Giant Dzyaloshinskii-Moriya interaction and ratchet motion of bimeronic excitations in two-dimensional magnets

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(Dated: November 23, 2021)

Topological magnetic excitations rooted in Dzyaloshinskii-Moriya interaction (DMI) are promising information carriers for next-generation memory and spintronic devices. The recently discovered two-dimensional (2D) magnets provide fertile new platforms for revealing rich physical phenomena with atomic-scale precision, as exemplified by the enhanced DMI associated with an effective electric field that breaks the inversion symmetry. Here we use first-principles calculations to establish a conceptually different compositional engineering approach to induce giant DMI in a representative CrMnI3 2D magnet, and the underlying mechanism is rooted in the spontaneous inversion symmetry breaking in the bipartite system. Using atomistic magnetics simulations, we further reveal that bimeronic excitations can emerge upon cooling a spin random state or perturbing a ferromagnetic state. Strikingly, the bimeron can exhibit unidirectional soliton-like propagation, and such a ratchet phenomenon is highly desirable for racetrack memories. These findings may prove instrumental in developing novel devices for quantum information based on 2D magnets.

The past decades have witnessed the entry and fertilization of the concept of topology in various realms of condensed matter physics, where geometric phase is introduced to effectively characterize the topologically nontrivial electronic and magnetic properties of solids. Different from the electronic band topology [1], the geometric phase of magnetic structures evolves in real space and manifestation of the corresponding topology is more vivid and intuitive. As a compelling example, the well-known nontrivial magnetic entity of skyrmion cannot be adiabatically deformed into a trivial ferromagnetic state, a distinct and striking property that can be exploited for novel spintronic applications [2]. These promising technological potentials, in turn, have propelled intensive research efforts in searching for more desirable hosting materials, developing better imaging techniques, and achieving more precise control of such topological magnetic quasiparticles [3–5].

Although topological magnetic structures encompass abundant forms [4], their underlying formation mechanisms are predominantly rooted in the same quantum mechanical and relativistic origin, namely, the Dzyaloshinskii-Moriya interaction (DMI) [6, 7]. Historically, the DMI was introduced to interpret the weak ferromagnetism in prototypical antiferromagnets such as α-Fe2O3 [6], but subsequently, it was found that the DMI can also play an essential role in stabilizing the vortex-like configurations in model ferromagnetic systems [8]. The DMI exchange energy has a chiral form \( D_{ij} \cdot (S_i \times S_j) \), thus favoring perpendicular spin alignment within the plane normal to the DM vector \( D_{ij} \). This antisymmetric exchange coupling of two magnetic moments arises in materials with simultaneous spin-orbit coupling (SOC) and structural inversion symmetry breaking. Recently, layered van der Waals magnets [9], with CrI3 [10] and CrGeTe3 [11] as the representatives, have been experimentally thinned to the mono- or few-layer limit while preserving their long-range ferromagnetic (FM) order. As the long-sought members in the portfolio of 2D materials, such 2D magnets provide fertile new platforms for revealing rich physical phenomena with atomic-scale precision, and can also serve as new building blocks in various magneto-devices with higher integration, lower energy consumption, and faster processing [12]. Nevertheless, the DMI is absent in the exchange coupling of two nearest neighbored magnetic sites in pristine CrI3 [13], CrGeTe3 [13], and many other subsequently discovered 2D magnets [14, 15], due to their inherent crystalline symmetry [16]. To induce substantially large DMIs, various symmetry-breaking scenarios have been proposed, such as by applying a vertical electric field [17] or invoking Janus structures [18–20] or heterojunctions [21]. In essence, these approaches share the same commonality, namely, by creating an effective electric field perpendicular to the 2D magnets [as sketched in Fig. 1(a)], conceptually similar to those extensively studied at interfaces of ferromagnetic and heavy metal films [22, 23]. The topological magnetic entities associated with such DMIs are typically constrained to be Néel type. Novel centro-symmetry breaking schemes are desired to further enrich and expand the territory of 2D topological magnetism.

