi–p_z and Te–p_x,y orbitals are different, it is expected that the topological properties of ML MnBi2X2Te2 in K space can be tuned by applying external biaxial strain ε. Interestingly, different from most known QAH insulators, it is found that multiple topological phase transitions can emerge under a reasonable ε-region. As shown in Fig. 2D, when 0.0% < ε < 2.4%, the ML MnBi2X2Te2 maintains the high-C insulator phase with C = +2. When ε > 2.4%, the first phase transition appears, induced by the band inversion at the Q point on the Γ–K line (Fig. 2D, Left Inset). Due to threefold rotation symmetry, there are three equivalent Q points in the first Brillouin zone. Therefore, three band inversions happen at the same time, leading to the decrease of C by −3 (i.e., the ML MnBi2X2Te2 is converted from a C = +1 phase to a C = −1 one).

When 2.4% < ε < 5.0%, the ML MnBi2X2Te2 maintains the low-C insulator phase with C = −1. When ε = 5.0%, the second phase transition appears, induced by the band inversion at the Γ-point (Fig. 2D, Right Inset). Consequently, the C further changes from −1 to 0 (i.e., ML MnBi2X2Te2 is eventually converted to a trivial insulator phase) (more details are in SI Appendix, Fig. S9). Similarly, the diagram of the +2 → −1 → 0 topological phase transition is also found in ML MnBi2Se2Te2 but in different ε-regions (SI Appendix, Figs. S10 and S11).

**Spin Hamiltonian and Magnetic Interaction in ML MnBi2X2Te2.**

The highly localized feature of Mn-3d states (SI Appendix, Fig. S6) means that the magnetic property of ML MnBi2X2Te2 can be captured using the spin Hamiltonian with the general form (42, 43)

\[
H = \frac{1}{2} \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_i \sum_{\alpha, \beta} D_{\alpha \beta} S^\alpha_i S^\beta_i + \sum_i A_{\alpha \beta} S^\alpha_i S^\beta_i, \tag{1}
\]

where \(S^\alpha_i, S^\beta_i\) represent the spin operators; \(i, j\) are the magnetic sites; and \(\alpha, \beta\) run over three cartesian indices. The first and second terms represent the exchange interaction and single-ion anisotropy (SIA), respectively, and the magnetic coupling matrix \(J\) and \(A\) can be obtained from the DFT calculations. Similar to ML MnBi2Te2 (40), the symmetric part of the nearest-neighbor (NN) exchange matrix of ML MnBi2X2Te2 is diagonal with the isotropic exchange strength \(J = (J^{xx} + J^{yy} + J^{zz})/3\). The calculated \(J = −2.27\) meV reflects an FM interaction, originating from the nearly ~90° superexchange interaction (44, 45). Due to the symmetry of ML MnBi2X2Te2, the SIA matrix \(A\) is diagonal with \(A^{xx} = A^{yy}\), and the effective SIA parameter is \(A = A^{xx} − A^{zz}\). The calculated \(A = −0.17\) meV indicates that the \(z\) direction is the preferred spin orientation, consistent with the MAE calculations. Based on the isotropic exchange interaction and out-of-plane SIA, the Curie temperature \(T_C\) of ML MnBi2X2Te2 is estimated to be ~28 K (SI Appendix, Fig. S13). For ML MnBi2Se2Te2, the smaller magnetic coupling strengths \(J\) and \(A\) lead to a lower \(T_C\) ~23 K (SI Appendix, Figs. S12 and S13).

