Anisotropic Dzyaloshinskii-Moriya interaction protected by $D_{2d}$ crystal symmetry in two-dimensional ternary compounds

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Magnetic skyrmions, topologically protected chiral spin swirling quasiparticles, have attracted great attention in fundamental physics and applications. Recently, the discovery of two-dimensional (2D) van der Waals (vdW) magnets has aroused great interest due to their appealing physical properties. Moreover, both experimental and theoretical works have revealed that isotropic Dzyaloshinskii–Moriya interaction (DMI) can be achieved in 2D magnets or ferromagnet-based heterostructures. However, 2D magnets with anisotropic DMI haven’t been reported yet. Here, via using first-principles calculations, we unveil that anisotropic DMI protected by $D_{2d}$ crystal symmetry can exist in 2D ternary compounds $M$Cu$X_2$ ($M$: 3d transition metal (TM), $X$: group VIA). Interestingly, by using micromagnetic simulations, we demonstrate that ferromagnetic (FM) antiskyrmions, FM bimerons, antiferromagnetic (AFM) antiskyrmions, and AFM bimerons can be realized in the $M$Cu$X_2$ family. Our discovery opens up a avenue to creating antiskyrmions and bimerons with anisotropic DMI protected by $D_{2d}$ crystal symmetry in 2D magnets.

INTRODUCTION

Topological non-trivial magnetic structures such as chiral domain walls1, merons2, bimerons3,4, and skyrmions5,6 have attracted great research interest due to their rich physical properties and widespread application prospects in spintronic devices. Among these spin textures, magnetic skyrmions have been extensively studied due to their small size, low energy consumption, and low driving current density7,8. Magnetic skyrmions with non-collinear spin configurations can be induced by antisymmetric exchange coupling-Dzyaloshinskii-Moriya interaction (DMI) in structures with inversion symmetry breaking and spin-orbit coupling (SOC)9–10. The antisymmetric exchange interaction can be written as $E_{DMI} = D_{ij} (S_i \times S_j)$, where $D$ is the DMI vector and $S_i$ and $S_j$ represent the spins of sites $i$ and $j$. Notably, magnetic skyrmions have been observed in non-centrosymmetric B20 bulk materials such as MnSi, FeGe, and FeCoSi6,11 and interfacial systems of multilayers such as Ir(111)/Fe, Ta/CoFeB, and Pt/Co15–21 with C$_{2h}$ crystal symmetry. On the other hand, anisotropic DMI are reported in ultrathin epitaxial Au/Co/W(110) with C$_{2v}$ crystal symmetry22, and antiskyrmions with anisotropic DMI are reported in acentric tetragonal Heusler compounds with $D_{2d}$ crystal symmetry23,24 and non-centrosymmetric tetragonal structure with S$_8$ crystal symmetry25. In parallel with the development of the hot study of skyrmions in these traditional bulk and multilayer thin films, 2D magnets, e.g., Fe$_2$GeTe$_2$26, CrI$_3$27, CrGeTe$_3$28, MnSe$_2$29, and VSe$_2$30 with long-range magnetic orderings have been extensively reported in the last few years, which have been providing an ideal platform to study fundamental properties of magnetism such as magneto-optical and magnetoelectric effect for ultra-compact spintronics in reduced dimensions. Moreover, recent works have proposed that Néel-type skyrmions with isotropic DMI can be realized in 2D Janus magnets, e.g., MnXY31, CrXY32, and multiferroic structures, e.g., CrN33, BaTiO$_3$/SrRuO$_3$34, In$_2$Se$_3$/MnBi$_2$Se$_3$Te$_2$35. However, it is worth noting that anisotropic DMI has not been reported yet in 2D magnets. Different from previous materials with isotropic DMI vectors along with x and y directions, $M$Cu$X_2$ with special crystal symmetry has an anisotropic DMI vector. Skyrmion Hall effect will cause FM skyrmions with opposite topological charges to propagate in opposite directions, instead of moving parallel to injected current. Antiskyrmions Hall angle is strongly dependent on the direction of applied current related to the internal spin texture of antiskyrmion. When applying spin-polarized current to drive an antiskyrmion, the propagation direction of the antiskyrmion will follow the current direction without topological skyrmion Hall effect36–41. Therefore, it is possible to achieve the zero antiskyrmion Hall angle in a critical current direction.