This Letter makes multifold contributions. First, we employ a conceptually different design scheme to induce giant DMI in intrinsic yet physically realistic 2D magnets. Here, the underlying physical mechanism does not rely on the establishment of an effective electric field to
break the centrosymmetry; instead, the DMI arises from the inherent asymmetry of the magnetic species in the systems [as illustrated in Fig. 1(b)]. Specifically, our first-principles calculations reveal that giant Bloch-type DMI exists in a prototypical CrMnI$_6$ monolayer [24], which is isostructural to CrI$_3$ but with bipartite asymmetric. Salient new magnetic properties are also shown to emerge upon such compositional engineering, including easy in-plane ferromagnetism, as well as anisotropic bilinear and pronounced biquadratic exchange couplings. Secondly, we use atomistic magnetics simulations to demonstrate that paired meronic excitations (bimerons) can be selected upon cooling or controllably excited. Most strikingly, a solitonic bimeron can be driven to move unidirectionally by thermally excited magnons, revealing a ratchet feature in the interior of the in-plane FM racetrack [25, 26]. This exotic phenomenon, predicted for the first time in 2D magnets, can be further exploited for memory and spintronic device applications. These findings may not only provide instrumental insights to understanding chiral exchange interactions and topological magnetism in the ultrathin regime, but also help to accelerate the applications of 2D magnets in quantum devices such as in racetrack memories [27].

**Giant DMI in the bipartite CrMnI$_6$ monolayer.**—The CrMnI$_6$ monolayer shown in Fig. 1(c) can be obtained by substituting one sublattice of chromium in the CrI$_3$ monolayer with manganese, and the ordered structure with bipartite spin lattice (Cr$^{3+}$ and Mn$^{3+}$ ions with three and four $\mu_B$ respectively) as shown in Fig. 1(d) is energetically favored [24]. The corresponding Curie temperature was estimated to be enhanced compared to CrI$_3$ upon substitution. Because of the coexistence of centro-asymmetry and large SOC, the system is expected to harbor pronounced DMI.

To investigate the anisotropy of exchange couplings in the monolayered CrMnI$_6$ system, we first assume an atomistic spin Hamiltonian with bilinear (BL) terms,

$$H_{BL} = -\sum_{i} A_{i,zz} S_i^z S_j^z - \sum_{i<j} S_i S_j^{\text{sym}} - \sum_{i<j} D_{ij} \cdot (S_i \times S_j) - \mu_B \sum_i B \cdot S_i. \tag{1}$$

The first term represents the single-ion anisotropy arising from atomic SOC, which is reduced to a single scalar due to the three-fold rotational symmetry of the system [28]. The second and third terms correspond to the symmetric and antisymmetric (DMI) parts of the exchange couplings, respectively, with the nearest neighbor (NN) and next nearest neighbor (NNN) terms indicated in Fig. 1(d). The last term accounts for the Zeeman energy due to the external magnetic field $B$.

We perform density functional theory calculation with a mean-field onsite Coulomb repulsion correction for the localized $d$-orbitals (DFT+$U$), to attain the energetics of the monolayered CrMnI$_6$ [29–43]. For the magnetic degrees of freedom, we consider four specifically designed non-collinear spin configurations following the powerful four-state method [28, 44], from which the exchange parameters can be conveniently extracted with DFT accuracy [13, 45–47]. The calculated bilinear exchange parameters are summarized in Table S1 [48]. One can see that the symmetric exchange couplings are predominantly ferromagnetic, but exhibit highly anisotropic characteristics, as reflected by the distinctly different diagonal values in the $J^{\text{sym}}$ tensors and some non-vanishing off-diagonal components. For the antisymmetric part (DMI), more insights can be drawn regarding both directions and magnitudes. For directions, our calculated DM vectors are consistent with the Moriya’s rules [16], namely, the symmetry-forbidden components are numerically negligible as well ($<0.01$ meV). For magnitudes, the $|D|/J_{\text{iso}}$ ratio [49] has been widely used to evaluate the competition between the DMI and $J_{\text{iso}}$, which respectively favor canted and collinear alignments of the magnetic moments, with the trace average $J_{\text{iso}} = \text{Tr}(J^{\text{sym}})/3$. For the three considered bilinear exchange coupling strengths, the $|D|/J_{\text{iso}}$ values reach 0.31 (Cr-Mn), 0.44 (Cr-Cr), and 0.82 (Mn-Mn) respectively, the latter two substantially exceeding the corresponding ratios in the prototypical Fe/Ir(111) system [50] and other

