Dzyaloshinskii-Moriya interaction and magnetic skyrmions induced by curvature

Yonglong Ga,1,2 Qirui Cui,1 Jinghua Liang,1 Dongxing Yu,1 Yingmei Zhu1, Liming Wang1

and Hongxin Yang1,2,*

1Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, China

2Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China

Abstract

Realizing sizeable Dzyaloshinskii-Moriya interaction (DMI) in intrinsic two-dimensional (2D) magnets without any manipulation will greatly enrich potential application of spintronics devices. The simplest and most desirable situation should be 2D magnets with intrinsic DMI and intrinsic chiral spin textures. Here, we propose to realize DMI by designing periodic ripple structures with different curvatures in low-dimensional magnets and demonstrate the concept in both one-dimensional (1D) CrBr$_2$ and two-dimensional (2D) MnSe$_2$ magnets by using first-principles calculations. We find that DMIs in curved CrBr$_2$ and MnSe$_2$ can be efficiently controlled by varying the size of curvature $c$, where $c$ is defined as the ratio between the height $h$ and the length $l$ of curved structure. Moreover, we unveil that the dependence of first-principles calculated DMI on size of curvature $c$ can be well described by the three-site Fert-Lévy model. At last, we uncover that field-free magnetic skyrmions can be realized in curved MnSe$_2$ by using atomistic spin model simulations based on first-principles calculated magnetic parameters. The work will open a new avenue for inducing DMI and chiral spin textures in simple 2D magnets via curvature.
Introduction.—The antisymmetric exchange coupling, Dzyaloshinskii-Moriya interaction (DMI) [1,2], arising from inversion symmetry breaking and spin-orbit coupling (SOC), plays an essential role in stabilizing topological protected non-collinear chiral magnetic configurations [3-7]. The presence of DMI is confirmed in the noncentrosymmetric B20 material MnSi [8], in which magnetic skyrmions are first observed. Recently, many reports have further demonstrated DMI can be induced at interfaces of 3d/5d metal [9-11], metal/oxides [12-15], or light elements such as graphene [16,17], oxygen and hydrogen, etc. [18,19]. Notably, experimental progresses have demonstrated that magnetic order of two-dimensional (2D) magnets can persist down to monolayer, which is useful for the study of fundamental physics and for engineering spintronic devices [20-22]. As for non-collinear magnetism, several effective methods are proposed to obtain sizeable DMI in 2D materials, such as in 2D Janus magnets [23-25], intrinsic 2D multiferroics [26,27] and van der Waals Ferromagnet-Based heterostructures, etc. [28-32], in which inversion symmetry breaking plays a significant role.

In practice, it is inevitable to induce ripples for a 2D material either free standing or on a substrate as long as the size of 2D material is large enough, which has been reported in graphene [33,34], and MoS$_2$ [35,36], etc. If such a curved system is magnetic, it is highly possible to achieve DMI in pure 2D magnets. Here, we propose to realize sizeable DMI by designing periodic ripple structures with different curvatures in low-dimensional magnets. We demonstrate the concept in both 1D CrBr$_2$ and 2D MnSe$_2$ structures, in which DMI can be efficiently tuned by magnitude of curvatures. Moreover, we validate that the variation of curvature-dependent DMI can be well interpreted by Fert-Lévy mechanism. Interestingly, using atomistic spin model simulations based on first-principles calculated magnetic parameters, we find that field-free chiral magnetic skyrmions can be realized in 2D MnSe$_2$. These examples provide a new routine towards realizing chiral magnetic skyrmions in 2D magnets with long-range magnetic orderings, e.g., CrGeTe$_2$ [37], CrI$_3$ [38] and Fe$_3$GeTe$_2$ [39], etc.