The antisymmetric part of the exchange matrix represents the DM interaction (30, 31), whose strength can be written as \(D_{\alpha \beta} = \varepsilon_{\alpha \beta \gamma} (J^{\alpha \beta} − J^{\beta \alpha})/2\), where \(\varepsilon_{\alpha \beta \gamma}\) is the Levi–Civita symbol. The asymmetric structure leads to a nonzero DM interaction (46–51). For the ML MnBi2X2Te2, the calculated NN DM vector \(D_{01} = (0.02, 0.52, −0.29)\) in units of millielectron volts of the \(\mathrm{Mn}_0\)–\(\mathrm{Mn}_1\) pair (marked in Fig. 2B) lies in a plane perpendicular to the x axis (within a tiny numerical error), consistent with the restriction of the Moriya rule (31), due to a mirror plane passing through the middle point of the line between \(\mathrm{Mn}_0\) and \(\mathrm{Mn}_1\). Considering the \(C_{6v}\) symmetry, all six NN DM vectors of each Mn have a unified form \(D_{ij} = D_{\parallel} (\varepsilon_{\alpha \beta \gamma} e_{\alpha} \times e_{\beta} + 1) e_{\gamma}\), where \(e_{\alpha}\) and \(e_{\beta}\) are the unit vectors along the \(\mathrm{Mn}_0\)–\(\mathrm{Mn}_1\) bond and z axis, respectively. Therefore, the six DM vectors surround each Mn in a staggered counterclockwise configuration (49–51). For ML MnBi2Se2Te2, a relatively weaker DM interaction with \(D_{01} = (−0.02, 0.24, −0.10)\) in units of millielectron volts is obtained because the weaker electronegativity of Se than S causes weaker antisymmetric distortion in ML MnBi2Se2Te2 than in ML MnBi2S2Te2. Compared with the DM interaction in the surface of MnBi2Te4 (52), the intrinsic DM interactions in ML MnBi2X2Te2 are sufficiently strong, which is the crucial ingredient for forming SK.

**RK-SK State and Magnetic Phase Diagram in ML MnBi2X2Te2.**

Based on the above understanding, Eq. 1 can be simplified to

\[
H = \frac{1}{2} \sum_{\langle i,j \rangle} J S_i \cdot S_j + \frac{1}{2} \sum_{\langle i,j \rangle} D_{ij} \cdot (S_i \times S_j) + \sum_i A S_i^x S_i^z, \tag{2}
\]

where \(\langle i,j \rangle\) represents the summation runs over the NN sites \(i, j\). As the DM interaction prefers to make the spin mutual perpendicularity, the interplay between the DM interaction and the collinear magnetic interaction may generate noncollinear magnetic structures (e.g., SK) (12–16). The competition between different magnetic interactions can be quantified using the factor \(\alpha = 4\sqrt{\mathcal{A}/\pi D}\) (53, 54). The calculated \(\alpha = 1.33\) in ML MnBi2S2Te2 suggests the possibility for the existence of Néel-type SK in this system. Starting from Eq. 2, we have performed the MC simulations to calculate the spin textures (55) in R space for ML MnBi2X2Te2. As shown in Fig. 3A, the ground state of ML MnBi2X2Te2 has a spin spiral (SS) configuration. To stabilize SK, the Zeeman term \(H_z = g \mu_B S B\), should be included, where \(g\) is the Landé factor and \(\mu_B\) is the Bohr magneton. As shown in Fig. 3B, due to the relatively strong DM interaction, the isolated SK with a radius \(R_{SK}\) ~3.4 nm appears under \(B_z = 0.1\) T, with the swirling number \(S = −1\), and its spin direction continuously rotates from the −z direction (in the center) to the x−y direction (in the boundary). Importantly, the nature of high-C phase in ML MnBi2S2Te2 gives rise to the fundamentally different
The topological phase transition of ML MnBi$_2$S$_2$Te$_2$ in $R$ space. Spin textures of ML MnBi$_2$S$_2$Te$_2$ under (A) $B_z = 0.0$ T and (B) $B_z = 0.1$ T. The color map corresponds to the spatial distribution of the out-of-plane spin component ($S_z$). (C) Isotropic exchange strength $J$ and SIA parameter $A$ of ML MnBi$_2$S$_2$Te$_2$ as a function of $\varepsilon$. (D) In-plane component $D_0$ and out-of-plane component $D_1$ of the DM vector $D_0$ for ML MnBi$_2$S$_2$Te$_2$ as a function of $\varepsilon$. (E) Magnetic phase diagram of ML MnBi$_2$S$_2$Te$_2$ as a function of $\varepsilon$ and $B_z$. The gray-shaded area shows the regions where the $R$K-$SK/SkX$ with $\varepsilon$-tunable $N_{CBS}$ can appear.

