

Topological spin texture in Janus monolayers of the chromium trihalides Cr(I, X)₃Changsong Xu^{1,*}, Junsheng Feng^{2,3,*}, Sergei Prokhorenko¹, Yousra Nahas¹, Hongjun Xiang^{3,4,†} and L. Bellaïche^{1,‡}¹Physics Department and Institute for Nanoscience and Engineering, University of Arkansas, Fayetteville, Arkansas 72701, USA²School of Physics and Materials Engineering, Hefei Normal University, Hefei 230601, People's Republic of China³Key Laboratory of Computational Physical Sciences (Ministry of Education), State Key Laboratory of Surface Physics, and Department of Physics, Fudan University, Shanghai 200433, People's Republic of China⁴Collaborative Innovation Center of Advanced Microstructures, Nanjing 210093, People's Republic of China

(Received 25 July 2019; revised manuscript received 18 September 2019; accepted 28 January 2020; published 10 February 2020)

Topological magnetic states are promising for ultradense memory and logic devices. Recent progress in two-dimensional magnets encourages the idea to realize topological states, such as skyrmions and merons, in freestanding monolayers. However, monolayers such as CrI₃ lack Dzyaloshinskii-Moriya interactions (DMIs) and thus do not naturally exhibit skyrmions/merons but rather a ferromagnetic state. Here we propose the fabrication of Cr(I, X)₃ Janus monolayers, in which the Cr atoms are covalently bonded to the underlying I ions and top-layer Br or Cl atoms. By performing first-principles calculations and Monte Carlo simulations, we identify strong enough DMIs, which leads to not only helical cycloid phases, but also to topologically nontrivial states, such as the intrinsic domain wall skyrmions in Cr(I, Br)₃ and the magnetic-field-induced bimerons in Cr(I, Cl)₃. Microscopic origins of such spin textures are revealed as well.

DOI: [10.1103/PhysRevB.101.060404](https://doi.org/10.1103/PhysRevB.101.060404)

Introduction. Novel spin textures, such as skyrmions and merons, are nanoscale spin clusters with topological stability, and are promising for advanced spintronics [1–3]. One requirement toward such applications is that the hosting materials should be thin films, so that the nanosize of the spin clusters can be taken full advantage of. Besides previous studies on skyrmionic states of bulk MnSi [4–7], recent works focused on ultrathin films, such as skyrmion states in FeGe [8,9] and rare-earth ion garnet [10,11] and meron states in Co₈Zn₉Mn₃ [12], which all take advantage of the Dzyaloshinskii-Moriya interaction (DMI) arising from relativistic spin-orbit coupling (SOC). However, no skyrmionic state has ever been reported to intrinsically exist in free-standing *monolayers*, to the best of our knowledge. Two-dimensional (2D) semiconducting magnets, such as monolayer CrI₃ [13], are recently attracting much attention due to their novel physics and rich applications [14]. The ferromagnetic monolayer CrI₃ crystallizes in honeycomb lattice made of edge-sharing octahedra. Its ferromagnetic order is stabilized by an out-of-plane anisotropy [13], which arises from single ion anisotropy (SIA) and Kitaev-type exchange coupling that both result from the SOC of its heavy ligands [15,16]. However, the ingredient DMI is absent between the most strongly coupled first-nearest-neighbor (1st NN) Cr-Cr pairs, because the inversion center between the two Cr atoms prevents its existence [17].