Computational details.—Our first-principles calculations are performed within the
framework of density-functional theory (DFT) as implemented in the Vienna \textit{ab initio} simulation package (VASP) [40]. We choose the Predev-Burke-Ernzerhof (PBE) functionals of generalized gradient approximation (GGA) [41] to deal with the exchange-correlation energy. Besides, Projector augmented plane wave (PAW) method [42,43] is adopted to deal with the interaction between nuclear electrons and valence electrons. In order to correctly describe the $3d$ electrons, we employ the GGA+$U$ method [44] with an effective $U = 2$ eV and $U = 3$ eV for Mn and Cr as reported in previous studies [45,46], respectively. We build up a $7 \times 1 \times 1$ CrBr$_2$ and $6\sqrt{3} \times 1 \times 1$ MnSe$_2$ supercells to construct magnetic structures with different curvatures. The energy cutoff for plane wave expansion is set to 520 eV. $\Gamma$-centered $7 \times 1 \times 1$ for 1D CrBr$_2$ and $2 \times 18 \times 1$ for 2D MnSe$_2$ k-point meshes are adopted for the Brillouin zone (BZ) integration to relax supercell, respectively. All the structures are fully relaxed until the Hellmann-Feynman force acting on each atom and convergence criteria for energy are $0.001$ eV/Å and is $1 \times 10^{-7}$ eV,
1-Dimensional CrBr$_2$.—For simplification of analysis, we first investigate curvature-dependent DMI in 1D magnet CrBr$_2$. Similar to CuCl$_2$ and CuBr$_2$, bulk CrBr$_2$ has the monoclinic space group C12/m1 [47-49]. In each CrBr$_2$ layer, the ribbons made up of edge-sharing CrBr$_4$ squares along b-axis. It has been not experimentally reported whether one-dimensional CrBr$_2$ can be exfoliated from bulk so far. In our paper, we use the simple 1D structure to demonstrate the dependence of DMI on size of curvature $c$. The obtained 1D CrBr$_2$ has D$_{2h}$ crystal symmetry which preserved spatial inversion symmetry, as shown in Fig. 1(a). For achieving ripples, a $7 \times 1 \times 1$ supercell is constructed and the length $l$ of entire supercell is adjusted gradually in our calculations. Next, we keep two unit cells (located at the edges of...
supercell) fixed in z direction and relax all other atoms to obtain final curved structures (Figs. 1(b)-1(d)). In order to articulate our ideas, we present the schematic diagram (Fig. 5) consisting of sine and piecewise functions, in which the inversion symmetry with symmetry center $i$ of sine function (red dotted line) will be broken in the piecewise function (blue line). This piecewise structure is an approximation for the effect of substrate on curvature. With this premise, we study how magnetic parameters change under different size of curvature $c$ for a series of curved structures (Figs. 1(a)-1(d)). Firstly, we study curvature-dependent magnetic properties of 1D CrBr$_2$. Figure 6(a) shows the ferromagnetic (FM) and antiferromagnetic (AFM) configurations. It is found that the calculated energy difference between AFM and FM evidently decreases with increasing $c$, as shown in Fig. 6(c). Moreover, the spin ordering tends to be AFM when $c$ is larger than 15.4%. In Fig. 7(a), we also investigate absolute value of magnetic moment per Cr atoms as $c$ increases. In general, the magnetic moment variation of each magnetic atom is relatively weak and tends to decrease with increase of curvature.

Figure 1(g) shows the result of DMI for curved 1D CrBr$_2$ with different $c$. Obviously, DMI is strongly dependent on size of curvature $c$ and the calculated DMI strength increases with the increment of size of curvature $c$. Here, positive $D$ represents spin configuration of anticlockwise (ACW) chirality, negative one represents clockwise (CW) chirality. For total

FIG. 3. Summing over all M-X-M trimers based on Fert-Lévy model (Eq. (3)) in (a) 1D CrBr$_2$ and (b) 2D MnSe$_2$, respectively. Insets (i) and (j) represent structures with/without inversion symmetry, in which both $\theta$ ($\theta_i$, $i = 1,2$) and $a$ are bond angle of each M-X-M trimer and bond length between the nearest neighbor M atoms.
DMI $D$ of 1D CrBr$_2$ with different size of curvature $c$, its value varies from 0 to 1.0 meV/f.u.. Otherwise, In Figs. 1(e)-1(f) the total energy $E(q)$ and DMI energy $\Delta E_{DMI}(q)$ including SOC are presented as a function of spin spiral vector $q$ along $\bar{\Gamma} - \bar{X}$ direction in 2D BZ for $c = 30.5\%$ and $c = 34.1\%$, respectively. The extracted DMI are found to be 0.93 and 1.00 meV/f.u., respectively, suggesting that DMI of 1D CrBr$_2$ is directly associated with the existence of curvature. Considering the fact that 2D magnets, such as CrGeTe$_3$, CrI$_3$, Fe$_3$GeTe$_2$, and MnSe$_2$ [50] with long-range ferromagnetic orderings have been synthesized experimentally, we take the simplest one MnSe$_2$ as an example to further investigate the feasibility of tunable DMI in 2D magnets.

2-Dimensional MnSe$_2$.—It is recently reported that monolayer 2D MnSe$_2$ with D$_{3d}$ crystalsymmetry is a room temperature ferromagnet [50]. The optimized MnSe$_2$ structure is presented in Fig. 2(a). It can be seen that each Mn atom is surrounded by six non-magnetic Se atoms that make up an octahedron. Similar to 1D CrBr$_2$, we construct a $6\sqrt{3} \times 1 \times 1$ supercell to obtain ripple structures, in which Mn atoms of three unit cells (located at initial and terminal of the supercell) are fixed in the $z$ direction and the other atoms are relaxed (Figs. 2(b)-2(d)). To understand magnetism associated with curved structures, we construct FM and AFM states in Fig. 6(b). It is found that the energy difference between AFM and FM suddenly decreases and then keeps increasing as the $c$ gradually increases (Fig. 6d). We also compute the magnetic moment of Mn atoms from Mn1 to Mn12, as shown in Fig. 7d. Compared to 1D CrBr$_2$, there is a distinct difference that the magnetic moments of Mn atoms located at peaks and troughs

FIG. 4. (a) Atomistic spin model simulated spin configurations for a curved MnSe$_2$ with size of curvature $c$ of 31.6\%, (b) zoom in of an isolated skyrmion profile.
have large variation due to the locally severe structural deformation, while others don’t change much.