In experiments, in order to discover 2D materials, a lot of efforts have been devoted to finding materials with characteristics of weak interlayer bonds, which allow their exfoliation down to a single layer by mechanical or liquid-phase approach42,43. Peeling off the three-dimensional layered van der Waals materials, such as CrGeTe$_3$44, CrI$_3$45, and Fe$_2$GeTe$_2$46, etc. Bulk FeCuTe$_2$ is a layered magnetic material stacked by weak interlayer van der Waals interaction, and it has been reported experimentally44–46. It is reported bulk FeCuTe$_2$ has a layered structure, and the unit cell parameters are as follows: $a = 3.93 \, \text{Å}$, $c = 6.078 \, \text{Å}$10. The layered compound FeCuTe$_2$ presents an antiferromagnetically ordered state below $T_N = 254 \, \text{K}$47. Based on first-principles calculations, we further determine the magnetic properties of bulk FeCuTe$_2$. The calculated results show that the unit cell parameters $a$ and $c$ of bulk FeCuTe$_2$ are 3.964 and 6.176 Å, respectively. Moreover, we also compare energies with different magnetic orderings as shown in Supplementary Fig. 10. Bulk FeCuTe$_2$ prefers AFM V in the ground state. From these results, one can see that theoretical results are consistent with the experimental report. Before studying the properties of monolayer FeCuTe$_2$, we theoretically simulate the exfoliation energy when blocks were gradually peeled into a two-dimensional structure as shown in Supplementary Fig. 1. The calculated cleavage energy is 0.46 J/m$^2$, which is comparable to that of graphene or phosphene48,49, indicating the high possibility that layered 2D FeCuTe$_2$ can be exfoliated from...
RESULTS AND DISCUSSION

Crystal structure of monolayer MCuX$_2$

Top and side views of the crystal structure of single-layer MCuX$_2$ (M3d TM; X=group VIA) are represented in Fig. 1a–c. It exhibits a tetrahedral structure with M-X$_{top}$-M and M-X$_{bot}$-M configurations along x and y directions, and the face-centered coordinates are occupied by Cu atoms in the middle layer. The MCuX$_2$ structure belonging to $D_{2d}$ (No.115) crystal symmetry has the symmetry generators: identity operation $E$: (x, y, z); mirror plane $\rightarrow$ (−x, −y, z); fourfold roto-inversion $I_C$: (x, y, z) → (y, −x, −z); and twofold axis $C_2$: (x, y, z) → (x, −y, z), which leads to its feature with intrinsic inversion symmetry broken. According to the Moriya rules\textsuperscript{10}, the induced DMI sign should be opposite along x and y directions.

Via first-principles calculations, we obtain the basic magnetic parameters of MCuX$_2$ ternary compounds as shown in Table 1 (Computational details are presented in the experimental method). The $J_1$ and $J_2$ indicate the exchange coupling constants between NN and NNN atoms. In most systems, $J_1$ is orders larger than $J_2$. There are frustration coming from competing exchange interactions between $J_1$ and $J_2$ in the classical Heisenberg model on the tetrahedra structure except for VCuS$_2$ and VCuSe$_2$. Interestingly, for MCuX$_2$, when M changes from V to Mn and from Fe to Ni, the systems are tuned from FM to G-type AFM, respectively. In tetrahedra crystal, the five $d$ orbitals of transition metal ions split into two groups lower $e$ ($d_{x^2}$, $d_{y^2}$, $d_{z^2}$) and higher $t_2$ ($d_{xy}$, $d_{xz}$, $d_{yz}$) levels due to the influence of the crystal field. The $t_2$ $\leftrightarrow$ $e$ super-exchange interaction favors the appearance of FM coupling, while the $e$ $\leftrightarrow$ $e$ direct exchange and $t$ $\leftrightarrow$ $t$ direct exchange prefer AFM coupling. When the $d$ orbit is more than half-filled with electrons, the AFM coupling mainly benefits from $t$ $\leftrightarrow$ $t$ direct exchange. Thus, in the MCuX$_2$ family with $d$ orbit no less than half-filled, the competitive AFM coupling is stronger. Similar results are also depicted in the zinc-blende binary transition metal compounds\textsuperscript{50}. Collecting all 3d TM atoms magnetic moments in MCuX$_2$ monolayer [see Table 1]. We can find that Mn atoms have the highest magnetic moments and they monotonically decrease on both sides of Mn. It is obvious that the overall trend across the 3d TM row obeys Hund’s rule\textsuperscript{51}.