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**FIG. 1.** (a) and (b) Schematics of two different approaches to induce DMIs in monolayered magnets (silver/green balls: magnetic cations, orange balls: nonmagnetic anions). (a) The prevailing strategy, relying on an effective vertical electric field $E_{\text{eff}}$. (b) Creating an asymmetric bipartite lattice as proposed here. (c) Top view of the atomic structure of monolayered CrMnI$_6$, showing the Cr and Mn ions centered in edge-sharing $I_6$ octahedra. (d) The honeycomb spin lattice with the NN and NNN exchange couplings indicated.
chiral magnets hosting topological spin textures \[19\]. These findings indicate that giant chiral magnetic interactions emerge in this system of broken inversion symmetry, a salient feature that is highly desirable for exploring exotic topological magnetism.

**Magnetic anisotropy and biquadratic coupling.**—Another important physical aspect to investigate is the magnetocrystalline anisotropy energy (MAE), which indicates the easy magnetization orientations. We employ DFT+U calculations to trace the total energy evolution of the FM state upon rotating the spin quantization axis. It is found that the total energy of the system remains nearly isotropic within the \(xy\) plane, but is lower than that of the out-of-plane FM state, suggesting that the system favors in-plane magnetization. These features can be correctly predicted by the BL model [Eq. (1)], indicating its capacity to depict the in-plane physics. We therefore use the in-plane FM configuration as the reference state to calculate the MAE using this model. The overall energy evolution trend can be qualitatively well reproduced [Fig. 2(a)], indicating that the MAE mainly stems from competition between bilinear spin-spin interactions. On the other hand, there exists pronounced discrepancy at the quantitative level, which hints on the existence of some complex higher-order spin-spin interactions.

Given the protection of time-reversal symmetry \[51\], the likely leading higher-order term is the biquadratic (BQ) coupling \[52, 53\]. To reveal the potential existence and microscopic origin of such BQ interactions, we systematically investigate the element-resolved single-ion and pair-resolved ion-ion contributions. The single-ion and NN exchange coupling are both found to be bilinear \[54\]. Then we study the NNN pairs by rotating the spins of one sublattice synchronically from \(x\) to \(z\) while fixing the other sublattice magnetized along \(y\) \[55\]. By comparing the energy evolutions between the DFT+U and BL model calculations, we see quantitative consistency upon rotating spins of the Cr sublattice. In stark contrast, pronounced differences exist when the spins of the Mn sublattice are rotated [Fig. 2(b)], strongly pointing to the likely source of the MAE discrepancy shown in Fig. 2(a). Microscopically, this contrast could originate from their different 3d-electron fillings in the presence of an octahedral crystal field [Fig. 1(c)]. Both ions have three \(d\) electrons occupying the low-lying \(t_{2g}\) orbitals in the spin majority channel, while the Mn\(^{3+}\) ions (\(d^4\)) have an extra \(d\) electron populating the higher-energy \(e_g\) orbitals in the same spin channel. The exchange coupling of the \(e_g\) electrons within a Mn-Mn pair in return gives rise to pronounced BQ spin-spin interaction.

We can now further improve the spin Hamiltonian in Eq. (1) on solid physics basis. Unlike earlier studied systems with isotropic BQ exchange couplings \[52, 53\], one distinctly new feature of the present 2D system is that the BQ coupling must be highly anisotropic, because it contributes substantially to the MAE. Here we adopt a simple and physically intuitive anisotropic form,

\[
H_{\text{BQ}} = \frac{1}{2} \sum_{\{i,j\}} K_{\text{Mn-Mn}}^{\text{zz}} S_{iz}^2 S_{jz}^2,
\]

with \(K_{\text{Mn-Mn}}^{\text{zz}}\) being the only coefficient to be determined. With the inclusion of this BQ coupling, the energy evolution trend upon rotating the Mn sublattice spins can be significantly improved, as shown in Fig. 2(b). As a crucial crosscheck, it also improves the description of the MAE quantitatively, as confirmed in Fig. 2(a). The optimally compromised value that can simultaneously improve the quantitative accuracies in both Fig. 2(a) and (b) is -0.023 meV, with the negative sign indicating a BQ contribution to in-plane magnetization. The coefficient seems to be weak at the first glance, but given the large magnetic moments of Mn (\(S=2\)), the BL [\(J_{\text{BQ}} S^2 = 1.4\) meV] and BQ [\(K_{\text{Mn-Mn}}^{\text{zz}} S^4 = -0.37\) meV] coupling strengths between NN Mn-Mn pairs are readily comparable.