In Fig. 2(i), we display the calculated results of DMI for 2D MnSe$_2$ with curvature. In 2D MnSe$_2$ structure, the global tendency of DMI is increasing as $c$ gradually increases, similar as that of 1D CrBr$_2$. In 2D MnSe$_2$, there are two DMI vectors when spin spiral vector $q$ propagates along directions of $\bar{T} − \bar{X}$ and $\bar{T} − \bar{Y}$ in the 2D BZ, in which the calculated values are in the range of 0-0.88 meV/f.u. and 0-26.29 meV/f.u., respectively. Obviously, compared to that $\bar{T} − \bar{Y}$ direction, the DMI strength along with $\bar{T} − \bar{X}$ direction is almost negligible. Thus, we focus on the DMI vector in the $\bar{T} − \bar{Y}$ direction below. The energy dispersion $E(q)$ and the DMI energy $\Delta E_{DMI}(q)$ associated with spin spiral vector $q$ for $c = 28.4\%$ and $c = 31.6\%$ are shown in Figs. 2(e)-2(h). The extracted DMI parameters $D$ by taking the slope of dispersion in the vicinity of ground state are 23.4 and 26.3 meV/f.u., respectively. For convenience in comparison, we also convert the total DMI per supercell to per magnetic atom as the effective DMI $D_{eff}$ (Fig. 1(g) and Fig. 2(i)). The $D_{eff}$, 0.21 and 0.24 meV/atom in 2D MnSe$_2$ induced by curvature with $c = 28.4\%$ and $c = 31.6\%$, which are comparable to those of Co/Ru(0001) [51] or Co/graphene interfaces [16,17], are highly possible to stabilize magnetic chiral spin textures.
in 2D magnets with long-range magnetic orderings.

Discussion.—To further understand the variation of curvature-induced DMI above, the DMI effect of curved structure can be modelled as the sum of each M-X-M trimer, where M and X represent the magnetic and nonmagnetic atoms, respectively. Based on three-site Fert-Lévy model [52], the hoping of spins between two magnetic atoms \((i\) and \(j\)) can induce DMI through a nonmagnetic atom \((l)\) with SOC. The theoretically formulism of antisymmetric DMI can be expressed as:

\[
\vec{D}_{ijl}(\vec{R}_{li}, \vec{R}_{lj}, \vec{R}_{lj}) = -V_{soc} \frac{(\vec{R}_{li} \cdot \vec{R}_{lj})(\vec{R}_{li} \times \vec{R}_{lj})}{|\vec{R}_{li}| |\vec{R}_{lj}| |\vec{R}_{lj}|},
\]

where \(\vec{R}_{li}\), \(\vec{R}_{lj}\), and \(\vec{R}_{lj}\) are the corresponding distance vectors between atoms in the three-site model. \(V_{soc}\) is a SOC-governed material parameter, its expressions can be written as \([135\pi\lambda_d\Gamma^2(\sin(Z_d\pi/10))/(32k_F^2E_F^2)]\), in which \(\lambda_d\), \(k_F\), \(E_F\), and \(Z_d\) are the SOC constant, Fermi vector, Fermi energy and the number of \(d\) electrons, respectively. \(\Gamma\) is the interaction parameter between the localized spins and the spins of conduction electrons. In a curved magnet, the total DMI can be obtained by summing over all M-X-M trimers and we can further simplify the above Eq. (1) as:

FIG. 6. (a) and (b) are FM and AFM spin configurations of CrBr\(_2\) and MnSe\(_2\). (c) and (d) are the energy differences between AFM and FM for CrBr\(_2\) and MnSe\(_2\) under different size of curvature c, respectively.
\[ \vec{D} = -V_{soc} \sum_{i=1}^{n} \frac{2 \sin(2\theta_i) \sin^2(\frac{1}{2}\theta_i)}{a_i^3}, \]  

(2)

where \( i \) is the index of summation, \( \theta \) and \( a_i \) represent bond angle of each M-X-M trimer and bond length between the nearest neighbor M atoms in whole curved structure, respectively. In order to describe curvature dependence of DMI in Fert-Lévy model, we assume that \( V_{soc} \) is a constant for a given system. Then we can provide a quantitative understanding for curvature-induced DMI based on Eq. (2). Insets \( i \) in Figs. 3(a) and 3(b) present the schematic structures with inversion symmetry, which will lead to the offset of total DMI. However, the inversion symmetry breaking arising from curvature, will induce an uncompensated DMI in the noncentrosymmetric three-site interaction (insets \( j \) in Figs. 3(a) and 3(b)). For 1D CrBr\(_2\), in order to build up the relationship between curvature and DMI, the calculated \( \theta_i \) and \( a_i \) of each Cr-Br-Cr trimer are brought into the Eq. (2). The result shows that \( D/2V_{soc} \) increases with increasing size of curvature \( c \), as is shown in Fig. 3(a). Similar to that of 1D CrBr\(_2\), we can determine the \( \theta_i \) and \( a_i \) of each Mn-Se-Mn trimer in curved 2D MnSe\(_2\) and one can see that \( D/2V_{soc} \) increases as size of curvature \( c \) increases when \( \theta_i \) and \( a_i \) are brought into Eq. (2) as
shown in Fig. 3(b). These results are in line with given results from first-principles calculations. More interestingly, notice that the whole curve structure is composed of several similar M-X-M trimers in our studied system, and we find that $\theta_i$ and $a_i$ of each M-X-M trimer present a small tendency of change as a whole in curved structure with increasing size of curvature $c$. Therefore, we propose a simple model, in which average bond angle $\bar{\theta}$ of Cr-Br-Cr (Mn-Se-Mn) triples and average bond length $\bar{a}$ of the nearest neighbor Cr (Mn) atoms in whole curve magnet are described as key parameters to further examine the relationship between curvature and DMI. We plot the dependences of $\bar{\theta}$ and $\bar{a}$ with respect to size of curvature $c$ as shown in Figs. 8(a) and 8(b). We can further simplify Eq. (2) as:

