Ferroelectrically controlled topological magnetic phase in a Janus-magnet-based multiferroic heterostructure

Qirui Cui,^{1,2} Yingmei Zhu,¹ Jiawei Jiang,¹ Jinghua Liang,¹ Dongxing Yu,¹ Ping Cui,^{1,2} and Hongxin Yang^{1,*}

¹Key Laboratory of Magnetic Materials and Devices, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, China

and Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China

²Faculty of Science and Engineering, University of Nottingham Ningbo China, Ningbo 315100, China

(Received 14 April 2021; revised 10 August 2021; accepted 10 September 2021; published 5 October 2021)

Electric control of topological magnetic phases has attracted extensive attention due to its potential applications in energy-efficient spintronic devices. Here, using first-principles calculations and atomistic spin model simulations, we demonstrate that electric control of topological magnetic phases can be realized in Janus-magnet-based multiferroic heterostructure, i.e., $\text{MnBi}_2\text{Se}_2\text{Te}_2/\text{In}_2\text{Se}_3$. The loops of vortices and antivortices can be transformed into skyrmions with diameter of only 4 nm via ferroelectricity reversal, which is originated from the change of magnetic anisotropy. For heterostructure with up polarization, loops of vortices and antivortices are further tuned to bimeron solitons by applying in-plane magnetic field. Our results thus pave the way for achieving highly tunable topological magnetism in atomic-thickness heterostructure, which can be useful in nonvolatile data encoding and storage with low-energy consumption.

DOI: [10.1103/PhysRevResearch.3.043011](https://doi.org/10.1103/PhysRevResearch.3.043011)

I. INTRODUCTION

The magnetic skyrmion, as a topologically protected spin texture, is highly promising for nonvolatile information carrier in next-generation spintronic devices with high-storage density and low-energy consumption because it has small size, stable configuration, and can be driven by low-threshold current [1–8]. One key parameter for realizing skyrmion in materials is Dzyaloshinskii-Moriya interaction (DMI), which is originated from spin-orbit coupling (SOC) and inversion symmetry breaking (ISB) [9–12]. In the last decades, many efforts have been devoted to constructing ferromagnetic/heavy-metal heterostructures (HSs) with perpendicular magnetic anisotropy (PMA) and large DMI to realize skyrmion [13–17]. Recently, through constructing Janus structures to introduce ISB, the large DMI can be realized in two-dimensional (2D) magnetic materials leading to the formation of skyrmion [18–21]. Compared with multilayers of ferromagnetic/heavy metal, a single 2D magnet has simpler structure, smaller size, and is more beneficial for reducing defects. Besides skyrmion, the bimeron is another topologically nontrivial spin texture consisting of a vortex and an antivortex, which can be constructed by rotating the spin direction of skyrmion by an angle of 90° [22–24]. The bimeron can also be used as a nonvolatile information carrier for spintronic devices, based on materials with in-plane (IP) magnetic anisotropy (IMA) [25,26].

Another crucial task for practical application of topological spin textures, such as skyrmion and bimeron, is searching or developing a method that can effectively control the mobility and morphology of these spin textures. Particularly, using electric field is considered as a more energy-dissipationless method compared with spin-polarized current, thermal excitation, and external magnetic field (B_{ext}) [27–31]. However, since electric field does not break time-reversal symmetry directly, realizing control of magnetism by electricity is difficult [28]. Notably, previous works have demonstrated that the multiferroic system which combines ferroelectricity (FE), ferromagnetism, and ISB provides an ideal platform for electric control of chirality [32,33], creation/annihilation [34], density, and thermal stability [35] of topological magnetism through switching the FE polarization. Here, using first-principles calculations and atomistic spin model simulations, we unveil that a type of FE-controlled topological magnetism can be realized in Janus-magnet-based multiferroic heterostructure, $\text{MnBi}_2\text{Se}_2\text{Te}_2(\text{MBST})/\text{In}_2\text{Se}_3$. The loops of vortices and antivortices can be transformed into skyrmions by switching the direction of ferroelectric polarization of In_2Se_3 . Furthermore, in MBST/ In_2Se_3 with up polarization, bimeron solitons can be obtained by applying IP magnetic field. Further analysis unravels that the variation of interlayer charge transfers induced by FE reversal gives rise to the change of magnetic anisotropy, which is responsible for the topological magnetic phase transition in system.

II. COMPUTATIONAL METHODS

Our first-principles calculations are performed using the Vienna *Ab initio* Simulation Package (VASP), which is based on the density-functional theory (DFT) [36–38]. The exchange-correlation interaction is described by the

*hongxin.yang@nimte.ac.cn

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

FIG. 1. Top and side views of the crystal structure of (a) MBST and (b) In_2Se_3 . (c) Side views of MBST/ $P\uparrow$ and MBST/ $P\downarrow$ HSs. Green, purple, blue, brown, and red balls represent Se, Bi, Mn, Te, and In elements, respectively. The orange vectors in (c) indicate two spin configurations with opposite chirality used to extract the in-plane DMI strength.

generalized gradient approximation (GGA) based on the Perdew-Burke-Ernzerhof function [39,40]. The GGA + U method is applied ($U_{\text{eff}} = 3$ eV) to describe strongly correlated $3d$ electrons of Mn [41,42]. The projector-augmented wave method is adopted [43], and the cutoff energy is set to 420 eV. The Brillouin zone is sampled using $24 \times 24 \times 1$ Γ -center k -point grids. The vacuum space of at least 15 Å is used to avoid interactions between periodic replicas. The electronic convergence is performed with a tolerance of 10^{-7} eV. All structures are fully relaxed until the Hellmann-Feynman force on each ion is less than 10^{-3} eV/Å. The interlayer van der Waals interaction is corrected by the DFT-D3 method [44]. Dipole corrections are included in calculations of multiferroic HS, and for obtaining the electric polarization of systems, we directly integrate ρ_z over the supercell, where ρ represents the local charge density and z represents coordinates along the out-of-plane (OOP) direction [45]. More computational details can be found in the Appendix.

III. RESULTS AND DISCUSSION

The crystal structure of Janus MBST monolayer is shown in Fig. 1(a). MBST consists of septuple Te-Bi-Te-Mn-Se-Bi-Se layers, with atoms in each layer forming a 2D triangular lattice. MBST can be constructed by replacing triple Te-Bi-Te layers of MnBi_2Te_4 (MBT) monolayer with triple Se-Bi-Se layers [46], where MBT monolayer is a topologically trivial ferromagnetic semiconductor [47,48]. The crystal symmetry of MBST is C_{3v} , and intrinsic ISB allows for DMI between Mn atoms. The calculated magnetic moment of MBST is $5\mu_B$ per unit cell, indicating that the Mn atom has a high spin configuration. Furthermore, we demonstrate the dynamic and thermal stability of MBST by calculating the phonon dispersions (see Appendix, 1), elucidating absence of imaginary frequency in Brillouin zone, and *ab initio* molecular dynamic simulations (AIMD) (see Appendix, 2). Notably, similar ferroelectric heterostructures, such as $\text{WS}_2/\text{In}_2\text{Se}_3$ and $\text{WSe}_2/\text{In}_2\text{Se}_3$, have been successfully fabricated in experiments by dry transfer methods [49,50], and Janus monolayers of transition-metal dichalcogenides, i.e., MoSSe and PtSSe , can be synthesized by controlling the reaction atmosphere [51,52]. These results highlight the possibility of construction of MBST/ In_2Se_3 HS in experiments.