Here we propose an approach that consists in fabricating Janus monolayers of chromium trihalides Cr(I, X)₃ (X =

Br, Cl). One example of Janus monolayers is the transition metal dichalcogenides MoSSe, which originates from the well-known monolayer MoS₂ but with one layer of S being substituted by Se. Such Janus monolayer is experimentally achievable, since MoSSe has been reproducibly obtained using different methods [18,19], which strongly suggests the feasibility of also creating Cr(I, X)₃ Janus monolayers. In this Rapid Communication, we apply density functional theory (DFT) and parallel tempering Monte Carlo (PTMC) simulations to study the magnetic interactions and to reveal novel spin textures of two Janus monolayers, namely, Cr(I, Br)₃ and Cr(I, Cl)₃, as well as the prototype CrI₃ [see structural schematics in Figs. 1(a) and 1(b)]. As we will show, these Janus monolayers exhibit not only strong enough DMI, but also a decrease in magnetic anisotropy energy (MAE), which both contribute to stabilizing helical, skyrmion, and even bimeron states. We also demonstrate that the presence of Kitaev interaction and the application of magnetic field both benefit to the emergence of novel spin textures in these Cr(I, X)₃ Janus monolayers.

Hamiltonian and magnetic parameters. We consider the following Hamiltonian for describing the magnetic interactions of the Cr(I, Br)₃ and Cr(I, Cl)₃ Janus monolayers, as well as the prototype CrI₃ monolayer, using the generalized matrix form up to the second-nearest neighbors (2nd NN) for the spins, **S**:

$$\mathcal{H} = \frac{1}{2} \sum_{n=1,2} \sum_{(i,j)_n} \mathbf{S}_i \cdot \mathcal{J}_{n,ij} \cdot \mathbf{S}_j + \sum_i \mathbf{S}_i \cdot \mathcal{A}_{ii} \cdot \mathbf{S}_i, \quad (1)$$

where the first term is the exchange coupling that runs over all first (*i, j*)₁ and second (*i, j*)₂ NN Cr pairs, respectively, and the second term represents SIA that runs over all Cr

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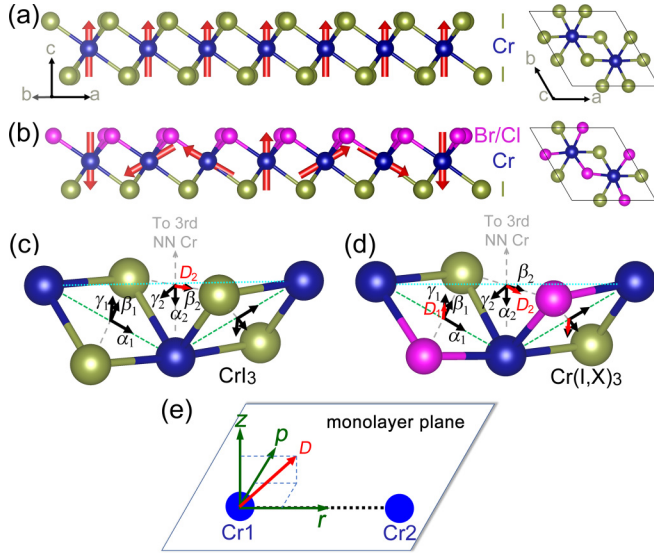


FIG. 1. Panels (a) and (b) display the side and top views of CrI_3 and Cr(I, X)_3 , respectively. The red arrows in (a) and (b) schematize the FM and helix spin configurations, respectively; (c) and (d) illustrate the eigenvectors $\{\alpha\beta\gamma\}$ of Kitaev interactions and the directions of DMI vectors of CrI_3 and Cr(I, X)_3 , respectively; (e) schematizes the DM vector being decomposed into its $\{r p z\}$ components.