$C_{\text{inner}} = -2$ and $C_{\text{outer}} = +2$ inside and outside the SK region, respectively, leading to the appearance of $R$K-SK with $N_{CBS} = C_{\text{inner}} - C_{\text{outer}} = -4$ (Fig. 3B). Since the estimated penetration depth ($\sim 3.6$ nm) of CBS in ML MnBi$_2$S$_2$Te$_2$ (56) is comparable with the $R_{\text{SK}}$ of SK, the $R$K-SK state can exist, although a tiny hybridization gap ($\sim 10$ meV) may exist among these CBSs (SI Appendix, Fig. S16). Different from the ML MnBi$_2$S$_2$Te$_2$, the calculated $\alpha = 2.38$ in ML MnBi$_2$Se$_2$Te$_2$ gives rise to the absence of the SK state (SI Appendix, Figs. S10, S11, and S15).

The tunable magnetic interactions via strain may further modulate the topological spin textures of ML MnBi$_2$S$_2$Te$_2$. Generally, the increase of $\varepsilon$ will increase the bond angle $\theta_{\text{Mn} - X - \text{Mn}}$, weakening the FM superexchange interaction (57), and increase the localization of Mn-3$d$ states, weakening the $p$-$d$ hybridization between Mn and its NN ligands. These result in the decrease of $J$ with the increase of $\varepsilon$, as shown in Fig. 3C. A similar weakening behavior is found for $A$, originating from the weakening of $p$-$d$ hybridization and crystal-field splitting. As shown in Fig. 3D, it is surprisingly found that in contrast to $J$ and $A$, the in-plane $D$ ($D_0$) is insensitive to the variable $\varepsilon$, and the out-of-plane $D$ ($D_1$) can even be largely increased when $\varepsilon$ increases. Since the DM interaction derives from the SOC effect on the isotropic exchange interaction, its strength is determined by the exchange interaction of adjacent magnetic sites as well as the SOC-induced orbital coupling between the occupied and empty Mn-3$d$ states (31, 58). Therefore, although the exchange interaction weakens as the $\varepsilon$ increases, the reduced energy gap between occupied and empty states may enhance their interorbital coupling, resulting in the unexpected $\varepsilon$-dependent behaviors of the DM interaction in Fig. 3D. For ML MnBi$_2$Se$_2$Te$_2$, a similar $\varepsilon$-dependent magnetic interaction is observed (SI Appendix, Fig. S12).

The dramatically different $\varepsilon$-dependent $J/A$ and $D$ strongly indicate that the noncollinear magnetic structure will be rather sensitive to $\varepsilon$. Fig. 3E shows the magnetic phase diagram of ML MnBi$_2$S$_2$Te$_2$ as a function of $\varepsilon$ and $B_z$. When $B_z = 0.0$ T, the ML MnBi$_2$S$_2$Te$_2$ maintains the SS phase, and the width of the worm-like pattern decreases as the $\varepsilon$ increases (SI Appendix, Fig. S14). Under a moderate $B_z$ (e.g., $B_z \sim 0.2$ T), the sparsely isolated SKs appear when $0.0\% < \varepsilon < 1.5\%$, as the relatively weak DM interaction leads to $\alpha > 1.0$. When $1.5\% < \varepsilon < 3.0\%$, $\alpha < 1.0$, the SKs with high density arrange in a lattice form (i.e., the SkX phase appears); when $\varepsilon > 3.0\%$, the contribution of DM interaction becomes more important, leading to the appearance of SS spin texture, and the system enters the phase of coexistence of SK and SS states. Since $B_z$ tends to align spins in the $z$ direction, the size of SK could be tunable by $B_z$ (e.g., it will gradually decrease as the $B_z$ increases) (SI Appendix, Fig. S14). Therefore, when $B_z > 0.2$ T and $0.0\% < \varepsilon < 1.5\%$, the system will eventually enter the FM phase. When $B_z > 0.2$ T and $\varepsilon > 1.5\%$, $\alpha < 1.0$, the $B_z$ will eliminate the worm-like pattern and results in SkX phase. 