In_2Se_3 monolayer is a 2D van der Waals material with room-temperature FE [45,53] and small lattice mismatch with

MBST [54], and its crystal structure is shown in Fig. 1(b). One can see that the In_2Se_3 monolayer consists of five triangular atomic layers stacked in sequence of Se-In-Se-In-Se, and the asymmetric position of Se atoms in the central layer gives rise to the IP and OOP polarizations simultaneously. When In_2Se_3 is used as substrate to construct HS with MBST, the reversal of OOP polarization in the In_2Se_3 monolayer can induce distinct interfacial coupling, further resulting in the variation of magnetic properties of MBST. Next, we consider 12 possible stacking configurations of MBST/ In_2Se_3 HS as discussed in Appendix, 3. We find that the most stable configuration is the outmost Te and Bi atoms of MBST sitting at the hollow and top-In sites of In_2Se_3 , respectively, for both polarized-up ($P\uparrow$) and -down ($P\downarrow$) states, and their side views are shown in Fig. 1(c). The equilibrium interlayer distance of MBST/ $P\uparrow$ and $P\downarrow$ are 2.93 and 2.74 Å, respectively. The total energy of MBST/ $P\uparrow$ is higher by 0.053 eV/u.c. than MBST/ $P\downarrow$, due to the weaker interfacial coupling between MBST and In_2Se_3 with upward polarization [55]. The electric polarization for MBST/ $P\uparrow$ and MBST/ $P\downarrow$ HS is 0.073 and -0.081 eÅ/u.c., respectively, and the magnitude of that of the In_2Se_3 monolayer is 0.096 eÅ/u.c., which implies that the ferroelectricity of HS mainly comes from In_2Se_3 . Based on the climbing-image nudged elastic band method, we obtain that the energy barrier of ferroelectric phase transitions of In_2Se_3 is 0.083 eV. Recent experiments have demonstrated that an OOP field around 1 V/nm can overcome such a barrier and flip the OOP polar order [56]. Therefore, it can be expected that a similar electric field is large enough for realizing the reversal of polar orientation of MBST/ In_2Se_3 HS.

To investigate magnetic properties of systems, we adopt the following spin Hamiltonian:

$$H = -J \sum_{\langle i,j \rangle} \mathbf{S}_i \mathbf{S}_j - K \sum_i (S_i^z)^2 - \sum_{\langle i,j \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) - \mu_{Cr} B \sum_i S_i^z, \quad (1)$$

where $\mathbf{S}_i$ is a unit vector representing local spin of the i th Mn atom, and $\langle i,j \rangle$ represents summation of all nearest-neighbor (NN) Mn pairs. J , K , and $\mathbf{D}_{ij}$ represent the Heisenberg exchange coupling, magnetic anisotropy, and DMI, respectively. The methods to calculate these parameters are given detail in Appendix, 4, and their results are shown in Table I. We adopt the sign convention that $J > 0$

TABLE I. Calculated nearest-neighboring exchange coupling J , magnetocrystalline anisotropy K_{MCA} , magnetic shape anisotropy K_{MSA} , effective anisotropy K_{eff} , and IP DMI component $d_{\parallel}$ of MBST, MBST/ $P\uparrow$, and MBST/ $P\downarrow$ in units of meV.

	J_{xx}	J_{yy}	J_{zz}	J	K_{MCA}	K_{MSA}	K_{eff}	$d_{\parallel}$
MBST	1.657	1.650	1.668	1.658	0.045	-0.205	-0.160	0.307
MBST/ $P\uparrow$	1.323	1.318	1.304	1.315	0.006	-0.204	-0.198	0.323
MBST/ $P\downarrow$	1.626	1.627	1.662	1.638	0.287	-0.203	0.084	0.217

($J < 0$) represents the ferromagnetic (antiferromagnetic) coupling, $K > 0$ ($K < 0$) indicates PMA (IMA), and $d_{\parallel} > 0$ ($d_{\parallel} < 0$) favors clockwise (anticlockwise) spin configuration. We first calculate the NN exchange coupling for magnetization along positive directions of x , y , and z axis, i.e., J_{xx} , J_{yy} , and J_{zz} . The SOC effects are taken into account. One can see that the exchange coupling of MBST is ferromagnetic, and J_{xx} , J_{yy} , and J_{zz} are almost identical, which indicates that the exchange coupling for MBST is nearly isotropic. Accordingly, we define an isotropic parameter J as the average of J_{xx} , J_{yy} , and J_{zz} , which represents the coefficient in the normally used $-J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$ term. We also notice that the magnitude of J of MBST is 1.658 meV, which is obviously larger than $J = 0.922$ meV when SOC is not considered. Therefore, the SOC effects cannot be neglected in calculations of exchange coupling for MBST systems. The FE substrate does not change the type of exchange coupling of MBST, and J of MBST/ $P\uparrow$ ($P\downarrow$) is 1.315 (1.638) meV. The effective anisotropy K_{eff} contains two parts: magnetocrystalline anisotropy K_{MCA} and magnetic shape anisotropy K_{MSA} , where the first part arises from SOC and the second part is caused by magnetostatic dipole-dipole interaction; i.e., $K_{\text{eff}} = K_{\text{MCA}} + K_{\text{MSA}}$. One can see that MBST, MBST/ $P\uparrow$, and MBST/ $P\downarrow$ all have positive K_{MCA} . Interestingly, the K_{MCA} of HS is significantly enhanced from 0.006 to 0.287 meV when the polarization direction of In_2Se_3 is tuned from up to down. Contrary to K_{MCA} , K_{MSA} favors the IP alignment of magnetization and basically does not change in three systems. For MBST and MBST/ $P\uparrow$, K_{MSA} reaches to -0.205 and -0.204 meV, respectively, whose magnitudes are much larger than K_{MCA} . Therefore, K_{eff} are negative for both MBST and MBST/ $P\uparrow$. However, due to the giant enhancement of K_{MCA} , K_{eff} in MBST/ $P\downarrow$ becomes positive. Therefore, the IMA-to-PMA transition is achieved in HS by FE modulation. The transition also indicates that it is very important to investigate K_{MSA} in calculations of magnetic anisotropy for magnetic thin films. We also test the influence of U_{eff} on magnetic anisotropy and find that the IMA-to-PMA transition induced by FE reversal is robust to U_{eff} in the range of 2 to 5 eV (see Table II). Moreover, we find that MBST, MBST/ $P\uparrow$, and MBST/ $P\downarrow$ all process sizable DMI reaching to 0.307, 0.323, and 0.217 meV, respectively. We further calculate the layer-resolved SOC energy difference ΔE_{soc} between the opposite chiral spin configurations (see Fig. 2). The detailed computational methods are given in Appendix, 5. For MBST/ In_2Se_3 HS, the DMI still comes from the MBST layer. The Mn layer basically does not contribute to the DMI strength, whereas the most adjacent Te and Bi layers provide dominant contribution, which means

TABLE II. K_{MCA} , K_{MSA} , and K_{eff} of HS with $U_{\text{eff}} = 2/4/5$ eV.