$$\bar{D} = -V_{soc} \frac{2\sin(2\bar{\theta})\sin^2(\frac{\bar{\theta}}{2})}{\bar{a}^3}. \quad (3)$$

Overall, one can see that $D/2V_{soc}$ increases for both 1D CrBr$_2$ and 2D MnSe$_2$ with increasing size of curvature $c$ when $\bar{\theta}$ and $\bar{a}$ are brought into Eq. (3), as shown in Figs. 9(a) and 9(b). This is also consistent with given results from first-principles calculations. Thus, our findings based on both first-principles calculations and Fert-Lévy model demonstrate that curvature is an effective way to realized DMI in 1D CrBr$_2$ and 2D MnSe$_2$.

**Realization of skyrmions in 2D MnSe$_2$**—To explore the formation of such chiral spin textures in curved 2D MnSe$_2$, we apply atomistic spin model simulations [53] for the magnetization dynamics simulation based on magnetic parameters with size of curvature $c$ of 31.6% from first-principles calculations. To obtain final spin configurations, the atomistic
Landau-Lifshitz-Gilbert (LLG) equation is given:

$$\frac{\partial \mathbf{s}_i}{\partial t} = -\frac{\gamma}{(1+\lambda^2)} \left[ \mathbf{s}_i \times \mathbf{H}_{eff} + \lambda \mathbf{s}_i \times (\mathbf{s}_i \times \mathbf{H}_{eff}^i) \right],$$

(4)

Here $\gamma$ is the gyromagnetic ratio, $\lambda$ is the Gilbert damping constant and $\mathbf{s}_i$ is the unit vector, which is defined as $\frac{\mu_i}{|\mu_i|}$, where $\mu_i$ represents the magnetic moment of $i$th magnetic atom. $\mathbf{H}_{eff}^i$ is the effective magnetic field on each $\mathbf{s}_i$ and can be obtained by following equation:

$$\mathbf{H}_{eff}^i = -\frac{1}{\mu_i} \frac{\partial \mathbf{H}}{\partial \mathbf{s}_i},$$

in which $\mathbf{H}$ represents the total spin Hamiltonian. In simulations, we set a 60-nm width square with periodic boundary condition from an initial random state to obtain final spin textures. These parameters including effective exchange constant $J_{eff}$, effective DMI parameter $D_{eff}$, magnetic anisotropy $K$, dipole-dipole interaction $K_{Dip}$ and magnetic moment of Mn atoms are 14.94 meV, 0.24 meV, 0.17 meV, -0.176 meV and 3.58 $\mu_B$, respectively, and the methods to calculate $J_{eff}$, $K$, $D_{eff}$ and $K_{Dip}$ are described in the supplementary material. In addition, we apply the theoretical formula $\tilde{R} = \pi D \sqrt{\frac{A}{16AR^2 - \pi^2D^2K}}$ [54] to predict the radius $\tilde{R}$ of skyrmion, which is close to 20 nm. We can find that the size of chiral structure is much larger than the ripple wavelength. Thus, we used the flat crystal structure to simulate our chiral magnetic structures in the atomic magnetic simulations. In simulations, Figure 4 shows the spin

FIG. 9. Plotted theoretical curve and calculated results by using Fert-Lévy model (Eq. (2)). (a) and (b) indicate that the $D/2\mathcal{V}_{soc}$ as a function of average bond angle $\bar{\theta}$ and average bond length $\bar{a}$ in 1D CrBr$_2$ and 2D MnSe$_2$, respectively.
textures of curved MnSe$_2$ in $c = 31.6\%$. Interestingly, magnetic skyrmion spin texture with diameter of around 20 nm emerges under zero magnetic field as shown in Figs. 4(a) and 4(b). The calculated topological charge $Q$ is -1 for skyrmion by using the formula $Q = \frac{1}{4\pi} \int S \cdot (\partial_x S \times \partial_y S) dx dy$, where $S$ represents the unit vector, $x$ and $y$ are the coordinates.

Conclusion.—In summary, we obtain sizeable DMI in both 1D CrBr$_2$ and 2D MnSe$_2$ by using ripple structures with different curvatures via using first-principles calculations. We unveil that quite large DMI can be induced in curved structure, in which breaking of spatial inversion plays an important role. In addition, with the help of three-site Fert-Lévy model, the DMI dependence on the magnitude of curvature is successfully interpreted. Via atomistic spin model simulations, we further uncover that magnetic skyrmions can be induced without external field in curved 2D MnSe$_2$. Considering the experimental progress in 2D magnets, such as CrGeTe$_3$, CrI$_3$ and Fe$_2$GeTe$_2$, etc., our results open up new opportunities for development of spintronics.

