Multifunctional two-dimensional van der Waals Janus magnet Cr-based dichalcogenide halides

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Two-dimensional van der Waals Janus materials and their heterostructures offer fertile platforms for designing fascinating functionalities. Here, by means of systematic first-principles studies on van der Waals Janus monolayer Cr-based dichalcogenide halides CrYX (Y = S, Se, Te; X = Cl, Br, I), we find that CrSX (X = Cl, Br, I) are the very desirable high Tc ferromagnetic semiconductors with an out-of-plane magnetization. Excitingly, by the benefit of the large magnetic moments on ligand S2⁻ ions, the sought-after large-gap quantum anomalous Hall effect and sizable valley splitting can be achieved through the magnetic proximity effect in van der Waals heterostructures CrSBr/Bl₂Se₃/CrSBr and MoTe₂/CrSBr, respectively. Additionally, we show that large Dzyaloshinskii–Moriya interactions give rise to skyrmion states in CrTeX (X = Cl, Br, I) under external magnetic fields. Our work reveals that two-dimensional Janus magnet Cr-based dichalcogenide halides have appealing multifunctionalities in the applications of topological electronic and valleytronic devices.

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INTRODUCTION

The discovery of ferromagnetism at a finite temperature in atomically thin van der Waals (vdW) monolayers (MLs)1–3 has spurred a surge of experimental and theoretical interests in understanding the two-dimensional (2D) magnetism and furthermore in producing emergent functionalities in 2D vdW heterostructures4–9. Fundamentally, the well-known Mermin–Wagner theorem based on the isotropic Heisenberg exchange model10 suggested the absence of either FM or antiferromagnetic (AFM) order in 2D systems at nonzero temperature, and hence the early findings of FM order in CrI₃ ML1 and Cr₂Ge₂Te₆ bilayer11 were rather surprising. Recent studies indicated that magnetic anisotropies play a major role in the establishment of their long-range magnetic ordering. The main advantage of these 2D magnetic vdW materials is their integrability with other vdW functional materials. Through the magnetic proximity effect, many spintronic, valleytronic, and optoelectronic properties can be achieved in heterostructures by stacking diverse 2D vdW MLs or ultrathin films5,12,13. For example, the quantum anomalous Hall effect (QAHE) and axion insulator phase can be realized by integrating 2D FM semiconductors with thin films of topological insulators14–16. When transition metal dichalcogenide (TMDC) MLs are in contact with 2D FM semiconductors, a valley splitting can be produced for valleytronic and optoelectronic manipulations17,18, as recently predicted by first-principles calculations. This is partially inspired by the experimental observation of high-temperature (~200 K) ferromagnetism in 1T-CrTe₃ ML18–20. We identify that CrSX (X = Cl, Br, I) are the very attractive out-of-plane FM semiconductors with high Curie temperatures (~176 K) and large magnetic moments on S²⁻ ions. Excitingly, through the magnetic proximity effect, the sought-after large-gap QAHE can be achieved in CrSBr/Bl₂Se₃/CrSBr. For the same reason, a sizable valley splitting of 37.9 meV, corresponding to a magnetic field of 379 T21, can be generated in MoTe₂/CrSBr. Furthermore, we find that large DMI leads to magnetic skyrmion states in CrTeX (X = Cl, Br, I) under an appropriate external magnetic field by virtue of Monte Carlo (MC) simulations. Our work highlights the remarkable multifunctionalities of these 2D vdW Janus ML magnets that are promising for applications in the next-generation topotronic and valleytronic devices.
RESULTS

Electronic and magnetic properties of CrYX MLs

Figure 1a shows the crystal structure of Janus ML CrYX (Y = S, Se, Te; X = Cl, Br, I). Magnetic CrX3+ cations form a triangular lattice and are sandwiched by Y2− and X− anions. Similar to CrI3 and CrGe2Te5, MLs 1,11, CrX3+ cations in CrYX are surrounded by the distorted edge-shared octahedrons formed by Y2− and X− anions, which implies FM nearest-neighbor (NN) exchange interactions. As a direct result of different sizes of Y2− and X− anions, CrSCI and CrTeI have the smallest and largest in-plane lattice constants (Supplementary Table 1), respectively. Since no imaginary frequency is found in the calculated phonon spectra of all CrYX MLs (Supplementary Fig. 1), they should be dynamically stable.