U_{eff}	MBST/ $P\uparrow$			MBST/ $P\downarrow$		
	K_{MCA}	K_{MSA}	K_{eff}	K_{MCA}	K_{MSA}	K_{eff}
2	0.008	-0.198	-0.190	0.340	-0.197	0.143
4	0.015	-0.209	-0.194	0.250	-0.209	0.041
5	0.024	-0.214	-0.190	0.230	-0.214	0.016

that the spin tilting is caused by spin-orbit scattering from heavy elements [12].

Once magnetic parameters are obtained, we perform atomistic spin model simulations by using VAMPIRE [57] in square geometry with a side length of 60 nm. In all simulations, the periodic boundary is used along x and y axes, and the initial magnetization state is set to random. More computational details are given in Appendix, 6. The magnetic topology of spin textures can be defined by the topological charge: $Q = \frac{1}{4\pi} \int \mathbf{m} \cdot (\partial_x \mathbf{m} \times \partial_y \mathbf{m}) dx dy$, where $\mathbf{m}$ is the normalized magnetization. Both skyrmion and bimeron can be attributed to $Q = \pm 1$. The sign of Q determines the orientation of transverse motion of these topological quasiparticles in the skyrmion Hall effect [3,24]. Figure 3 shows spin textures of MBST, MBST/ $P\uparrow$, and MBST/ $P\downarrow$. For pristine MBST, the loops of vortices and antivortices appear [see Fig. 3(a)], which arise from the competition between DMI and weak IMA [58]. By applying an IP magnetic field along the positive direction of the x axis ($B \rightarrow$), the vortex-antivortex pairs generate and become ordered along the direction of external field [see Fig. 4(a)]. Interestingly, the bimeron soliton with diameter of around 9 nm emerges [see Fig. 3(b)] while $B \rightarrow$ increases to 60 mT. Compared with MBST, denser loops of vortices and antivortices emerge [see Fig. 3(c)] in MBST/ $P\uparrow$, since the ferromagnetic exchange coupling is weaker. Moreover, loops in MBST/ $P\uparrow$ can also be transformed into bimeron solitons gradually via enhancing IP magnetic field [see Figs. 3(d) and 4(b)]. Due to the same outer magnetization, bimerons with opposite topological charges ($Q = \pm 1$) can coexist [23].

The reversal of electric polarization in In_2Se_3 from up to down tunes magnetic anisotropy of HS from IP to OOP

FIG. 2. Layer-resolved localization of the SOC energy difference ΔE_{soc} between opposite chiral spin configurations in MBST (black bar) monolayer, MBST/ $P\uparrow$ (red bar) and MBST/ $P\downarrow$ (blue bar) HS.

FIG. 3. The spin textures for MBST monolayer, MBST/ $P\uparrow$ and MBST/ $P\downarrow$ HSs in real space. Corresponding external fields are labeled above each panel. The color map indicates the out-of-plane spin component of Mn atoms.

while the DMI slightly decreases, which indicates that there is obvious variation of topological magnetism. For 2D magnets processing hexagonal lattice, it is convenient to introduce the dimensionless parameter, $\kappa = (\frac{4}{\pi})^2 \frac{2J K_{\text{eff}}}{3d_{\parallel}^2}$ [59–61]. The detailed derivation of κ is given in Appendix, 7. Notably, κ is independent of the magnet thickness but only determined by the magnetic parameters, i.e., J , K_{eff} , and $d_{\parallel}$, which are directly obtained from first-principles calculations. When $0 < \kappa < 1$, the magnetic ground state of the system exhibits spin spiral, whereas for $\kappa > 1$, the isolated skyrmion could be generated in the background of ferromagnetism. For MBST/ $P\downarrow$, κ is 3.15, which is close to the critical value ($\kappa = 1$) of transition to the spiral state. Therefore, isolated skyrmions are hopefully stabilized. Atomistic spin model simulations demonstrate that the multiple Néel-type skyrmions with diameter of around 4 nm appear in MBST/ $P\downarrow$ [see Fig. 3(e)] under zero magnetic field. We thus obtained the FE-controlled topological mag-

netic structures in MBST/ In_2Se_3 HS. The distinct chiral spin textures in MBST/ $P\uparrow$ and MBST/ $P\downarrow$ are potentially used as “0” and “1” bit carriers for realizing the binary data encoding and storage. Strikingly, the size of skyrmion in MBST/ $P\downarrow$ is very small, which can largely enhance the integration level of corresponding devices. The skyrmions with opposite topological charges ($Q = \pm 1$) appear at distinct domains, and applying an 80 mT OOP magnetic field along the positive direction of the z axis ($B\uparrow$) to shrink the blue domain, the skyrmions with $Q = +1$ are annihilated [see Fig. 3(f)].

Since the FE-controlled magnetic phase transition is dominantly caused by large variation of MCA, we elucidate the physical mechanism of magnetic anisotropy energy (MAE) of MBST/ In_2Se_3 HS. From layer-resolved MAE [see Fig. 5(a)], one can find that the MAE of HS just comes from MBST, and the PMA of Te which is adjacent to the Mn atom is enhanced significantly while the polarization of In_2Se_3 reverses, which responds for K_{MCA} increasing. For further understanding MAE change of Te atom, we calculate the MAE from orbital hybridization as shown in Figs. 5(b) and 5(c). One can see that the hybridization between p_y and p_z provides IMA, while hybridization between p_y and p_x gives rise to PMA. The PMA contribution arises from p_y and p_x and is obviously increased when the polarization direction of In_2Se_3 reverses from up to down. Moreover, we analyze the MAE change sourcing from $5p$ orbitals of Te based on second-order perturbation theory [62–64]. MAE can be expressed as

$$\text{MAE} = \xi^2 \sum_{\sigma\sigma'} \sum_{\sigma''\sigma'''} \frac{(2\delta_{\sigma\sigma'} - 1)(|\langle \sigma'' | L_z | u^{\sigma'} \rangle|^2 - |\langle \sigma'' | L_x | u^{\sigma'} \rangle|^2)}{E_u^{\sigma''} - E_o^{\sigma''}}, \quad (2)$$

FIG. 4. Spin textures of (a) MBST monolayer under in-plane magnetic field of 30 mT and (b) MBST/ $P\uparrow$ HS under IP magnetic field of 40 mT. The color map indicates the OOP spin component of Mn atoms.