Note added. After submission of this work, one article appeared on complimentary studies discussing curvature-induced magnetic parameters in curved magnets CrI$_3$ [55].

Acknowledgments. This work was supported by National Natural Science Foundation of China (Grants No. 11874059 and No. 12174405), Key Research Program of Frontier Sciences, CAS (Grant No. ZDBS-LY-7021), Ningbo Key Scientific and Technological Project (Grant No. 2021000215), Pioneer and Leading Goose R&D Program of Zhejiang Province under Grant No. 2022C01053, Zhejiang Provincial Natural Science Foundation (Grant No. LR19A040002), and Beijing National Laboratory for Condensed Matter Physics (Grant No. 2021000123).

APPENDIX: Calculations of magnetic parameters

1. Dzyaloshinskii-Moriya interaction (DMI) $D$

DMI is beneficial to form the non-collinear magnetic textures, in which the neighboring spins tends to align perpendicular to each other. The energy term can be described as:

$$E_{DM} = \sum_{i,j} D_{ij} \cdot (S_i \times S_j),$$

(2)
where $D_{ij}$ represents the DMI vector, $S_i$ and $S_j$ are the spins of nearest neighbor magnetic atoms. We adopt the qSO method based on generalized Bloch theorem and treat SOC within the first-order perturbation theory [26,56,57], in which the total energy $E(q)$ as the function of spin-spiral vector $q$ is calculated along the high symmetry direction of $\bar{T} - \bar{X}$ and $\bar{T} - \bar{Y}$ in our studied system. To extract the DMI parameter, a $\Gamma$-centered $7 \times 1 \times 1$ for 1D CrBr$_2$ and $2 \times 18 \times 1$ k mesh for 2D MnSe$_2$ are considered to compute the spin spiral energy $E(q)$ in the interval of $q$ from $-0.1 \frac{2\pi}{a}$ to $0.1 \frac{2\pi}{a}$ by a self-consistent way. The nonlinear spin spiral structure $m = (\sin(q \cdot r_i), 0, \cos(q \cdot r_i))$ rotates along the axis of spin spiral parallel the y axis, where $q$ represents the spin spiral vector and $r_i$ is the location of $i$th atom (When the rotation axis is x axis, the nonlinear spin spiral structure $m = (0, \sin(q \cdot r_i), \cos(q \cdot r_i))$). Then, DMI energy can be written as:

$$E_{DMI}(q) = \sum_{i,j} D_y \sin(q \cdot r_{ij}),$$

(3)

where $D_y$ represents the DM vector perpendicular to the propagation direction of spin spirals, and $r_{ij}$ is the unit vector between sites $i$ and $j$. Finally, the DMI parameter can be obtained by taking the slope of $\Delta E_{DMI}(q)$ with respect to spin spiral vector $q$, where $\Delta E_{DMI}(q)$ is the energy difference between $E(q)$ and $E(-q)$. Furthermore, final form of DMI energy can be simplified as:

$$\Delta E_{DMI}(q) = E(q) - E(-q) = 2\sum_{i,j} D_y \sin(q \cdot r_{ij}),$$

(4)

For small $q = (q,0,0)$ vector, we have the relationships:

$$\Delta E_{DM}(q) = 2q \sum_{i} D_y r_{ij,x} = Dq,$$

(5)

where

$$D = 2\sum_{i,j} D_y r_{ij,x}.$$  

(6)

Here, the direction of DM vector depends on the propagation direction of spin spiral $q$ of the whole Brillouin zone.

2. Effective DMI value $D_{eff}$

In our calculations, we convert the total DMI $D$ per supercell to per magnetic atom as the effective $D_{eff}$. For 1D CrBr$_2$, $D = 14\pi D_{eff}$; For 2D MnSe$_2$, $D = 36\pi D_{eff}$. 

13
3. Magnetocrystalline anisotropy energy $K$

$K$ is defined as the energy difference between in-plane (100) and out-of-plane (001) magnetized axis:

$$K = E_{100} - E_{001} \quad (7).$$

4. Effective exchange coupling constant $J_{\text{eff}}$

As is shown in Fig. 6(b), we set different magnetic configurations FM and AFM for MnSe$_2$. We consider the Heisenberg model on a hexagonal lattice,

$$H = \sum_{ij} J_{\text{eff}} \vec{S}_i \cdot \vec{S}_j \quad (8)$$

Then effective exchange coupling constant can be solved by following formula:

$$E_{\text{FM}} = -\frac{1}{2} \times 12 \times (6J_{\text{eff}}) + E_{\text{other}}, \quad (9)$$

$$E_{\text{AFM}} = -\frac{1}{2} \times 12 \times (-2J_{\text{eff}}) + E_{\text{other}}, \quad (10)$$

$$J_{\text{eff}} = \frac{E_{\text{AFM}}-E_{\text{FM}}}{48}, \quad (11)$$

where the positive or negative value corresponds to FM or AFM coupling.