DMI of monolayer MCuX$_2$

Figure 2 shows the calculated NN-DMI of MCuX$_2$ structures based on the chirality-dependent total energy difference approach\textsuperscript{52}. It is found that all systems have anisotropic DMI and $d_z$ and $d_x$ have opposite signs along x and y directions, which is consistent with the DMI analysis at the beginning. Besides, the DMI strength varies from 0 to 15 meV/atom. These DMIs are very large compared to many state-of-the-art FM/HM heterostructures and 2D Janus structures, e.g., Co/Pl ($\sim$3.0 meV\textsuperscript{52} and Fe/Ir(111) ($\sim$1.7 meV\textsuperscript{53}) thin films and 2D MnTe ($\sim$2.63 meV\textsuperscript{31}). In addition, it is worth

| Family   | $a$(Å) | $J_1$ (meV) | $J_2$ (meV) | $K$ (meV) | $\mu_X$ ($\mu_B$) | $R$ (nm) |
|----------|--------|-------------|-------------|-----------|-------------------|---------|
| VCuS$_2$ | 3.994  | 24.852      | 1.017       | 0.035     | 2.052             | 22.034  |
| VCuSe$_2$| 4.105  | 28.666      | 1.278       | 0.025     | 2.143             | 31.356  |
| CrCuS$_2$| 4.083  | 100.108     | −11.574     | 0.316     | 3.381             | −        |
| CrCuSe$_2$| 4.188 | 104.129     | −10.620     | 0.280     | 3.525             | −        |
| CuTe$_2$ | 4.325  | 101.504     | −4.675      | 0.708     | 3.670             | −        |
| MnCuS$_2$| 4.044  | 35.842      | −1.838      | 0.185     | 4.183             | −        |
| MnCuSe$_2$| 4.119 | 54.848      | −5.741      | 0.131     | 4.249             | 17.14   |
| NiCuS$_2$| 4.100  | 21.805      | −5.430      | −5.910    | 4.268             | 4.000   |
| NiCuSe$_2$| 3.881 | −89.103     | 2.020       | −0.198    | 3.562             | −        |
| CoCuS$_2$| 3.946  | −77.876     | 9.775       | −0.088    | 3.545             | 14.583  |
| CoCuSe$_2$| 4.025 | −50.894     | 10.96       | −2.064    | 3.437             | −        |
| MnCuSe$_2$| 3.957 | −39.310     | 8.996       | 0.435     | 2.346             | 19.231  |
| MnCuTe$_2$| 4.060 | 9.177       | −7.802      | 1.977     | 2.151             | −        |
| NiCuSe$_2$| 3.868 | −17.366     | 2.164       | −0.266    | 0.996             | −        |
| NiCuTe$_2$| 3.877 | −15.516     | 0.141       | −0.241    | 0.968             | 17.797  |
| NiCuTe$_2$| 3.633 | −0.175      | 0.174       | −0.559    | 0.260             | −        |
noting that the DMI of VCuTe$_2$ reaches up to 15.2 meV/atom. In order to verify the correctness of the DMI, we calculated the variation of DMI when the U values of the V atom were 2, 3, and 4 eV, which were 13.2, 15.2, and 11.6 meV/atom, respectively. Notably, CoCuTe$_2$ is different from other antiferromagnetic structures. We find that this structure tends to the Strip-AFM structure with a different lattice constant in x and y direction. Meantime, CoCuTe$_2$ also has the kind of anisotropic DMI in Fig. 2, which is $-0.37$ and $4.2$ meV/atom along x and y directions, respectively.