FIG. 5. (a) Layer-resolved MAE of MBST/P↑ and MBST/P↓ HSs. (b), (c) MAE coming from $5p$ orbitals hybridization, and (d), (e) projected density of states of Te which is adjacent to Mn atom in MBST/In₂Se₃ HS.

where $|o^\sigma\rangle$ and $|u^{\sigma'}\rangle$ are occupied states with spin σ and unoccupied states with spin σ' , respectively, whose energy eigenvalues are E_o^σ and $E_u^{\sigma'}$. $(2\delta_{\sigma\sigma'} - 1)(|\langle o^\sigma | L_z | u^{\sigma'} \rangle|^2 - |\langle o^\sigma | L_x | u^{\sigma'} \rangle|^2)$ represents the difference of SOC elements as given in Table III. Figures 5(d) and 5(e) show the projected density of states for Te. One can see that spin-up occupied $p_{x(y)}$ states ($p_{x(y)}^{u+}$) shift across the Fermi level (E_F) for HS with $P\downarrow$, which is indicated by the dashed black arrow in Fig. 5(e). The difference of SOC elements between $p_{x(y)}^{o+}$ and spin-up unoccupied $p_{y(x)}^{u+}$ ($p_{y(x)}^{u+}$) is 1 [see Table III], which responds for PMA coming from p_x and p_y hybridization. The shift of $p_{x(y)}^{o+}$ means that the value of $(E_u^{\sigma'} - E_o^\sigma)$ obviously decreases. Therefore, the positive MAE from p_x and p_y hybridization enhances. It is also found that peaks of spin-up and spin-down occupied p_z states (p_z^{o-} and p_z^{o+}) are close to each other. Since SOC elements difference of p_y^{u+} , p_z^{o-} and p_y^{u+} , p_z^{o+} have opposite signs and same amplitudes (see Table III), the MAE contribution from p_y and p_z barely changes. The fact that FE reversal-induced PMA enhancement is due to the shift of electronic states of Te inspires us to further investigate the interlayer charge transfers of HS. Bader analysis [65–68] shows that the transferred charge from MBST to In₂Se₃ obviously increases

TABLE III. The matrix differences between magnetization along positive directions of x and z in Eq. (2). u^+ , o^- , and o^+ represent unoccupied spin-up, occupied spin-down and spin-up states, respectively.

u^+	o^-			o^+		
	p_y	p_z	p_x	p_y	p_z	p_x
p_y	0	1	-1	0	-1	1
p_z	1	0	0	-1	0	0
p_x	-1	0	0	1	0	0

from 0.019 to 0.049e when FE polarization is reversed from up to down, resulting in occupied states of MBST shifting across E_F [see Fig. 5(e)]. Furthermore, the planar-average charge-density difference (see Fig. 6) clearly shows that charge transfers are mainly restricted in the interface of MBST/P↑ but spread over all In₂Se₃ layer for MBST/P↓ [69], which is consistent with charge-transfer analysis.

In summary, using first-principles calculations and atomistic spin model simulations, we demonstrate that by modulating FE polarization of MBST/In₂Se₃ HS, the IMA-to-PMA transition is achieved, resulting in that loops of vortices and antivortices are transformed into skyrmions with diameter of only 5 nm. Furthermore, the loops in MBST/P↑ can be turned to bimeron solitons via applying IP magnetic field. Detailed analysis elucidates that charge transfers between MBST and

FIG. 6. Planar-average charge-density difference of MBST/P↑ (left panel) and MBST/P↓ (right panel) HSs. The blue and black dots represent the positions of MBST and In₂Se₃ atoms, respectively.

FIG. 7. Calculated phonon band structure for the MBST monolayer.

In_2Se_3 play a crucial role in determining magnetic anisotropy of HS. When the FE polarization is reversed from up to down, the $p_{x(y)}^{o+}$ states of Te which is adjacent to Mn shifts across the E_F due to the increase of interlayer transferred charge, resulting in large enhancement of PMA contribution from Te. The high tunability of topological magnetism is promising to be applied in nonvolatile information handling with low-power consumption. Since the effect is robust, more nontrivial phenomena of controlling of topological magnetism can be expected in HSs consisting of Janus magnets and ferroelectric/antiferroelectric substrate.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China, Grants No.11874059 and No. 12174405; Key Research Program of Frontier Sciences, CAS, Grant No. ZDBS-LY-7021; Zhejiang Provincial Natural Science Foundation of China, Grant No. LR19A040002; and Beijing National Laboratory for Condensed Matter Physics.

APPENDIX

1. Phonon calculations

In phonon calculations, the real-space force constants are computed with a $4 \times 4 \times 1$ supercell and $5 \times 5 \times 1$ $\mathbf{k}$ -point grids using the VASP code. The force constant matrices and phonon dispersions are calculated using the PHONOPY code [70]. Figure 7 shows the phonon band structure of the monolayer $\text{MnBi}_2\text{Se}_2\text{Te}_2$ (MBST). There is no imaginary frequency, which demonstrates the dynamic stability of MBST.

2. Molecular dynamics

For verifying the thermal stability of MBST, we perform AIMD implemented in VASP. The canonical NVT ensemble is used with the Nosé thermostat [71]. For NVT ensemble, the number of particles (N), cell volume (V) and temperature (T) keeps same throughout simulations. A $4 \times 4 \times 1$ supercell with a $1 \times 1 \times 1$ $\mathbf{k}$ -point grid is adopted. Each time step is

set to 1 fs, and simulations are performed for 10 000 steps at 300 K. The evolution of the total energy and temperature are shown in Figs. 8(a) and 8(b). The top and side views [see Figs. 8(c) and 8(d)] of MBST after simulations indicate that the structure is still maintained at initial phase.

3. Heterostructure configurations

Considering structural symmetries of In_2Se_3 and MBST, we propose 12 stacking configurations of MBST/ In_2Se_3 HS (see Fig. 9). The total energies of optimized structures are shown in Fig. 10. Compared with the Se-terminated side, In_2Se_3 prefers to contact the Te-terminated side of MBST. For both MBST/ $P\uparrow$ and $P\downarrow$, the configuration 6, with outmost Te and Bi atoms of MBST sitting at the hollow and top-In sites of In_2Se_3 , is most stable.

4. Calculations of J , K , and $d_{\parallel}$

(i) For obtaining NN exchange coupling J , we compare the energy difference of ferromagnetic and antiferromagnetic states of a $2 \times 1 \times 1$ supercell, i.e., $J = (E_{\text{AFM}} - E_{\text{FM}})/8$. The SOC effects are taken into account. Moreover, to determine the influence of next-nearest-neighboring (NNN) interactions, we expand the unit cell of MBST and calculate the DFT total energies of three different magnetic configurations (see Fig. 11). The total energies of three magnetic configurations are given by