5. Dipole-dipole contribution to $K_{\text{Dip}}$

The dipole-dipole interaction contribution to $K$ is the sum of all magnetostatic dipole-dipole interactions up to infinity [58,59] and is calculated by following formula:

$$E_{\text{dip}} = -\frac{1}{2} \left(\frac{\mu_0}{4\pi V_{\text{cell}}}\right) \sum_{i,j=1}^{N} \frac{(m_i \cdot m_j) r_{ij}^{-3} (m_i \cdot r_{ij})(m_j \cdot r_{ij})}{r_{ij}^6}, \quad (12)$$

where $m_i$ and $m_j$ represent the unit vector of magnetization at position $r_i$ and $r_j$, $r_{ij}$ is the unit vector between site $i$ and $j$. In calculations, we choose a $150 \times 150 \times 1$ supercell to obtain the dipole-dipole interaction between two magnetic atoms in 2D MnSe$_2$. 


References

[1] I. Dzyaloshinsky, A Thermodynamic Theory of “Weak” Ferromagnetism of Antiferromagnetics, J. Chem. Phys. Solids 4, 241 (1958).

[2] T. Moriya, Anisotropic Superexchange Interaction and Weak Ferromagnetism, Phys. Rev. 120, 91 (1960).

[3] A. Thiaville, S. Rohart, É. Jué, V. Cros, and A. Fert, Dynamics of Dzyaloshinskii Domain Walls in Ultrathin Magnetic Films, Europhys. Lett. 100, 57002 (2012).

[4] 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 (2019).

[5] H. Jani, J. C. Lin, J. Chen, J. Harrison, F. Maccherozzi, J. Schad, S. Prakash, C. B. Eom, A. Ariando, T. Venkatesan, and P. G. Radaelli, Antiferromagnetic Half-Skyrmions and Bimerons at Room Temperature, Nature 590, 74 (2021).

[6] A. N. Bogdanov and D. A. Yablonsky, Thermodynamically Stable Vortexes in Magnetically Ordered Crystals-Mixed State of Magnetics, Sov. Phys. JETP 95, 178 (1989).

[7] X. Z. Yu, N. Kanazawa, Y. Onose, K. Kimoto, W. Z. Zhang, S. Ishiwata, Y. Matsui, and Y. Tokura, Near Room-Temperature Formation of a Skyrmion Crystal in Thin-Films of the Helimagnet FeGe, Nat. Mater. 10, 106 (2011).

[8] S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Skyrmion Lattice in a Chiral Magnet, Science, 323, 915 (2009).

[9] S. Heinze, K. von Bergmann, M. Menzel, J. Brede, A. Kubetzka, R. Wiesendanger, G. Bihlmayer, and S. Blügel, Spontaneous Atomic-Scale Magnetic Skyrmion Lattice in Two Dimensions, Nat. Phys. 7, 713 (2011).

[10] O. Boule, J. Vogel, H. Yang, S. Pizzini, D. de Souza Chaves, A. Locatelli, T. O. Mentes, A. Sala, L. D. Buda-Prejbeanu, O. Klein, M. Belmeguenai, Y. Roussigne, A. Stashkevich, S. M. Cherif, L. Aballe, M. Foerster, M. Chshiev, S. Auffret, I. M. Miron, and G. Gaudin, Room-Temperature Chiral Magnetic Skyrmions in Ultrathin Magnetic Nanostructures, Nat. Nanotechnol. 11, 449 (2016).

[11] W. Jiang, P. Upadhyaya, W. Zhang, G. Yu, M. B. Jungfleisch; F. Y. Fradin, J. E. Pearson, Y. Tserkovnyak, K. L. Wang, O. Heinonen, S. G. E. t. Velthuis, and A. Hoffmann, Blowing Magnetic Skyrmion Bubbles, Science, 349, 283 (2015).

[12] M. Schott, A. Bernard-Mantel, L. Ranno, S. Pizzini, J. Vogel, H. Bea, 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).

[13] H. Yang, O. Boule, 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).

[14] D. d. S. Chaves, F. Ajejas, V. Křižáková, J. Vogel, and S. Pizzini, Oxidation Dependence of the Dzyaloshinskii-Moriya Interaction in Pt/Co/MO_x trilayers (M=Al or Gd), Phys. Rev. B 99, 144404 (2019).

[15] H. T. Nembach, E. Jué, E. R. Evarts, and J. M. Shaw, Correlation between Dzyaloshinskii-Moriya Interaction and Orbital Angular Momentum at an Oxide-Ferromagnet Interface, Phys. Rev. B 101, 020409(R) (2020).

[16] H. Yang, G. Chen, A. A. C. Cotta, A. T. N’Diaye, S. A. Nikolaev, E. A. Soares, W. A. A.
Macedo, K. Liu, A. K. Schmid, A. Fert, and M. Chshiev, Significant Dzyaloshinskii-Moriya Interaction at Graphene-Ferromagnet Interfaces Due to the Rashba Effect, Nat. Mater. 17, 605 (2018).

[17] F. Ajejas, A. Gudin, R. Guerrero, A. Anadon Barcelona, J. M. Diez, L. de Melo Costa, P. Olleros, M. A. Nino, S. Pizzini, J. Vogel, M. Valvidares, P. Gargiani, M. Cabero, M. Varela, J. Camarero, R. Miranda, and P. Perna, Unraveling Dzyaloshinskii-Moriya Interaction and Chiral Nature of Graphene/Cobalt Interface, Nano Lett. 18, 5364 (2018).