To elucidate the microscopic energy source of DMI, we calculate the atomic resolved SOC energy difference $\Delta E_{SOC}$ with opposite chirality associated with DMI. Only the $\Delta E_{SOC}$ are presented along the x-direction in Fig. 3. We can see that M atom and Cu atom contribute a relatively large DMI when the X is the light element S with weak SOC. As the X varies from S to Te, the dominant contribution of the X element to DMI gradually improves in all MCu$_X$ monolayers due to the increase in SOC strength. Similar to the interface of HM/FM$_{52,54}$, in which $\Delta E_{SOC}$ is contributed mainly by heavy 5d metal elements of the interfacial location. In our systems, the Fert–Levy mechanism of DMI can be understood that the heavy chalcogen element plays a significant role in inducing the spin-orbit scattering between two magnetic atoms. In addition, we noticed that the $\Delta E_{SOC}$ of NiCuTe$_2$ is close to zero.

Fig. 2 The calculated anisotropic DMI of monolayer MCuX$_2$. Yellow and blue bars indicate the DMI components of NN magnetic atoms along x and y direction, respectively. Here, $d > 0$ ($d < 0$) represents the anticlockwise (clockwise) chirality.

Fig. 3 Anatomy of DMI for MCuX$_2$ monolayers. a–c Atomic resolved localization of SOC energy difference between clockwise and anticlockwise chirality for iron/cobalt/nickel (a), chromium/manganese (b), and vanadium ternary compounds.

about 4.100 and 7.265 Å, as shown in Supplementary Fig. 3. The main reason is that the magnetic moment of Ni atoms in the system is very small, which leads to a small contribution to DMI. Of course, we also check the different U value from 2 to 4 eV, very small magnetic moments are obtained.

Next, according to Moriya’s rules$^{10}$ and the structural symmetry analysis above, the DMI vector for each pair of NNN M atoms is parallel to their bonds because the twofold rotation axes are along the directions between two NNN magnetic atoms [see Supplementary Fig. 2]. In our calculated structure with $D_{2d}$ crystal symmetry, the staggered spin vector will rotate in the plane perpendicular to the propagation direction $<110>$. However, we ignore the NNN DMI in our theoretical calculations, because we find that the DMI between NN and NNN atoms differs by about two orders of magnitude by using the four-state energy-mapping analysis$^{55}$ e.g., VCuSe$_2$ and MnCuSe$_2$ (NNN DMI is $-0.082$ and $-0.075$ meV). Although the NNN DMI is neglected, we still observe the magnetic structure of Bloch-type helicoid from the results of the micromagnetic simulation.