External Field–Tunable $R$K-SK/SkX States and Topology Cross-Over from $K$ Space to $R$ Space. In general, the $R$K-SK state can appear in the coexistence phase of the QAHE insulator and SK as long as the CBSs with well-defined chirality surrounding the $R$K-SK can be well distinguished from the bulk states. Here, we focus on the $R$K-SK/SkX with tiny hybridization gap opening among the CBSs (i.e., their $R_{\text{SK}}$ should be at least comparable with the penetration depth of CBS (59)), so that the original chirality of CBSs can be largely maintained. As shown in the gray-shaded area of Fig. 3E, when $0.0\% < \varepsilon < 2.4\%$ and under an appropriate $B_z$, the ML MnBi$_2$S$_2$Te$_2$ maintains the $C = +2$ phase, and $N_{CBS} = -4$ results in the $R$K-SK encircled by four CBS modes (Fig. 3B). When $\varepsilon > 2.4\%$, ML MnBi$_2$S$_2$Te$_2$ is converted to the $C = -1$ phase; therefore, the $N_{CBS}$ will be changed from $-4$ to $+2$ (i.e., the $R$K-SK is now encircled by two CBS modes with opposite chirality). Interestingly, as shown in Fig. 3E, while the $N_{CBS} = -4$ state can exist either in isolated $R$K-SK or in $R$K-SkX in the different regions of the $\varepsilon$–$B_z$ phase diagram, the $N_{CBS} = +2$ state can solely exist in $R$K-SkX. Although the number and chirality of CBSs in $R$K-SK can be tunable via external fields, the contribution of CBSs to the global transport properties of the system may depend on the exact Fermi-level position (SI Appendix, section VIII, which includes ref. 60). It should be noted that these CBSs may be visible by the local $dI/dV$ spectrum in experiments, providing an additional degree of freedom for information storage during local manipulation of individual SK. For ML MnBi$_2$Se$_2$Te$_2$, the $R$K-SK/SkX states cannot exist as the QAHE phase and SK states cannot coexist (SI Appendix, Figs. S10, S11, and S15).

Other than the $R$K-SK/SkX states, as shown from Fig. 3E, it is interesting to find that the multiple-space topology cross-over
can be achieved in ML MnBi$_2$S$_2$Te$_2$ via tuning external fields. For example, as shown in Fig. 4, we choose four different ε-cases to calculate their K-space edge states and R-space spin textures. In K space, when ε = 0.0% and ε = 2.0%, the two “in-gap” chiral states with ε-tunable positive group velocity clearly show the topological nontrivial nature of ML MnBi$_2$S$_2$Te$_2$ in C = +2 phase. When ε = 3.0%, the number and group velocity of “in-gap” chiral states are changed to one and negative value, respectively, consistent with C = −1 phase of ML MnBi$_2$S$_2$Te$_2$ in this ε region; when ε = 6.0%, the vanishing chiral edge state represents a topological trivial state of ML MnBi$_2$S$_2$Te$_2$. On the other hand, in R space, when ε = 0.0%, the ML MnBi$_2$S$_2$Te$_2$ exhibits the trivial FM configuration. When ε = 2.0%, the Néel-type RK-SkX appears with $N_{CBS} = -4$. When ε = 3.0%, the Néel-type RK-SkX with $N_{CBS} = +2$ appears. When ε = 6.0%, the system transforms into the mixing phase with the coexistence of smaller-sized SKs and worm-like pattern SS as the result of stronger DM interaction. Therefore, other than the RK-SK states with tunable $N_{CBS}$, an ideal topology cross-over in multiple spaces, as proposed in Fig. 1, can also be well achieved in ML MnBi$_2$S$_2$Te$_2$. For ML MnBi$_2$Se$_2$Te$_2$, a similar multiple-space topology cross-over is also found but without the appearance of RK-SK states (SI Appendix, Figs. S10, S11, and S15).

Finally, it should be noted that the ML MnBi$_2$Te$_4$ has been successfully synthesized in the experiments (9, 61). On the other hand, the approaches for growing 2D Janus structures have also been successfully demonstrated in several different materials, creating many exotic applications (36–38). Therefore, we expect that the Janus ML MnBi$_2$X$_2$Te$_2$ system associated with the quantum phenomena we proposed here could stimulate the experimental efforts in the future.

### Outlook and Conclusion

To further control the skyrmion size, as the ML MnBi$_2$X$_2$Te$_2$ has out-of-plane polarization, we expect that the fine tuning of magnetic interactions may be achieved via the external electric field apart from external strain, leading to the effective manipulation of skyrmion size. In addition, despite the similar CBS that may exist in the domain walls between the regions with opposite magnetization, the magnetic domains are usually topologically trivial in R space, preventing the realization of RK joint topological matter, like RK-SK.