$$E_{\text{FM}} = -12J_1 - 12J_2 + E_{\text{other}},$$

$$E_{\text{AFM1}} = -4J_1 + E_{\text{other}},$$

$$E_{\text{AFM2}} = -4J_1 + 4J_2 + E_{\text{other}},$$

where J_1 and J_2 represent NN and NNN exchange coupling, respectively. Here, SOC effects are also considered, and the magnetization is set to be along the positive direction of the z axis. We find that $J_1 = 1.674$ meV and $J_2 = 0.164$ meV. The magnitude of J_1 is much larger than that of J_2 , and is very close to the that directly obtained from the 2×1 supercell. These results indicate that the NNN coupling is almost negligible in MBST systems. (ii) The magnetocrystalline anisotropy K_{MCA} is calculated as: $K_{\text{MCA}} = E_{M\rightarrow} - E_{M\uparrow}$, where $E_{M\rightarrow}$ and $E_{M\uparrow}$ represent the self-consistent total energy of system with magnetization along the positive direction of x and z axes, respectively, when SOC effect is included. The magnetic shape anisotropy K_{MSA} is the sum of all magnetostatic dipole-dipole interactions up to infinity [72,73] and is calculated by $K_{\text{MSA}} = E^{\text{dip}}/V_{\text{u.c.}} = -\frac{1}{2} \frac{\mu_0}{4\pi V_{\text{u.c.}}} \sum_{i \neq j}^N \frac{1}{r_{ij}^3} [\mathbf{M}_i \cdot \mathbf{M}_j - (\mathbf{M}_i \cdot \mathbf{r}_{ij})(\mathbf{M}_j \cdot \mathbf{r}_{ij}) \frac{3}{r_{ij}^2}]$, where $\mathbf{M}_i$ and $\mathbf{r}_{ij}$ represent local magnetic moments and vector connecting site i and j , respectively. A $250 \times 250 \times 1$ supercell is used in K_{MSA} calculation to ensure the reliability of results. Also, we would like to note that skyrmions and merons are small topological defects on the uniform magnetic background [4,6], which indicates that shape anisotropy can be considered as the contribution to effective magnetic anisotropy. This approximation has been used in micromagnetic simulations of complex spin textures, such as skyrmion and chiral domain wall [1,32], and gives good agreement with experiments [13]. (iii) The chirality-dependent total energy

FIG. 8. (a) The evolution of total energy and (b) temperature during the *ab initio* simulations for MBST monolayer at 300 K. The (c) top and (d) side views of MBST supercell after simulations of 10 ps.

difference approach is applied to obtain the DMI strength [12]. Since the out-of-plane components cancel each other, we only provide the in-plane component of DMI, $d_{\parallel}$. In calculations of DMI, a $4 \times 1 \times 1$ supercell with $6 \times 24 \times 1$ k -point grids is

adopted. To obtain $d_{\parallel}$, we calculate the energies of clockwise (CW) and anticlockwise (ACW) spin configurations as shown in Fig. 1(c) of the main text, and $d_{\parallel}$ equals $(E_{ACW} - E_{CW})/12$. (iv) In fact, the spin Hamiltonian for describing spin interactions between Mn atoms can be written as $H = \vec{S}_i \cdot \vec{J} \cdot \vec{S}_j$, where the exchange matrix has components

$$\vec{J} = \begin{bmatrix} J_{xx} & \Gamma_{xy} + D_z & \Gamma_{xz} - D_y \\ \Gamma_{xy} - D_z & J_{yy} & \Gamma_{yz} + D_x \\ \Gamma_{xz} + D_y & \Gamma_{yz} - D_x & J_{zz} \end{bmatrix}.$$

Here, the J and D represent Heisenberg exchange coupling and DMI, respectively. The Γ_{xy} , Γ_{xz} , and Γ_{yz} are the additional, symmetric, off-diagonal contributions to the exchange tensor. For determining Γ of MBST, such as Γ_{xy} , we compare the energies of a $2 \times 1 \times 1$ supercell with spin configurations of Mn pairs as $\{(S \ 0 \ 0), (0 \ S \ 0)\}$ and $\{(-S \ 0 \ 0), (0 \ S \ 0)\}$. For

FIG. 9. Top and side views of 12 stacking configurations for MBST/In₂Se₃ HS s.

FIG. 10. Total energies of 12 stacking configurations for (a) MBST/P $\uparrow$ and (b) MBST/P $\downarrow$ HSs. The dashed line indicates the most stable stacking.

FIG. 11. Three different spin configurations applied to calculate the NN and NNN exchange coupling parameters.

$\{(S\ 0\ 0), (0\ S\ 0)\}$, $E_1 = J_{xx} + J_{xy} + 4\Gamma_{xy}$, and for $\{(S\ 0\ 0), (0\ S\ 0)\}$, $E_2 = J_{xx} + J_{xy} - 4\Gamma_{xy}$. Accordingly, $\Gamma_{xy} = (E_1 - E_2)/8$. Γ_{xy} , Γ_{xz} , and Γ_{yz} of MBST are 0.0027, 0.0038, and 0.0018 meV, respectively. One can see that the magnitude of Γ is very limited and is much smaller compared with DMI components. These results indicate that Γ can be negligible for MBST systems.

5. Calculations of layer-resolved localization of the SOC energy difference ΔE_{soc} between opposite chiral spin configurations and layer-resolved MAE

The SOC is calculated for each atom layer in MBST and MBST/In₂Se₃ HSs. By comparing the atom-layer SOC energy between ACW and CW configurations, and in-plane (100) and out-of-plane (001) magnetic orientations, one can obtain layer-resolved ΔE_{soc} and MAE, respectively.

6. Atomic spin model simulations

All atomic spin model simulations are performed using the VAMPIRE package [57]. The hexagonal lattice of MBST is transformed into a lattice with an orthorhombic unit cell (see Fig. 12). Then, this unit cell is extended to a 60×60 -nm square with the periodic boundaries.

FIG. 12. The orthorhombic unit cell for MBST and MBST/In₂Se₃ HSs. The purple atoms represent Mn element.

FIG. 13. The spin textures for MBST monolayer with λ in range of 0.1 to 0.4. The color map indicates the out-of-plane spin component of Mn atoms.

The time evolution of atomic spin is described by the Landau-Lifshitz-Gilbert (LLG) equation. The LLG is given by: $\frac{\partial \mathbf{S}_i}{\partial t} = -\frac{\gamma}{(1+\lambda^2)}[\mathbf{S}_i \times \mathbf{B}_{\text{eff}}^i + \lambda \mathbf{S}_i \times (\mathbf{S}_i \times \mathbf{B}_{\text{eff}}^i)]$, where $\mathbf{S}_i$ is the unit vector of the spin moment of site i , γ is gyromagnetic ratio, and λ represents damping constant. $\mathbf{B}_{\text{eff}}^i$ represents the effective magnetic field on each spin and can be expressed as $\mathbf{B}_{\text{eff}}^i = -\frac{1}{\mu_s} \frac{\partial H}{\partial \mathbf{S}_i}$, where H is the spin Hamiltonian of MBST. We choose $\lambda = 0.2$ for MBST, considering magnetism originates from 3d electrons of Mn, and Bi and Te process strong SOC [74]. Moreover, we calculate spin textures of MBST with λ in the range of 0.1 to 0.4 (see Fig. 13). One can find that loops of vortices and antivortices emerge, which are robust to the value of λ .