Chiral spin textures of monolayer MCuX$_2$

Furthermore, we perform the atomistic micromagnetic simulation based on first-principles calculated materials parameters as shown in Table 1 by using the VAMPIRE software package$^{56}$. To get the dynamics of magnetization, the Landau–Lifshitz–Gilbert (LLG)
equation was used with the Langevin dynamics as follows:
\[
\frac{\partial \mathbf{S}_i}{\partial t} = -\gamma \left( \mathbf{S}_i \times \mathbf{H}_{\text{eff}} + \lambda \mathbf{S}_i \times (\mathbf{S}_j \times \mathbf{H}_{\text{eff}}) \right)
\]
where \(\mathbf{S}_i\) is the normal unit vector of \(i\)th magnetic atom, \(\gamma\) is the gyromagnetic ratio and \(\lambda\) is the Gilbert damping constant. The magnitude of the effective field is obtained by the equation:
\[
\mathbf{H}_{\text{eff}} = -\frac{\mu_i}{\mu_B} \frac{\partial H}{\partial \mathbf{S}_i}
\]
In all micromagnetic simulations, we relax a random state of 100-nm-wide with periodic boundary conditions as the initial state to get the ground states without an external magnetic field. For all FM systems, uniform FM and FM Néel chiral structures are observed from CrCuS\(_2\) to MnCuTe\(_2\) in Fig. 4 due to the enhancement of ferromagnetic exchange coupling. Besides, in the VCuSe\(_2\) monolayer, antiskyrmions with a diameter of 31 nm emerge on the large ferromagnetic domain. It is also important to know the topological charge \(Q\). Here, we apply the lattice-based approach to calculate the topological charge. First, we perform the atomic spin model simulations to obtain the spin vector \(\mathbf{S}\) on each lattice point. Furthermore, we use the formula: \(\mathbf{S} \cdot (\partial_x \mathbf{S} \times \partial_y \mathbf{S})\) to calculate the topological charge density of each lattice point. Finally, we integrate the charge density of the zone holding the topological magnetic quasiparticle to obtain the final topological charge. The topological charge is \(+1\) in the zoomed antiskyrmion in Fig. 4d. More interestingly, we also realize the FM anti-bimerons arising from strong in-plane magnetic anisotropy in MnCuTe\(_2\). In all AFM systems, the topological charge of each AFM antiskyrmion with antiparallel NN spin alignment is 0. It can be decomposed into two identical FM antiskyrmion sublattices, where the FM sublattice pairs have opposite topological charges \(+1\) and \(-1\). In FeCuSe\(_2\), AFM antiskyrmion and AFM antiskyrmionium are presented in Fig. 4i, j. It is well known that lots of topologically magnetic textures can be induced by exchange frustration\(^{57,58}\). Notably, previous work have demonstrated that DMI and exchange frustration can stabilize skyrmionium in CrGe(Se,Te)\(_3\) Janus monolayer when \(J_1/J_2\) is within a certain range\(^{59}\). Similar to AFM antiskyrmion, AFM

Fig. 4  Spin configurations of ground states of monolayers M\(\text{CuX}_2\) in real-space. Spin configuration of a 100 nm × 100 nm square and zoom of isolated chiral magnetic profile of V\(\text{CuS}_2\) (a, b); V\(\text{CuSe}_2\) (c, d); Mn\(\text{CuS}_2\) (e, f); Mn\(\text{CuTe}_2\) (g, h); Fe\(\text{CuSe}_2\) (i, j); Ni\(\text{CuSe}_2\) (k, l); Co\(\text{CuS}_2\) (m, n) and Co\(\text{CuSe}_2\) (o, p). The color map indicates the out-of-plane spin component.
antiskyrmion ion with zero topological charge refers to a magnetic texture that can be viewed as two nested AFM antiskyrmions due to the frustration caused by opposite sign exchange coupling $J_1$ and $J_2$. In addition, we note that isolated bimerons and multiple bimerons are observed in CoCuS$_2$ and MnCuTe$_2$, respectively. The main reason is that FM exchange coupling can be consistent with an effective FM external field. When we reduce the $J_1$ of CoCuS$_2$, multiple bimerons appear, as is shown in Supplementary Fig. 9. Similar to that of CoCuS$_2$, when we increase $J_1$ or external magnetic field in the VCuS$_2$ system, respectively, one can observe that isolated antiskyrmions emerge, as is shown in Supplementary Fig. 11. Meantime, we also adopt open boundary conditions to simulate the spin textures of FM MnCuSe$_2$ and AFM CoCuSe$_2$, as shown in Supplementary Fig. 6. One can see that the simulated FM antiskyrmions in MnCuSe$_2$ and AFM antiskyrmions in CoCuSe$_2$ under open boundary conditions have almost the same size and topological properties with periodic boundary conditions. Furthermore, we also take into account the effect of dipolar interaction on chiral magnetic textures. For antiferromagnetic systems, magnetic dipolar interaction diminishes due to the cancellation of the magnetic moment of coupled sublattices. Thus, we simulated VCuSe$_2$ and MnCuSe$_2$ monolayers based on first-principles calculated magnetic parameters, one can see that the calculated spin configurations of VCuSe$_2$ and MnCuTe$_2$ are consistent with previous results as shown in the Supplementary Fig. 7.