[18] G. Chen, A. Mascaraque, H. Jia, B. Zimmermann, M. Robertson, R. L. Conte, M. Hoffmann, M. A. G. Barrio, H. Ding, R. Wiesendanger, E. G. Michel, S. Blügel, A. K. Schmid, and K. Liu, Large Dzyaloshinskii-Moriya Interaction Induced by Chemisorbed Oxygen on a Ferromagnet Surface, Sci. Adv. 6, eaba4924 (2020).

[19] G. Chen, M. Robertson, M. Hoffmann, C. Ophus, A. L. Fernandes Cauduro, R. Lo Conte, H. Ding, R. Wiesendanger, S. Blügel, A. K. Schmid, and K. Liu, Observation of Hydrogen-Induced Dzyaloshinskii-Moriya Interaction and Reversible Switching of Magnetic Chirality, Phys. Rev. X 11, 021015 (2021).

[20] D. Zhong, K. L. Seyler, X. Linpeng, R. Cheng, N. Sivadas, B. Huang, E. Schmidgall, T. Taniguchi, K. Watanabe, M. A. McGuire, W. Yao, D. Xiao, K.-M. C. Fu, and X. Xu, Van Der Waals Engineering of Ferromagnetic Semiconductor Heterostructures for Spin and Valleytronics, Sci. Adv. 3, e1603113 (2017).

[21] K. S. Burch, D. Mandrus, and J. G. Park, Magnetism in Two-Dimensional Van Der Waals Materials, Nature 563, 47 (2018).

[22] M. Gibertini, M. Koperski, A. F. Morpurgo, and K. S. Novoselov, Magnetic 2d Materials and Heterostructures, Nat. Nanotechnol. 14, 408 (2019).

[23] 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).

[24] 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).

[25] 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).

[26] J. Liang, Q. Cui, and H. Yang, Electrically Switchable Rashba-Type Dzyaloshinskii-Moriya Interaction and Skyrmion in Two-Dimensional Magnetolectric Multiferroics, Phys. Rev. B 102, 220409(R) (2020).

[27] 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).

[28] 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, 5930 (2020).

[29] C.-K. Li, X.-P. Yao, and G. Chen, Writing and Deleting Skyrmions with Electric Fields in a Multiferroic Heterostructure, Phys. Rev. Research 3, L012026 (2021).
[30] Y. Wu, S. Zhang, J. Zhang, W. Wang, Y. L. Zhu, J. Hu, G. Yin, K. Wong, C. Fang, C. Wan, X. Han, Q. Shao, T. Taniguchi, K. Watanabe, J. Zang, Z. Mao, X. Zhang, and K. L. Wang, Neel-Type Skyrmion in WTe$_2$/Fe$_3$GeTe$_2$ Van Der Waals Heterostructure, Nat. Commun. 11, 3860 (2020).

[31] T.-E. Park, L. Peng, J. Liang, A. Hallal, F. S. Yasin, X. Zhang, K. M. Song, S. J. Kim, K. Kim, M. Weigand, G. Schütz, S. Finizio, J. Raabe, K. Garcia, J. Xia, Y. Zhou, M. Ezawa, X. Liu, J. Chang, H. C. Koo, Y. D. Kim, M. Chshiev, A. Fert, H. Yang, X. Yu, and S. Woo, Néel-Type Skyrmions and Their Current-Induced Motion in Van Der Waals Ferromagnet-Based Heterostructures, Phys. Rev. B 103, 104410 (2021).

[32] M. Yang, Q. Li, R. V. Chopdekar, R. Dhall, J. Turner, J. D. Carlström, C. Ophus, C. Klewe, P. Shafer, A. T. N’Diaye, J. W. Choi, G. Chen, Y. Z. Wu, C. Hwang, F. Wang, and Z. Q. Qiu, Creation of Skyrmions in Van Der Waals Ferromagnet Fe$_3$GeTe$_2$ on (Co/Pd)$_n$ Superlattice, Sci. Adv. 6, eabb5157 (2020).

[33] A. Fasolino, J. H. Los, and M. I. Katsnelson, Intrinsic Ripples in Graphene, Nat. Mater. 6, 858 (2007).

[34] J. C. Meyer, A. K. Geim, M. I. Katsnelson, K. S. Novoselov, T. J. Booth, and S. Roth, The Structure of Suspended Graphene Sheets, Nature 446, 60 (2007).

[35] J. Brivio, D. T. Alexander, and A. Kis, Ripples and Layers in Ultrathin MoS$_2$ Membranes, Nano Lett. 11, 5148 (2011).

[36] P. Miro, M. Ghorbani-Asl, and T. Heine, Spontaneous Ripple Formation in Mos(2) Monolayers: Electronic Structure and Transport Effects, Adv. Mater. 25, 5473 (2013).

[37] C. Gong, L. Li, Z. Li, H. Ji, A. Stern, Y. Xia, T. Cao, W. Bao, C. Wang, Y. Wang, Z. Q. Qiu, R. J. Cava, S. G. Louie, J. Xia, and X. Zhang, Discovery of Intrinsic Ferromagnetism in Two-Dimensional Van Der Waals Crystals, Nature 546, 265 (2017).