7. The κ for MBST monolayer

The dimensionless parameter κ is calculated by $\kappa = (\frac{4}{\pi})^2 \frac{AK}{D^2}$, where A is exchange stiffness, and K and D are the magnetic anisotropy and DMI parameter in micromagnetic format, respectively. Based on the Heisenberg model, the exchange energy can be expressed as $E_{\text{ex}} = J \sum_{i,\delta} (\delta_i \cdot \nabla \mathbf{m}_i)^2$, where J is the magnitude of exchange coupling, and δ_i is the vector between neighboring local spin $\mathbf{m}_i$. In micromagnetic format, $E_{\text{ex}} = A \int dV [(\nabla m_x)^2 + (\nabla m_y)^2 + (\nabla m_z)^2]$, where A is the exchange stiffness. For MBST monolayer, one can obtain $A = \frac{\sqrt{3}J}{t_F}$, where t_F is the assumed thickness of magnetic film. The micromagnetic anisotropy is calculated as $K = \frac{K_{\text{eff}}}{V} = \frac{2K_{\text{eff}}}{\sqrt{3}a^2 t_F}$, where a is the lattice constant. The micromagnetic DMI strength D is calculated as $D = \frac{d_{\parallel} \sqrt{3}}{at_F}$ [12], where $d_{\parallel}$ is the IP component of DMI. Therefore, κ can be rewritten as $\kappa = (\frac{4}{\pi})^2 \frac{2JK_{\text{eff}}}{3d_{\parallel}^2}$.

- [1] J. Sampaio, V. Cros, S. Rohart, A. Thiaville, and A. Fert, Nucleation, stability and current-induced motion of isolated magnetic skyrmions in nanostructures, *Nat. Nanotechnol.* **8**, 839 (2013).
- [2] R. Wiesendanger, Nanoscale magnetic skyrmions in metallic films and multilayers: A new twist for spintronics, *Nat. Rev. Mater.* **1**, 1 (2016).
- [3] W. Jiang, X. Zhang, G. Yu, W. Zhang, X. Wang, M. B. Jungfleisch, J. E. Pearson, X. Cheng, O. Heinonen, K. L. Wang, and Y. Zhou, Direct observation of the skyrmion Hall effect, *Nat. Phys.* **13**, 162 (2017).
- [4] A. Fert, A. Reyren, and V. Cros, Magnetic skyrmions: Advances in physics and potential applications, *Nat. Rev. Mater.* **2**, 1 (2017).
- [5] J. Jena, B. Göbel, V. Kumar, I. Mertig, S. Felser, and S. Parkin, Evolution and competition between chiral spin textures in nanostripes with D_{2d} symmetry, *Sci. Adv.* **6**, eabc0723 (2020).
- [6] X. Zhang, Y. Zhou, K. M. Song, T. E. Park, J. Xia, M. Ezawa, X. Liu, W. Zhao, G. Zhao, and S. Woo, Skyrmion-electronics: Writing, deleting, reading and processing magnetic skyrmions toward spintronic applications, *J. Phys.: Condens. Matter.* **32**, 143001 (2020).
- [7] K. M. Song, J. S. Jeong, B. Pan, X. Zhang, J. Xia, S. Cha, T. E. Park, K. Kim, S. Finizio, J. Raabe, and J. Chang, Skyrmion-based artificial synapses for neuromorphic computing, *Nat. Electron.* **3**, 148 (2020).
- [8] W. Legrand, D. Maccariello, F. Ajejas, S. Collin, A. Vecchiola, K. Bouzehouane, N. Reyren, V. Cros, and A. Fert, Room-temperature stabilization of antiferromagnetic skyrmions in synthetic antiferromagnets, *Nat. Mater.* **19**, 34 (2020).
- [9] I. Dzyaloshinsky, A thermodynamic theory of “weak” ferromagnetism of antiferromagnetics, *J. Phys. Chem. Solids* **4**, 241 (1958).
- [10] T. Moriya, Anisotropic superexchange interaction and weak ferromagnetism, *Phys. Rev.* **120**, 91 (1960).
- [11] A. Fert and P. M. Levy, Role of Anisotropic Exchange Interactions in Determining the Properties of Spin-Glasses, *Phys. Rev. Lett.* **44**, 1538 (1980).
- [12] H. Yang, A. Thiaville, S. Rohart, A. Fert, and M. Chshiev, Anatomy of Dzyaloshinskii-Moriya Interaction at Co/Pt Interfaces, *Phys. Rev. Lett.* **115**, 267210 (2015).
- [13] O. Boulle, J. Vogel, H. Yang, S. Pizzini, D. de Souza Chaves, A. Locatelli, T. O. Montes, A. Sala, L. D. Buda-Pregbeanu, O. Klein, M. Belmeguenai, Y. Roussigné, A. Stashkevich, S. M. Chérif, L. Aballe, M. Foerster, M. Chshiev, F. Auffret, L. M. Miron, and G. Gaudin, Room-temperature chiral magnetic skyrmions in ultrathin magnetic nanostructures, *Nat. Nanotechnol.* **11**, 449 (2016).
- [14] A. Soumyanarayanan, M. Raju, A. G. Oyarce, A. K. Tan, M. Y. Im, A. P. Petrović, P. Ho, K. H. Khoo, M. Tran, C. K. Gan, F. Ernult, and C. Panagopoulos, Tunable room-temperature magnetic skyrmions in Ir/Fe/Co/Pt multilayers, *Nat. Mater.* **16**, 898 (2017).
- [15] H. Yang, O. Boulle, V. Cros, A. Fert, and M. Chshiev, Controlling Dzyaloshinskii-Moriya interaction via chirality dependent atomic-layer stacking, insulator capping and electric field, *Sci. Rep.* **8**, 12356 (2018).
- [16] M. Raju, A. Yagil, A. Soumyanarayanan, A. K. Tan, A. Almoalem, F. Ma, O. M. Auslaender, and C. Panagopoulos, The evolution of skyrmions in Ir/Fe/Co/Pt multilayers and their topological Hall signature, *Nat. Commun.* **10**, 1 (2019).
- [17] B. Cui, D. Yu, Z. Shao, Y. Liu, H. Wu, P. Nan, Z. Zhu, C. Wu, T. Guo, P. Chen, H. A. Zhou, L. Xi, W. Jiang, H. Wang, S. Liang, H. Du, K. L. Wang, W. Wang, K. Wu, X. Han, G. Zhang, H. Yang, and G. Yu, Néel-type elliptical skyrmions in a laterally asymmetric magnetic multilayer, *Adv. Mater.* **33**, 2006924 (2021).
- [18] J. Liang, W. Wang, H. Du, A. Hallal, K. Garcia, M. Chshiev, A. Fert, and H. Yang, Very large Dzyaloshinskii-Moriya interaction in two-dimensional Janus manganese dichalcogenides and its application to realize skyrmion states, *Phys. Rev. B* **101**, 184401 (2020).
- [19] Y. Zhang, C. Xu, P. Chen, Y. Nahas, S. Prokhorenko, and L. Bellaiche, Emergence of skyrmionium in a two-dimensional CrGe(Se, Te)₃ Janus monolayer, *Phys. Rev. B* **102**, 241107(R) (2020).
- [20] Q. Cui, J. Liang, Z. Shao, P. Cui, and H. Yang, Strain-tunable ferromagnetism and chiral spin textures in two-dimensional Janus chromium dichalcogenides, *Phys. Rev. B* **102**, 094425 (2020).
- [21] C. Xu, J. Feng, S. Prokhorenko, Y. Nahas, H. Xiang, and L. Bellaiche, Topological spin texture in Janus monolayers of the chromium trihalides Cr(I, X)₃, *Phys. Rev. B* **101**, 060404(R) (2020).
- [22] N. Gao, S. G. Je, M. Y. Im, J. W. Choi, M. Yang, Q. C. Li, T. Y. Wang, S. Lee, H. S. Han, K. S. Lee, W. Chao, C. Hwang, J. Li, and Z. Q. Qiu, Creation and annihilation of topological meron pairs in in-plane magnetized films, *Nat. Commun.* **10**, 1 (2019).
- [23] K. W. Moon, J. Yoon, C. Kim, and C. Hwang, Existence of in-Plane Magnetic Skyrmion and Its Motion Under Current Flow, *Phys. Rev. Appl.* **12**, 064054 (2019).
- [24] B. Göbel, A. Mook, J. Henk, I. Mertig, and O. A. Tretiakov, Magnetic bimerons as skyrmion analogues in in-plane magnets, *Phys. Rev. B* **99**, 060407(R) (2019).
- [25] L. Shen, J. Xia, X. Zhang, M. Ezawa, O. A. Tretiakov, X. Liu, G. Zhao, and Y. Zhou, Current-Induced Dynamics and Chaos of Antiferromagnetic Bimerons, *Phys. Rev. Lett.* **124**, 037202 (2020).
- [26] X. Li, L. Shen, Y. Bai, J. Wang, X. Zhang, J. Xia, M. Ezawa, O. A. Tretiakov, X. Xu, M. Mruczkiewicz, M. Krawczyk, Y. Xu, R. F. L. Evans, R. W. Chantrell, and Y. Zhou, Bimeron clusters in chiral antiferromagnets, *npj Comput. Mater.* **6**, 1 (2020).
- [27] F. Matsukura, Y. Tokura, and H. Ohno, Control of magnetism by electric fields, *Nat. Nanotechnol.* **10**, 209 (2015).
- [28] P. J. Hsu, A. Kubetzka, A. Finco, N. Romming, K. von Bergmann, and R. Wiesendanger, Electric-field-driven switching of individual magnetic skyrmions, *Nat. Nanotechnol.* **12**, 123 (2017).
- [29] M. Schott, A. Bernand-Mantel, L. Ranno, S. Pizzini, J. Vogel, H. Béa, C. Baraduc, S. Auffret, G. Gaudin, and D. Givord, The skyrmion switch: Turning magnetic skyrmion bubbles on and off with an electric field, *Nano Lett.* **17**, 3006 (2017).
- [30] P. Huang, M. Cantoni, A. Kruchkov, J. Rajeswari, A. Magrez, F. Carbone, and H. M. Rønnow, In situ electric field skyrmion creation in magnetoelectric Cu₂OSeO₃, *Nano Lett.* **18**, 5167 (2018).
- [31] C. Ma, X. Zhang, J. Xia, M. Ezawa, W. Jiang, T. Ono, S. N. Piramanayagam, A. Morisako, Y. Zhou, and X. Liu, Electric field-induced creation and directional motion of domain walls and skyrmion bubbles, *Nano Lett.* **19**, 353 (2018).