**Bimeronic topological excitations.**—The BL-BQ spin Hamiltonian combining Eqs. (1) and (2) now provides an accurate description of the bipartite spin lattice of monolayered CrMnI\(_6\). To explore potential chiral magnetic structures, we perform atomistic magnetics simulations by numerically integrating the Landau-Lifshitz-Gilbert (LLG) equation \[37\], which can depict the precession and damping of localized spins in the presence of exchange and external magnetic fields \[56\]. We start the LLG simulations from a randomly magnetized configuration within a \(36\times36\) supercell which mimics the paramagnetic state. The damping term in the LLG equation dissipates energy of the spin system and drives it to lower energy state, effectively modeling a cooling process \[57\]. To trace the real space geometric phase of the dynamic spin structure, we calculate the topological charge \(Q\) \[41\] evolution during the simulation. As shown in Fig. 3(a), \(Q\) fluctuates drastically during the initial transient period of the simulation, and then converges to lower values.
indicating the emergence of topological chiral spin textures upon cooling. To directly visualize the whirling of the spin vectors, selective snapshots at \( t = 10 \) and 17 ps are plotted. The spin structures clearly show the existence of vortex- or antivortex-like local excitations in the cycloidal background, with different topological charges. Given that the material strongly favors in-plane magnetization, we assign these nontrivial magnetic structures to stem from merons \([58]\) instead of skyrmions that require out-of-plane magnetization \([4]\). We also note that the pairing of two meronic excitations (also called bimerons) illustrated in Fig. 3(b, c), involving one vortex plus one antivortex, fulfills the generic vorticity conservation condition, which is qualitatively analogous to the topological defects of a 2D system below the critical temperature of the Berezinskii–Kosterlitz–Thouless transition \([59]\). Such bimeronic excitations have been predicted or observed in other easy in-plane magnets \([47, 57, 60–62]\). Similar nontrivial chiral magnetic structures are observed in several independent simulations from different paramagnetic initial states, further confirming that such bimeronic low-energy excitations are the intrinsic property of the monolayered \( \text{CrMnI}_6 \) governed by its giant DMI and in-plane magnetization. However, such bimeronic quasiparticles selected upon cooling may still possess stochastic uncertainty with regard to their topological charge, size, shape, position, and lifetime when connecting with practical spintronic applications. To overcome such limitations, it is necessary to explore approaches of writing and deleting an individual bimeron in a more controlled fashion \([61, 63]\). Our additional comprehensive simulations \([64]\) show that an isolated bimeron can be excited from the in-plane FM background with the aid of a pulse-like, confined vertical magnetic field, and can also be conveniently erased by applying a small in-plane magnetic field. Such operating conditions are physically clearly realistic \([63]\), and the topological excitations with well-defined spatial locations and soliton-like features may exhibit exotic bimeronic dynamics in racetrack geometry.

**Thermally-driven ratchet motion of bimeron.**—To study the dynamics of bimeron quasiparticles, we create a \( 100 \times 36 \) supercell in a FM racetrack geometry, and imprint a single Bloch-type bimeron with LLG relaxed configuration. Afterwards, a mild homogeneous thermal field (0.5 K) is additionally exerted to the system. The evolutions of the corresponding spin energy and topological charge are traced, as shown in Fig. 4(a). The spin energy increases by \( \sim 0.04 \) meV/site in a few picoseconds, and then fluctuates within a very small energy range, implying that the system has been driven to a steady state. The topological charge conserves the value of -1 throughout the simulation period of 200 ps, implying that the profile of the bimeron is robust and no extra topological excitations are triggered by the thermal field.