sites. \mathcal{J} and \mathcal{A} are 3×3 matrices, for which the elements can be extracted using the four-state energy mapping method and DFT (see Supplemental Material (SM) [20] for details). The \mathcal{J} matrices can be further decomposed into an isotropic parameter J' , the symmetric Kitaev term K [15], and the antisymmetric DMI vector \mathbf{D} [21]; and the \mathcal{A} matrix simply reduces to its A_{zz} components by symmetry. The Hamiltonian can then be rewritten as

$$\mathcal{H} = \frac{1}{2} \sum_{n=1,2} \sum_{(i,j)_n} \{J'_n \mathbf{S}_i \cdot \mathbf{S}_j + K_n S_i^\gamma S_j^\gamma + \mathbf{D}_{n,ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)\} + \sum_i A_{zz} (S_i^z)^2, \quad (2)$$

where $J' = (J'_\alpha + J'_\beta)/2$, $K = J'_\gamma - J'$, and S^γ is the γ component of \mathbf{S} in a local basis $\{\alpha, \beta, \gamma\}$ that results from the diagonalization of the symmetric part of \mathcal{J} [see Figs. 1(c), 1(d) and SM [20] for details about Kitaev interaction). The directions of the DMI vectors \mathbf{D} are illustrated in Figs. 1(c) and 1(d). Specifically, the \mathbf{D}_1 vectors of both Cr(I, X)_3 are perpendicular to the Cr_2IX plane, which satisfy Moriya's rule [17] and thus testify to the accuracy of our calculations. The \mathbf{D}_2 vectors are basically pointing from one bridging I to the other I or X. Note that we further define another isotropic parameter as $J = (J_{xx} + J_{yy} + J_{zz})/3$, which is equivalent to the J coefficient involved in the commonly used $J\mathbf{S}_i \cdot \mathbf{S}_j$ term. Such isotropic J is thus different from J' and its ratio with the DMI parameter ($|D/J|$) can provide information about the existence of magnetic skyrmions or not.

Let us now look at the precise values of the magnetic parameters obtained from DFT and the four-state method [15,21,23,24]. As shown in Table I, both J_1 and J_2 yield negative values, which imply ferromagnetism (FM) for all investigated systems, which is consistent with the measured

TABLE I. Magnetic parameters of CrI_3 , Cr(I, Br)_3 , and Cr(I, Cl)_3 monolayers [22]. Note that $S = 3/2$ is used when extracting the magnetic parameters. The energy unit is in meV.

Mag. Para.	CrI_3	Cr(I, Br)_3	Cr(I, Cl)_3
J_1	-2.218	-1.800	-0.983
K_1	0.847	0.505	0.422
D_1	0	0.270	0.191
$ D_1/J_1 $	0	0.150	0.194
J_2	-0.638	-0.673	-0.754
K_2	0.078	0.075	0.094
D_2	0.135	0.114	0.138
$ D_2/J_2 $	0.212	0.169	0.183
A_{zz}	-0.262	-0.124	-0.029

ferromagnetism of CrX_3 , with $X = \text{Cl, Br, and I}$ [13,25,26]. The J_1 , K_1 , and A_{zz} parameters all decrease in magnitude when the X ion of Cr(I, X)_3 varies from I to Cl, via Br. This decrease in J_1 results from the shrinking of lattice constants [27], while that of K_1 and A_{zz} root in the weakening of the SOC strength (as consistent with the location of I, Br, and Cl in the periodic table). On the other hand, the J_2 and K_2 coefficients show no significant changes with the X ion. Moreover, the Janus monolayers Cr(I, Br)_3 and Cr(I, Cl)_3 exhibit remarkable D_1 values of 0.270 and 0.194 meV, respectively, while the prototype CrI_3 has no finite D_1 , as aforementioned. Such values, altogether with J_1 , yield large $|D_1/J_1|$ ratios of 0.150 for Cr(I, Br)_3 and 0.194 for Cr(I, Cl)_3 , which are within the typical range of 0.1–0.2 known to generate skyrmionic phases [2]. Note that a very recent theoretical study proposed the application of electric field to monolayer CrI_3 , in order to break the inversion center and induce DMI [28]. Although this method is clever, the electric field has a weak effect in generating DMI, as an extremely large electric field of 2 V/nm leads to a small $|D_1/J_1|$ of 0.071, which hinders the actual creation of skyrmions. Notably, our three investigated systems also show non-negligible D_2 values, which leads to $|D_2/J_2|$ ratios comparable to their 1st NN counterparts, as detailed in Table I. The significant $|D_1/J_1|$ and $|D_2/J_2|$ ratios of Cr(I, Br)_3 and Cr(I, Cl)_3 demonstrate that the fabrication of Janus monolayers is an effective way to induce considerable DMI and is thus promising to create magnetic skyrmions.