- [32] J. Liang, Q. Cui, and H. Yang, Electrically switchable Rashba-type Dzyaloshinskii-Moriya interaction and skyrmion in two-dimensional magnetoelectric multiferroics, *Phys. Rev. B* **102**, 220409(R) (2020).
- [33] C. Xu, P. Chen, H. Tan, Y. Yang, H. Xiang, and L. Bellaiche, Electric-Field Switching of Magnetic Topological Charge in Type-I Multiferroics, *Phys. Rev. Lett.* **125**, 037203 (2020).
- [34] W. Sun, W. Wang, H. Li, G. Zhang, D. Chen, J. Wang, and Z. Cheng, Controlling bimerons as skyrmion analogues by ferroelectric polarization in 2D van der Waals multiferroic heterostructures, *Nat. Commun.* **11**, 1 (2020).
- [35] L. Wang, Q. Feng, Y. Kim, R. Kim, K. H. Lee, S. D. Pollard, Y. J. Shin, H. Zhou, W. Peng, D. Lee, W. Meng, H. Yang, J. H. Han, M. Kim, Q. Lu, and T. W. Noh, Ferroelectrically tunable magnetic skyrmions in ultrathin oxide heterostructures, *Nat. Mater.* **17**, 1087 (2018).
- [36] G. Kresse and J. Hafner, *Ab initio* molecular dynamics for liquid metals, *Phys. Rev. B* **47**, 558 (1993).
- [37] G. Kresse and J. Furthmüller, Efficient iterative schemes for *ab initio* total-energy calculations using a plane-wave basis set, *Phys. Rev. B* **54**, 11169 (1996).
- [38] G. Kresse and J. Furthmüller, Efficiency of *ab-initio* total energy calculations for metals and semiconductors using a plane-wave basis set, *J. Comput. Mater. Sci.* **6**, 15 (1996).
- [39] Y. Wang and J. P. Perdew, Correlation hole of the spin-polarized electron gas, with exact small-wave-vector and high-density scaling, *Phys. Rev. B* **44**, 13298 (1991).
- [40] G. Kresse and D. Joubert, From ultrasoft pseudopotentials to the projector augmented-wave method, *Phys. Rev. B* **59**, 1758 (1999).
- [41] J. Li, J. Y. Ni, X. Y. Li, H.-J. Koo, M.-H. Whangbo, J. S. Feng, and H. J. Xiang, Intralayer ferromagnetism between $S = 5/2$ ions in MnBi_2Te_4 : Role of empty Bi p states, *Phys. Rev. B* **101**, 201408(R) (2020).
- [42] Y. Li, Z. Jiang, J. Li, S. Xu, and W. Duan, Magnetic anisotropy of the two-dimensional ferromagnetic insulator MnBi_2Te_4 , *Phys. Rev. B* **100**, 134438 (2019).
- [43] P. E. Blöchl, Projector augmented-wave method, *Phys. Rev. B* **50**, 17953 (1994).
- [44] S. Grimme, J. Antony, S. Ehrlich, and S. Krieg, A consistent and accurate *ab initio* parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu, *J. Chem. Phys.* **132**, 154104 (2010).
- [45] W. Ding, J. Zhu, Z. Wang, Y. Gao, D. Xiao, Y. Gu, Z. Zhang, and W. Zhu, Prediction of intrinsic two-dimensional ferroelectrics in In_2Se_3 and other $\text{III}_2\text{-VI}_3$ van der Waals materials, *Nat. Commun.* **8**, 14956 (2017).
- [46] J. Y. You, X. J. Dong, B. Gu, and G. Su, Electric field induced topological phase transition and large enhancements of spin-orbit coupling and Curie temperature in two-dimensional ferromagnetic semiconductors, *Phys. Rev. B* **103**, 104403 (2021).
- [47] J. Li, Y. Li, S. Du, Z. Wang, B. L. Gu, S. C. Zhang, K. He, W. Duan, and Y. Xu, Intrinsic magnetic topological insulators in van der Waals layered MnBi_2Te_4 -family materials, *Sci. Adv.* **5**, eaaw5685 (2019).
- [48] M. M. Otrokov, I. P. Rusinov, M. Blanco-Rey, M. Hoffmann, A. Y. Vyazovskaya, S. V. Eremeev, A. Ernst, P. M. Echenique, A. Arnau, and E. V. Chulkov, Unique Thickness-Dependent Properties of the Van Der Waals Interlayer Antiferromagnet MnBi_2Te_4 Films, *Phys. Rev. Lett.* **122**, 107202 (2019).
- [49] S. Yuan, W. F. Io, J. Mao, Y. Chen, X. Luo, and J. Hao, Enhanced piezoelectric response of layered $\text{In}_2\text{Se}_3/\text{MoS}_2$ nanosheet-based van der Waals heterostructures, *ACS Appl. Nano Mater.* **3**, 11979 (2020).
- [50] H. Yang, M. Xiao, Y. Cui, L. Pan, K. Zhao, and Z. Wei, Nonvolatile memristor based on heterostructure of 2D room-temperature ferroelectric $\alpha\text{-In}_2\text{Se}_3$ and WSe_2 , *Sci. China Inf. Sci.* **62**, 220404 (2019).
- [51] A. Y. Lu, H. Zhu, J. Xiao *et al.*, Janus monolayers of transition metal dichalcogenides, *Nat. Nanotechnol.* **12**, 744 (2017).
- [52] R. Sant, M. Gay, A. Marty *et al.*, Synthesis of epitaxial monolayer Janus SptSe , *npj 2D Mater. Appl.* **4**, 41 (2020).
- [53] Y. Zhou, D. Wu, Y. Zhu, Y. Cho, Q. He, X. Yang, K. Herrera, Z. Chu, Y. Han, M. C. Downer, H. Peng, and K. Lai, Out-of-plane piezoelectricity and ferroelectricity in layered $\alpha\text{-In}_2\text{Se}_3$ nanoflakes, *Nano. Lett.* **17**, 5508 (2017).
- [54] The optimized in-plane lattice constants of MBST and In_2Se_3 monolayers are 4.233 and 4.108 Å, respectively. Here, 1×1 In_2Se_3 monolayer is strained by 3.04% to match 1×1 MBST monolayer as shown in Fig. 6.
- [55] C. Gong, E. Kim, Y. Wang, G. Lee, and X. Zhang, Multiferroicity in atomic van der Waals heterostructures, *Nat. Commun.* **10**, 2657 (2019).
- [56] J. Xiao, H. Zhu, Y. Wang, W. Feng, Y. Hu, A. Dasgupta, Y. Han, Y. Wang, D. A. Muller, L. W. Martin, P. Hu, and X. Zhang, Intrinsic Two-Dimensional Ferroelectricity with Dipole Locking, *Phys. Rev. Lett.* **120**, 227601 (2018).
- [57] R. F. Evans, W. J. Fan, P. Chureemart, T. A. Ostler, M. O. Ellis, and R. W. Chantrell, Atomistic spin model simulations of magnetic nanomaterials, *J. Phys. Condens. Matter* **26**, 103202 (2014).
- [58] S. Z. Lin, A. Saxena, and C. D. Batista, Skyrmion fractionalization and merons in chiral magnets with easy-plane anisotropy, *Phys. Rev. B* **91**, 224407 (2015).
- [59] S. Rohart and A. Thiaville, Skyrmion confinement in ultrathin film nanostructures in the presence of Dzyaloshinskii-Moriya interaction, *Phys. Rev. B* **88**, 184422 (2013).
- [60] A. O. Leonov, T. I. Monchesky, N. Romming, A. Kubetzka, A. N. Boganov, and R. Wiesendanger, The properties of isolated chiral skyrmions in thin magnetic films, *New J. Phys.* **18**, 065003 (2016).
- [61] H. Jia, B. Zimmermann, and S. Blügel, First-principles investigation of chiral magnetic properties in multilayers: Rh/Co/Pt and Pd/Co/Pt , *Phys. Rev. B* **98**, 144427 (2018).
- [62] D. -S. Wang, R. Wu, and A. J. Freeman, First-principles theory of surface magnetocrystalline anisotropy and the diatomic-pair model, *Phys. Rev. B* **47**, 14932 (1993).
- [63] Q. Cui, J. Liang, B. Yang, Z. Wang, P. Li, P. Cui, and H. Yang, Giant enhancement of perpendicular magnetic anisotropy and induced quantum anomalous Hall effect in graphene/ NiI_2 heterostructures via tuning the van der Waals interlayer distance, *Phys. Rev. B* **101**, 214439 (2020).
- [64] B. Yang, X. Zhang, H. Yang, X. Han, and Y. Yan, Nonmetallic atoms induced magnetic anisotropy in monolayer chromium trihalides, *J. Phys. Chem. C* **123**, 691 (2019).