To further track the spatial variations of the bimeron, we selectively plot its snapshots in Fig. 4(b)-(d). Strikingly, the bimeron not only preserves its original geometry with the mutually bound vortex and antivortex, but its center of mass also exhibits a unidirectional trajectory of motion parallel to the direction of the magnetization, effectively forming a magnetic ratchet \([65]\). Such intriguing ratchet effects have been predicted or observed in other types of chiral magnetic structures such as domain walls \([66, 67]\) and skyrmions \([25, 26]\) within magnetic thin films, but so far has not been reported in any 2D magnet. In particular, the present demonstration also amounts to the first prediction of a ratchet effect surrounding merons. This exotic dynamic behavior can be qualitatively understood from two aspects. The first is

![FIG. 3. (a) Evolution of the topological charge in an LLG simulation starting from a randomly magnetized configuration. (b) and (c) Snapshot configurations. The in-plane and out-of-plane spin components are shown by the arrows and a rainbow color mapping, as also adopted in Figs. 4.](image)

![FIG. 4. Thermally-driven ratchet motion of a bimeron in racetrack geometry. (a) Evolutions of the total energy/site (left axis) and topological charge (right axis) during the LLG simulations. (b)-(d) Snapshots of the spin configurations.](image)
about the likely driving force: because no external charge
or spin current is applied to induce a spin transfer torque,
it is plausible that the thermally excited collective modes
in the form of magnons could emerge to provide the un-
derlying impetus propelling the bimeron to propagate
like a massive quasiparticle [68]. The second is about
the unique directionality, whose underlying mechanism
could be attributed to the concerted effects of asymmetry
as encoded in both the FM configuration and spin Hamil-
tonian. The former introduces strong anisotropy into
the energy landscape, effectively confining the bimeron
motion to 1D [69]. The latter, featured by sublattice-
imbalanced DMIs, cause further differences in the two
directions of motion along a 1D channel. The asymmet-
ric dynamics, along with the robust geometry and topo-
logical charge, characterizes such bimeronic excitations
as reliable information carriers in racetrack applications.

Discussions.—As a practical recipe to induce/enhance
chiral spin-spin interactions and stabilize topological
magnetic configurations, compositional engineering has
been extensively investigated in bulk or thin film materi-
als [70, 71]. In this Letter, we have explicitly extended
the eligibility and feasibility of this design scheme to 2D
magnets, which possess inherent advantage in more deli-
cate spin manipulation. Motivated by this proposal, our
ongoing experimental efforts have preliminarily shown
that layered (Cr, Mn)I₃ films can indeed be fabricated
via chemical vapor growth, and such samples exhibit
novel magnetic properties [72]. Alternatively, molecular-
beam epitaxy and in-situ doping [73, 74] are also power-
ful techniques for compositional engineering of 2D mag-
nets. Unlike previously studied spin systems [75–77],
the bimeron unidirectional transport predicted here does not
rely on specifically designed odd geometry or edges of the
materials. The required in-plane FM ground state and
sublattice-asymmetric DMI, are both intrinsic properties
of the bipartite system, making direct experimental vali-
dations more feasible with simpler setups.

Conclusions.—Based on symmetry considerations and
first-principles calculations, we have established a pow-
erful compositional engineering scheme to induce pro-
nounced DMI in 2D magnets. Staring from the CrI₃
monolayer, the introduction of a second magnetic species
Mn can effectively tune the Ising-type ferromagnetism
of the parent compound, giving rise to giant DMI
and highly anisotropic biquadratic exchange interac-
tions. The bipartite CrMnI₆ monolayer has been further
shown to exhibit nontrivial magnetic properties featured
by nanometric bimeronic topological excitations. Most
strikingly, the solitonic bimeron can achieve thermally-
driven unidirectional motion, which can be further ex-
loited for racetrack applications. Collectively, these
central findings characterize the representative bipartite
CrMnI₆ monolayer as a new and versatile member in the
family of 2D magnets both for intriguing topological mag-
netism and for conceptually new spintronic devices.

This work was supported by the National Natural
Science Foundation of China (Grant Nos. 11904350,
11634011, 11974323, 11722435, and 11804210), the
National Key R&D Program of China (Grant No.
2017YFA0303500), the Strategic Priority Research Pro-
gram of Chinese Academy of Sciences (Grant No.
XDB30000000), the Anhui Initiative in Quantum Infor-
mation Technologies (Grant No. AHY170000), and An-
hui Provincial Natural Science Foundation (Grant.
No. 2008085QA30). S. Z acknowledges Dr. C. S. Xu for kind
help in the four-state method related calculations.

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[43] See Supplementary Material for a more comprehensive coverage of the method. The Supplementary Material includes Refs. [30–43].

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