Another important factor that affects the formation of topological states is the MAE, which is defined as the energy difference between out-of-plane FM (zFM) and in-plane FM (xFM) states, $\epsilon = E_{z\text{FM}} - E_{x\text{FM}}$, where $\epsilon < 0$ (> 0 , respectively) favors out-of-plane (in-plane, respectively) FM. Supplemental Material Fig. S1(a) displays the MAE calculated using the model energy of Eq. (2) with the parameters of Table I [20]. The prototype CrI_3 , with $\epsilon = -0.693$ meV/Cr, favors strong out-of-plane anisotropy and Cr(I, Br)_3 , with $\epsilon = -0.218$ meV/Cr, exhibits mild out-of-plane anisotropy. In contrast, Cr(I, Cl)_3 shows weak in-plane anisotropy since $\epsilon = 0.111$ meV/Cr [29]. As we will see later, such differences in anisotropy between Cr(I, Br)_3 and Cr(I, Cl)_3 plays a role on the morphology of the spin textures.

Intrinsic domain wall skyrmion in Cr(I, Br)_3 . As detailed in Method section of SM, PTMC simulations using the

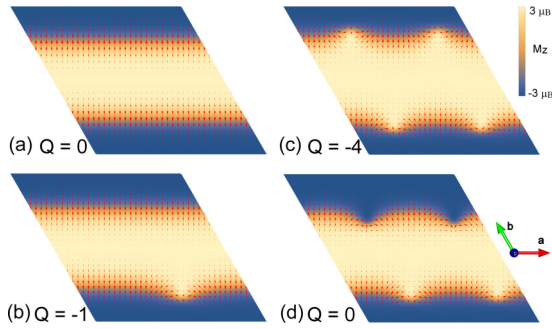


FIG. 2. Magnetic structures and topological charges of $\text{Cr}(\text{I}, \text{Br})_3$. (a) shows the out-of-plane Néel-type cycloid; (b) illustrates one DWS at one domain boundary; (c) and (d) each hosts four DWS and are also energetically degenerate but with different Q . The color map applies to the *out-of-plane* component of the spin vectors (“spin-in” and “spin-out,” for negative (dark) and positive (bright) z components, respectively) while the arrows characterize the *in-plane* components of the spin vectors.

Hamiltonian of Eq. (1) are performed over a $50 \times 50 \times 1$ supercell to find spin structures with low energies. These latter spin structures are then further relaxed with the conjugate gradient (CG) method, until arriving at energy minima. Such optimization scheme guarantees the converged spin structures to be either metastable states or the ground state at the temperature of 0 K.

Practically, the ground state of $\text{Cr}(\text{I}, \text{Br})_3$ is determined to be out-of-plane ferromagnetism (zFM), of which the energy is set to be zero. The first metastable state [the convention of terminology here is that the n th metastable state has the $(n + 1)$ th lowest energy] of $\text{Cr}(\text{I}, \text{Br})_3$ is an out-of-plane Néel-type cycloid structure, as shown in Fig. 2(a), with an energy of 0.056 meV/Cr. Its propagation direction is symmetrically equivalent along $(\mathbf{a} + 2\mathbf{b})$, $(2\mathbf{a} + \mathbf{b})$, and $(\mathbf{b} - \mathbf{a})$. Moreover, the in-plane components of the spin vectors and the propagation direction form angles of $\sim 5.9^\circ$ around the middle of domains (bright and dark zones) and $\sim 3.2^\circ$ near the domain walls. Such cycloid structure yields no finite topological charge Q that is defined as $Q = \frac{1}{4\pi} \int \mathbf{m} \cdot (\frac{\partial \mathbf{m}}{\partial x}) \times (\frac{\partial \mathbf{m}}{\partial y}) dx dy$, where \mathbf{m} is the unit vector lying along the local magnetic moment’s direction.