- [65] R. F. W. Bader, *Atoms in Molecules - A Quantum Theory* (Oxford University Press, Oxford, 1990).
- [66] G. Henkelman, A. Arnaldsson, and H. Jónsson, A fast and robust algorithm for Bader decomposition of charge density, *Comput. Mater. Sci.* **36**, 354 (2006).
- [67] E. Sanville, S. D. Kenny, R. Smith, and G. Henkelman, Improved grid-based algorithm for Bader charge allocation, *J. Comput. Chem.* **28**, 899 (2007).
- [68] W. Tang, E. Sanville, and G. Henkelman, A grid-based Bader analysis algorithm without lattice bias, *J. Phys. Condens. Matter* **21**, 084204 (2009).
- [69] F. Xue, Z. Wang, Y. Hou, L. Gu, and R. Wu, Control of magnetic properties of MnBi_2Te_4 using a van der Waals ferroelectric $\text{III}_2\text{-VI}_3$ film and biaxial strain, *Phys. Rev. B* **101**, 184426 (2020).
- [70] A. Togo and I. Tanaka, First principles phonon calculations in materials science, *Scr. Mater.* **108**, 1 (2015).
- [71] S. Nosé, A unified formulation of the constant temperature molecular dynamics methods, *J. Chem. Phys.* **81**, 511 (1984).
- [72] G. H. O. Daalderop, P. J. Kelly, and M. F. H. Schuurmans, First-principles calculation of the magnetocrystalline anisotropy energy of iron, cobalt, and nickel, *Phys. Rev. B* **41**, 11919 (1990).
- [73] A. Hallal, H. Yang, B. Dieny, and M. Chshiev, Anatomy of perpendicular magnetic anisotropy in Fe/MgO magnetic tunnel junctions: First-principles insight, *Phys. Rev. B* **88**, 184423 (2013).
- [74] Y. S. Hou and R. Q. Wu, Strongly Enhanced Gilbert Damping in 3d Transition-Metal Ferromagnet Monolayers in Contact with the Topological Insulator Bi_2Se_3 , *Phys. Rev. Appl.* **11**, 054032 (2019).