[38] B. Huang, G. Clark, E. Navarro-Moratalla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, W. Yao, D. Xiao, P. Jarillo-Herrero, and X. Xu, Layer-Dependent Ferromagnetism in a Van Der Waals Crystal Down to the Monolayer Limit, Nature 546, 270 (2017).

[39] Z. Fei, B. Huang, P. Malinowski, W. Wang, T. Song, J. Sanchez, W. Yao, D. Xiao, X. Zhu, A. F. May, W. Wu, D. H. Cobden, J. H. Chu, and X. Xu, Two-Dimensional Itinerant Ferromagnetism in Atomically Thin Fe$_3$GeTe$_2$, Nat. Mater. 17, 778 (2018).

[40] 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).

[41] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized Gradient Approximation Made Simple, Phys. Rev. Lett. 77, 3865 (1996).

[42] G. Kresse and J. Hafner, Ab Initio Molecular Dynamics for Liquid Metals, Phys. Rev. B 47, 558 (1993).

[43] G. Kresse and J. Furthmüller, Efficiency of Ab-Initio Total Energy Calculations for Metals and Semiconductors Using a Plane-Wave Basis Set, Comput. Mater. Sci. 6, 15 (1996).

[44] V. I. Anisimov, F. Aryasetiawan, and A. I. Lichtenstein, First-Principles Calculations of the Electronic Structure and Spectra of Strongly Correlated Systems: Thelda+Umethod, J. Phys.: Condens. Matter 9, 767 (1997).

[45] X. Sui, T. Hu, J. Wang, B.-L. Gu, W. Duan, and M.-s. Miao, Voltage-Controllable
Colossal Magnetocrystalline Anisotropy in Single-Layer Transition Metal Dichalcogenides, Phys. Rev. B 96, 041410(R) (2017).

[46] B. Yang, X. Zhang, H. Yang, X. Han, and Y. Yan, Strain Controlling Transport Properties of Heterostructure Composed of Monolayer CrI\(_3\), Appl. Phys. Lett. 114, 192405 (2019).

[47] L. Zhao, T. L. Hung, C. C. Li, Y. Y. Chen, M. K. Wu, R. K. Kremer, M. G. Banks, A. Simon, M. H. Whangbo, C. Lee, J. S. Kim, I. Kim, and K. H. Kim, Cubr2--a New Multiferroic Material with High Critical Temperature, Adv. Mater. 24, 2469 (2012).

[48] C. Wang, D. Yu, X. Liu, R. Chen, X. Du, B. Hu, L. Wang, K. Iida, K. Kamazawa, S. Wakimoto, J. Feng, N. Wang, and Y. Li, Observation of Magnetoelastic Effects in a Quasi-One-Dimensional Spiral Magnet, Phys. Rev. B 96, 085111 (2017).

[49] M. G. Banks, R. K. Kremer, C. Hoch, A. Simon, B. Ouladdiaf, J.-M. Broto, H. Rakoto, C. Lee, and M.-H. Whangbo, Magnetic ordering in the frustrated Heisenberg chain system cupric chloride CuCl2 Phys. Rev. B 80, 024404 (2009)

[50] D. J. O'Hara, T. Zhu, A. H. Trout, A. S. Ahmed, Y. K. Luo, C. H. Lee, M. R. Brenner, S. Rajan, J. A. Gupta, D. W. McComb, and R. K. Kawakami, Room Temperature Intrinsic Ferromagnetism in Epitaxial Manganese Selenide Films in the Monolayer Limit, Nano Lett. 18, 3125 (2018).

[51] M. Herve, B. Dupe, R. Lopes, M. Bottcher, M. D. Martins, T. Balashov, L. Gerhard, J. Sinova, and W. Wulfhekel, Stabilizing Spin Spirals and Isolated Skyrmions at Low Magnetic Field Exploiting Vanishing Magnetic Anisotropy, Nat. Commun. 9, 1015 (2018).

[52] A. Fert and P. M. Levy, Role of Anisotropic Exchange Interactions in Determining the Properties of Spin-Glasses, Phys. Rev. Lett. 44, 1538 (1980).

[53] 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).

[54] X.S. Wang, H.Y. Yuan, and X.R. Wang, A theory on skyrmion size. Commun. Phys. 1, 31 (2018).

[55] A. Edström, D. Amoroso, S. Picozzi, P. Barone, and M. Stengel, Curved Magnetism in CrI\(_3\). Phys. Rev. Lett. 128, 177202 (2022)

[56] L. M. Sandratskii, Insight into the Dzyaloshinskii-Moriya Interaction through First-Principles Study of Chiral Magnetic Structures, Phys. Rev. B 96, 024450 (2017).

[57] M. Heide, G. Bihlmayer, and S. Blügel, Describing Dzyaloshinskii–Moriya Spirals from First Principles, Phys. B (Amsterdam) 404, 2678 (2009).

[58] G. H. Daalderop, P. J. Kelly, and M. F. Schuurmans, First-Principles Calculation of the Magnetocrystalline Anisotropy Energy of Iron, Cobalt, and Nickel, Phys. Rev. B 41, 11919 (1990).

[59] A. Hallal, H. X. 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).