On the other hand, novel topological states with finite Q are found as the second and even higher metastable states. As shown in Fig. 2(b), near the lower bright-dark domain boundary, the spin vectors not only behave as the aforementioned Néel-type cycloid, but also rotate clockwise in the film plane and form a point defect. The in-plane components of such defect is reminiscent of a domain wall skyrmion (DWS) [30], or precisely, a slice of the latter. The in-plane rotation, together with the out-of-plane cycloidal behavior, renders $Q = -1$ and is thus topologically equivalent to the common axially symmetric skyrmion [4–7]. We thus refer to this special defect as a DWS for simplicity. Such DWS state of Fig. 2(b) has an energy of 343.5 meV/DWS with respect to the ground state and 63.8 meV/DWS with respect to the cycloid phase.

Moreover, it is found that either the upper or the lower domain boundary can host multiple DWS, as shown in Fig. 2(c). The $50 \times 50 \times 1$ supercell can thus host up to four DWS. Interestingly, when the in-plane components of the spin vectors

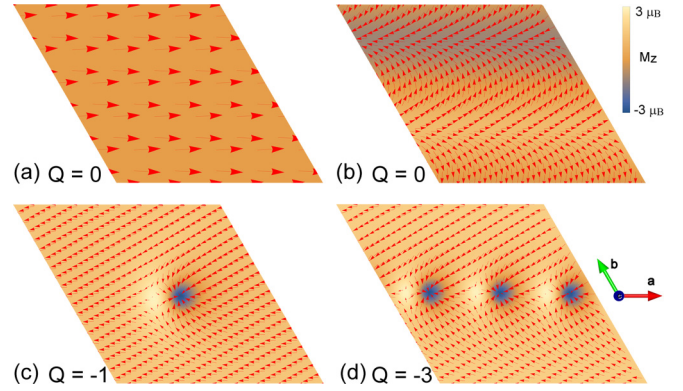


FIG. 3. Magnetic structures and topological charges of $\text{Cr}(\text{I}, \text{Cl})_3$. (a) shows the in-plane zigzag-canted FM state; (b) illustrates the in-plane cycloidal structure; (c) and (d) display one and three bimerons, respectively, with an out-of-plane magnetic field B of 0.8 T. See Fig. 2 for explanations of the color map and the vectors.

rotate anticlockwise, it forms a domain wall antiskyrmion, with $Q = +1$. As shown in Fig. 2(d), the upper domain boundary hosts two antiskyrmions and the lower one hosts two skyrmions, which results in the total topological charge being $Q = 0$. Interestingly, either a skyrmion or an antiskyrmion is found to cost an energy of 63.8 meV/DWS with respect to the cycloid state. In fact, the energy is linearly increasing with simply the number of skyrmions, as shown in Fig. S2(c). It is also found here that skyrmions and antiskyrmions cannot exist at the same domain wall, since they tend to annihilate each other, which would in fact lead to the transformation to the cycloidal state (resulting in $Q = 0$) [31].