In summary, we propose that the concept of the RK-SK state with external field–tunable $N_{CBS}$ may appear when the SK state exists in a QAH insulator under certain conditions. Beyond the conventional SK states that can mainly be used by creation and annihilation, the number and chirality of CBS provide additional degrees of freedoms for quantum-state manipulations. Combining DFT calculations and MC simulations, we predict the ML MnBi$_2$X$_2$Te$_2$ is a unique platform to realize the multiple-space topology cross-over from K-space QAHE to R-space SK/SkX states. Most importantly, the external field–tunable RK-SK/SkX states can appear in a certain region of the ε–B diagram. Our findings not only may provide an idea to design SK states beyond the traditional ones, but also, they may provide opportunities to explore interplay and cross-over phenomena between multiple-space topologies and generate conceptual dissipationless spintronic applications.

### Materials and Methods

**DFT Calculations**

The DFT calculations were performed using the Vienna ab initio Simulation Package (VASP) (62). In our calculations, the Perdew–Burke–Ernzerhof functional (63) was used to approximate the exchange correlation functional in the Kohn–Sham equation, and the projector-augmented wave method (64) was chosen to treat the core electrons. The energy cutoff for the plane-wave basis was chosen to be 400 eV, and the convergence criterion for the total energy was set to be 1.0 × 10$^{-5}$ eV in all the calculations. A 20-Å vacuum layer was adopted to avoid the influence of the periodic images. An 18 × 18 × 1 uniform Γ-centered k-point mesh was used to ensure the accuracy of our calculations. The rotationally invariant generalized gradient approximation (GGA) + U method (65) was used to correct the strong correlation effect derived from the partially occupied Mn-3d states. The value of effective Hubbard U$_{eff}$ was chosen to be 3.0 eV, which was verified to be accurate and suitable to describe the electronic structures of MnBi$_2$Te$_4$ family materials (7, 40).

The phonon dispersion in the FM configuration was obtained using the finite displacement method. First, the crystal structure was relaxed until the force of each atom is less than 0.001 eV/A. Second, the 5 × 5 × 1 supercell combined with the 3 × 3 × 1 uniform Γ-centered k-point mesh was used to...
perform self-consistent calculations to obtain the force constant matrix. Finally, the Fourier transform and diagonalization of the force constant matrix were performed using the Phonopy Package (66).

The thermodynamic stabilities of ML MnBi$_2$X$_2$Te$_2$ were confirmed by performing ab initio molecular dynamics simulations as implemented in VASP. We employed the NVT canonical ensemble and the Nose–Hoover thermostat (67, 68) to perform simulations. The energy cutoff for the plane-wave basis set was set to 350 eV, and the convergence criterion for the total energy was set to 1.0 × 10$^{-6}$ eV here. The 5 × 5 × 1 supercells combined with the single Γ-point were adopted to perform simulations, and the time step was set to be 2 fs. All simulations were performed for 8 ps (4,000 steps) to ensure that the systems reach equilibrium.

Tight-Binding Model and Edge-States Calculations. The tight-binding model was generated using the projected Wannier function approach as implemented in the Wannier90 Package (69). The edge state was calculated by iteratively solving the surface Green’s function of the seminfinite system (70).

Magnetic Interaction and MC Simulations. The magnetic interaction matrix was calculated using the four-states energy mapping method (71). Here, all matrix elements were obtained by performing the GGA + U calculations including the SOC effect and constraining the direction of magnetic moments. The 3 × 3 × 1 supercell was used to remove the interactions caused by the periodic boundary conditions; meanwhile, the 4 × 4 × 1 Γ-centered k-point mesh was adopted to maintain the same density of grid as in the unit cell calculations. The total energy was converged to < 1.0 × 10$^{-7}$ eV for each supercell to ensure the reliability of the calculated magnetic interaction parameters.

The R-space spin texture was obtained by performing the classical MC simulations, in which the metropolis algorithm was adopted to generate spin configurations according to the Boltzmann probability distribution (72).

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