In addition, we also calculate magnetic parameters $J_1$, $J_2$, $K$, and $D$ of monolayer VCuSe$_2$ when the tensile strain increases from 1 to 5%, as shown in Table 2. We find that NN and NNN FM exchange coupling strengths decrease a lot while DMI changes slightly, resulting in a large ratio of $D/J$. Therefore, antiskyrmion with a smaller diameter is achieved under tensile strain. The phase diagram of the VCuSe$_2$ monolayer under different stress and temperatures is shown in Fig. 5. For pristine VCuSe$_2$, the FM antiskyrmions embedded in the background of the large-size domain are observed [Fig. 4c]. It is found that stable chiral domain and magnetic antiskyrmions appear slowly when the temperature decreases, one can see that the chiral magnetic structures can be basically stabilized at 250 K, and these chiral structures also undergo some small changes as the temperature continues to decrease to the finite temperature. Moreover, as the strain increases, the density of antiskyrmions increases, and the size of the domain becomes much smaller than that of the pristine state. Supplementary Fig. 8 presents the results of the micromagnetic simulations for spin spiral length as a function of strain. We can clearly observe the strain dependence of spin spiral length with the increasing $D$ and decreasing exchange $J$, which is consistent with $|D/J|$.

In summary, we discover a group of 2D layered ternary compounds MCuX$_2$ (M:3d TM; X:group VIA) with anisotropic DMI protected by $D_{2d}$ crystal symmetry from first-principles calculations. We show that the anisotropic DMI can vary from 0 to 15 meV/atom in 2D layered ternary compounds MCuX$_2$, where $M$ represents 3d transition metal, and $X$ represents the VIA group element. Thanks to the large enough anisotropic DMI, we demonstrate that a series of FM (AFM) antiskyrmions, bimerons, and antiskyrmion ion can be realized without an external field in the MCuX$_2$ family. The discovery will provide a platform to find various FM/AFM anti-topological spin textures with crystal symmetry-protected anisotropic DMI. In addition, our calculations show that magnetic parameters of VCuSe$_2$ are sensible to strain, and the possibility of antiskyrmions formation up to a hundred Kelvin is demonstrated in 2D VCuSe$_2$. Our work will benefit both fundamental research and applications in the fields of 2D van der Waals materials and spintronics.

### METHODS

**DFT calculations**

First-principles calculations are carried out based on density functional theory (DFT) implemented in Vienna ab-initio Simulation Package (VASP). We adopt Perdew–Burke–Emzerhof (PBE) functionals of the generalized gradient approximation (GGA) as the exchange-correlation potential, and use projector augmented plane-wave (PAW) method to deal with the interaction between nuclear electrons and valence electrons. We set the cutoff energy of 520 eV for the plane-wave basis set, and $24 \times 24 \times 1$ k-point with F-centered meshes for the Brillouin zone integration. Partially occupied d orbitals of transition metal atoms are treated by GGA+U method with $U = 3$ eV for the 3d orbitals of metal and Cu elements. We set a vacuum layer with a thickness of 25 Å in the z-direction to ensure that there is no interaction between the periodic images. The convergence criteria of the total energy in the ion relaxation process and the Hellmann–Feynman force between atoms were set to be $1 \times 10^{-7}$ eV and 0.001 eV Å$^{-1}$, respectively. To describe our magnetic system, we adopt the following Hamiltonian model:

$$H = -J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - J_2 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_{\langle i,j \rangle} D_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) - K \sum_i (\mathbf{S}_i^2)^2$$

(2)

where $\mathbf{S}_i$ is the normal spin vector of the $i$th magnetic atom, the $J_1$ and $J_2$ represent exchange coupling constants between Nearest-Neighbor (NN)
and Next-Nearest-Neighbor (NNN) atoms, respectively. \( K \) is the magnetic anisotropy constant and \( D_i \) is the DMI vectors. The methods to calculate \( J \), \( K \), and \( D \) is described in the experimental section.