In order to reveal the microscopic origin of the aforementioned spin textures, DMI vectors are decomposed into their $\{r pz\}$ components. As shown in Fig. 1(e), the $\hat{\mathbf{r}}$ axis is along the Cr-Cr bond direction and thus in-plane; the $\hat{\mathbf{p}}$ axis is perpendicular to $\hat{\mathbf{r}}$ and also in-plane, and the $\hat{\mathbf{z}}$ axis is the out-of-plane direction. As shown in Supplemental Material Table SII, for $\text{Cr}(\text{I}, \text{Br})_3$, this decomposition gives $\mathbf{D}_1 = (0, -0.226, -0.147)$ meV for the Cr-Cr pair that is along $(\mathbf{a} - \mathbf{b})/2$ and $\mathbf{D}_2 = (0.080, -0.040, -0.071)$ meV. As detailed in Sec. SII and Table SIII of the SM, by switching these components on and off in the simulations, it is found that (i) D_1^r and D_2^r both favor the Néel-type cycloid of Fig. 2(a), which is consistent with previous knowledge [32]; (ii) D_1^z favors in-plane canting, which results in the 3.2° to 5.9° in-plane angles of the cycloid; (iii) D_2^z leads to a slight energy gain when creating a DWS on a cycloid, due to the changes in out-of-plane components of spins; while (iv) K_1 and A_{zz} are the driving forces to create a DWS on a cycloid, since they favor the extra out-of-plane components with respect to the perfect cycloid, which explains the energetic degeneracy between a clockwise and an anticlockwise DWS.

Magnetic-field-induced bimeron states in $\text{Cr}(\text{I}, \text{Cl})_3$. As consistent with its positive MAE, the ground state of $\text{Cr}(\text{I}, \text{Cl})_3$ is determined to be in-plane zigzag-canted FM, as shown in Fig. 3(a). The spin vectors at the A and B sites of the honeycomb lattice make an angle of 6.7° , which is in good agreement with the 6.2° that is directly obtained from DFT on a unit cell. Comparing to the xFM state, such canting

renders an in-plane perpendicular moment of $0.15\mu_B/\text{Cr}$, with an energy lowering of $0.024\text{ meV}/\text{Cr}$. Such zigzag-canted FM state is degenerate when the main component of the magnetization is along the \mathbf{a} direction (which is precisely our x direction), or along the \mathbf{b} and $(\mathbf{a} + \mathbf{b})$ directions.

The first metastable state is a mostly in-plane cycloidal structure superimposed with small out-of-plane components forming a waved spin pattern, as shown in Fig. 3(b) [33]. Note also that the zigzag canting of the ground state exists in such cycloid phase too. Such cycloidal structure has an energy higher by $0.026\text{ meV}/\text{Cr}$ from the ground state and is degenerate for propagation directions being along the $(\mathbf{a} + 2\mathbf{b})$, $(2\mathbf{a} + \mathbf{b})$, or $(\mathbf{b} - \mathbf{a})$ directions.

Skyrmionic states are not found to be “naturally” stable in $\text{Cr}(\text{I}, \text{Cl})_3$ within our Hamiltonian, partially due to the relatively strong in-plane anisotropy. An out-of-plane magnetic field B is then applied to compensate the in-plane anisotropy that arises from the original DFT-derived magnetic parameters (K and A_{zz}). It is numerically found that, for $B > 0.65\text{ T}$, zFM is energetically more favorable than in-plane FM. However, the magnetic field ($< 2\text{ T}$) does not polarize the spins to the fully out-of-plane direction, but rather renders the in-plane zigzag-canted FM now possessing an out-of-plane component. The previously metastable cycloid state cannot survive and transform to such FM state when $B > 0.2\text{ T}$.

Interestingly, topologically nontrivial bimeron states can be stabilized in $\text{Cr}(\text{I}, \text{Cl})_3$ with a compensative magnetic field between 0.5 and 1.3 T . A meron/antimeron can be viewed as half of a skyrmion/antiskyrmion (and thus has a half integer topological charge Q) embedded in a background made of in-plane dipoles [12,34]. Note also that a bimeron [35–38], which is a pair of merons or antimérons, is sometimes referred to as asymmetric skyrmion [39], double-pair skyrmion [40], or in-plane skyrmion [3,41] too. Although micrometer-sized merons have been widely reported in permalloys [42–45], merons with nanometric sizes have only been very recently observed in $\text{Co}_8\text{Zn}_9\text{Mn}_3$ [12]. The merons/bimerons not only induce emergent electromagnetism and thus novel Hall effects [1], but are also promising for the next generation of memory devices [43]. Practically, for a magnetic field of 0.8 T and as shown in Fig. 3(c), we identify two merons, one being a core-down vortex and the other one being a core-up antivortex. Such two merons form a close pair with a diameter of $\sim 10.5\text{ nm}$ and each has $Q = -\frac{1}{2}$. Such meron pair is thus precisely a bimeron with $Q = -1$. Due to the energy lowering from magnetic field, this bimeron state has an energy of $-63.7\text{ meV}/\text{bimeron}$ with respect to the zigzag FM phase of Fig. 3(a), while it is still metastable with an energy of $74.4\text{ meV}/\text{bimeron}$ with respect to ground state under 0.8 T magnetic field. Moreover, it is found that up to three bimerons

can be stabilized within the $50 \times 50 \times 1$ supercell and the energy also linearly increases with the topological charges in $\text{Cr}(\text{I}, \text{Cl})_3$, as shown in Fig. S2(c).

Again, we decompose DMI vectors into their $\{r, p, z\}$ components to investigate their specific effects on spin textures. As shown in Table SII, $\text{Cr}(\text{I}, \text{Cl})_3$ possesses $\mathbf{D}_1 = (0, -0.154, -0.113)\text{ meV}$ and $\mathbf{D}_2 = (0.095, -0.054, -0.085)\text{ meV}$. As detailed in Sec. SII and Table SIII of the SM, it is determined that (i) D_1^z leads to the zigzag canting in both the ground state of Fig. 3(a) and the cycloid phase of Fig. 3(b); (ii) D_1^p and D_2^p not only result in the out-of-plane waved spin pattern of the cycloid phase of Fig. 3(b), but also work together with the anisotropy and result in the bimerons of Figs. 3(c) and 3(d) (note that such anisotropy arises from interplay among SIA, Kitaev interaction, and the magnetic field); while (iii) other DMI components have either negligible or even no contributions.

In summary, we propose the fabrication of $\text{Cr}(\text{I}, X)_3$ ($X = \text{Br}, \text{Cl}$) Janus monolayers to induce large DMI and subsequent topological spin states. By combining DFT and MC simulations, we find that $\text{Cr}(\text{I}, \text{Br})_3$ can intrinsically host metastable domain wall skyrmion phases, while a bimeron state of $\text{Cr}(\text{I}, \text{Cl})_3$ can be stabilized by applying an out-of-plane magnetic field. The Kitaev interaction is found critical to stabilize DWS and bimerons, as it contributes to the MAE and adds to the disorder of the systems (see SM for details). Our study thus suggests a feasible approach to create topological spin textures in semiconducting magnets consisting of chromium trihalide Janus monolayers. Such presently predicted topological phases are not only useful for memory and logic devices, but can also be promising for energy storage [46].

Note added. Recently, we became aware of two recent theoretical works on prediction of skyrmionic states in MoS_2 -prototyped Janus monolayers of MnXY ($X, Y = \text{S}, \text{Se}$ and Te) [61,62].

Acknowledgments. This work is primarily supported by the Office of Department of Energy, Basic Energy Sciences under Contract No. ER-46612 (C.X. and L.B.). S.P. and Y.N., who contributed to the topological analysis, also acknowledge the financial support from DARPA Grant No. HR0011727183-D18AP00010 (TEE Program). H.X. is supported by NSFC (11825403), the Special Funds for Major State Basic Research (2015CB921700), Program for Professor of Special Appointment (Eastern Scholar), Qing Nian Ba Jian Program, and Fok Ying Tung Education Foundation. J.F. acknowledges the support from Anhui Provincial Natural Science Foundation (1908085MA10). The Arkansas High Performance Computing Center (AHPCC) is also acknowledged.

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