### Magnetic parameters

**Exchange coupling constant.** We construct a \( 2 \times 2 \times 1 \) supercell to study three different magnetic configurations, which are FM, G-type AFM where nearest-neighbor spins are aligned antiparallel and Stripe-type AFM where spins are ordered antiferromagnetically (ferromagnetically) along with \( x \) (y) axis (see Supplementary Fig. 4). Exchange coupling constant of nearest neighbor (NN) and next-nearest neighbor (NNN) magnetic atoms are obtained based on the following formula:

\[
E_{\text{FM}} = -\frac{1}{2} \times 4(4J_1 + 4J_2) + E_0
\]

\[
E_{\text{AFM}} = -\frac{1}{2} \times 4(-4J_2) + E_0
\]

\[
E_{\text{AFMII}} = \frac{1}{2} \times 4(4J_1 + 4J_2) + E_0
\]

\[
J_1 = \frac{E_{\text{AFMII}} - E_{\text{FM}}}{16}
\]

\[
J_2 = \frac{2 \times E_{\text{AFM}} - E_{\text{AFMII}} - E_{\text{FM}}}{32}
\]

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

**Magnetic anisotropy energy \( K \).** Magnetic anisotropy energy is defined as the energy difference between the in-plane magnetized [100] axis and the out-of-plane magnetized [001] axis:

\[
K = E_{100} - E_{001}
\]

**NN Dzyaloshinskii–Moriya interaction (NN-DMI) \( D \).** We performed DMI calculations using the chirality-dependent total energy difference method. First of all, a \( 4 \times 4 \times 1 \) supercell is constructed to obtain the charge distribution of the system’s ground state by solving the Kohn–Sham equations in the absence of spin-orbit coupling (SOC). Then, SOC is included and we set spin spirals to determine the self-consistent total energies in the clockwise and anticlockwise rotation. Finally, the energy difference between clockwise and anticlockwise rotation is calculated to obtain the anisotropic \( D \). The DMI can be obtained by the following formula:

\[
D = (E_{\text{FCI}} - E_{\text{SOC}})/8
\]

**NNN Dzyaloshinskii–Moriya interaction (NNN DMI).** We performed the four-state energy-mapping analysis. First of all, a \( 4 \times 4 \times 1 \) supercell is constructed to set all the spin configuration in the \( y \) direction, then change the spins between the two NN atoms. The DMI between NN \( M \) spins were calculated based on four spin configurations: (i) \( S_1 = (0, 0, 0), S_2 = (0, 0, 0), \) (ii) \( S_1 = (0, 0, 0), S_2 = (0, 0, -S) \); (iii) \( S_1 = (-S, 0, 0), S_2 = (0, 0, 0) \); (iv) \( S_1 = (-S, 0, 0), S_2 = (0, 0, -S) \). Next, according to spin interaction energy based different spin configurations, we can solve the in-plane component \( D_y \):

\[
D_y = (E_1 + E_2 - E_4)/4S^2
\]

**Phonon spectrum**

In calculations, based on the PHONOPY code, we use \( 3 \times 3 \times 1 \), \( 3 \sqrt{2} \times 3 \sqrt{2} \times 1 \) and \( 4 \times 4 \times 1 \) supercells with the frozen phonon approximation to calculate the phonon dispersions of single-layer MoS\(_2\). Supplementary Fig. 5 shows that \( \Gamma \) point of some of the MCL\(_2\) monolayers have very small imaginary frequencies within the entire wave-vector space, which can be attributed to a wavelength of particular mode.

### DATA AVAILABILITY

The datasets used in this article are available from the corresponding author upon request.

### CODE AVAILABILITY

Code that supports the findings of this study are available from the corresponding authors on reasonable request.

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AUTHOR CONTRIBUTIONS

H.Y. conceived the project. Y.G. performed the calculations. Y.G., Q.C., and H.Y. analyzed the results and wrote the manuscript. All the authors contributed to the discussion and the writing.

COMPETING INTERESTS

The authors declare no competing interests.

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