To explore the magnetic ground states of CrYX MLs, we consider a spin Hamiltonian consisting of Heisenberg exchange interactions, DMI and single ion anisotropy (SIA). This spin Hamiltonian is in the form of

\[
H = J_1 \sum \langle i,j \rangle \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum \langle i,j \rangle \mathbf{S}_i \cdot \mathbf{S}_j \nonumber + J_3 \sum \langle i,j \rangle \mathbf{S}_i \cdot \mathbf{S}_j + \sum \mathbf{D}_i \cdot (\mathbf{S}_i \times \mathbf{S}_j) - A \sum \langle i \rangle \mathbf{S}_i^2.
\]

Here, \(\mathbf{S}_i\) is the normalized spin vector at site \(i\); \(J_1, J_2, J_3\) are NN, second-NN, and third-NN Heisenberg exchange parameters, respectively; \(\mathbf{D}_i = (D_x, D_y, D_z)\) is the NN DMI vector and \(A\) is the SIA parameter. The Heisenberg exchange parameters \(J_i (i = 1, 2, 3)\), NN DM vector \(\mathbf{D}_i\) and SIA parameter \(A\) from DFT calculations are tabulated in Supplementary Table 2. As expected, the NN \(J_1\) is FM and dominates over the second-NN \(J_2\) and third-NN \(J_3\) for all CrYX (Fig. 1b). Since DMI is directly related to the spin–orbit coupling (SOC) magnitude of the NN DMI vector, \(|\mathbf{D}_i| = \sqrt{D_x^2 + D_y^2 + D_z^2}\) increases when \(Y(X)\) goes from S (Cl) to Te (I), as shown in Fig. 1c. It is interesting that \(|\mathbf{D}_i|/|\mathbf{J}_i|\) of the in-plane component of \(\mathbf{D}_i\) has a similar trend to \(|\mathbf{J}_i|\) when \(Y(X)\) varies (Fig. 1c). Particularly, \(|\mathbf{D}_{i,ij}|/|\mathbf{J}_i|\) of CrTeX (X = Cl, Br, I) is in the typical range of 0.1–0.2 that is known to generate magnetic skyrmions37. Lastly, CrSX (X = Cl, Br, I) and CrSeI have an out-of-plane SIA while the other five of CrYX have an in-plane SIA (Fig. 1d). Note that the present results of NN FM Heisenberg exchange interactions and in-plane SIA of CrSeBr and CrTeI Janus MLs are consistent with those obtained in a previous theoretical study 28.

Owing to the competition between Heisenberg exchange interactions, DMI and SIA, MC simulations reveal that CrYX exhibits very rich magnetic ground configurations (Fig. 1e). It has been reported that magnetic ground state of 2D magnets is mainly determined by a critical dimensionless factor \(|A/K|/D^2\) \((K, D)\) the stiffness parameter originating from FM Heisenberg exchange interactions)39. A large \(|A/K|/D^2\) yields a FM ground state, while a small one results in a spiral ground state38. First, CrSyX \((Y = Cl, Br, I)\) MLs have small DMI, out-of-plane SIA and hence an out-of-plane FM ground state with a Curie temperature up to 176 K (Supplementary Fig. 2). CrSeCl also has a FM ground state, but its magnetization is in plane due to its negative SIA. Second, CrSeBr and CrSeI have medium DMI, small SIA, and hence small \(|A/K|/D^2\). Therefore, they have a spin spiral ground state with a large periodic length. Finally, CrTeX \((X = Cl, Br, I)\) MLs have both large DMI and SIA. Their magnetic ground states have wormlike domains, similar to what was found in the previous studies of 2D Janus manganese dichalcogenides 29.

Considering the needed semiconducting nature of vdW ferromagnets for engineering emergent physical properties via the magnetic proximity effect in heterostructures 7,13,16, we first analyze the electronic properties of CrSX \((X = Cl, Br, I)\) MLs which have a FM ground state and out-of-plane magnetic anisotropy. Band structures in Fig. 2a–c show that all CrSX MLs are indirect-gap semiconductors. As the electronegativity weakens from Cl to Br and to I, the band gaps of CrSX decrease from 2.17 eV (CrSBr) to 1.77 eV (CrSBr) and to 0.81 eV (CrSI). The curves of density of state (Supplementary Fig. 3) suggest that the conduction bands mainly come from the d states of Cr while the valence bands have mixtures of Cr, S, and X states. Besides, the d–p hybridization between Cr and S atoms is stronger than that between Cr and X atoms near the Fermi level.

As the magnetic ions are mostly covered by nonmagnetic anions in vdW FM semiconductors, the spin polarization of the outer layer is typically very small. The magnetic proximity effect is hence weak.
in most vdW heterostructures. Interestingly, the magnetic moments on $S^2^\text{−}$ anion are about 0.14 $\mu_B$/S in CrS$_X$ MLs, the largest for 2D FM semiconductors reported so far (Fig. 3c)\textsuperscript{1,11,40−45}. Especially, this magnetic moment is much larger than that of $I^2^\text{−}$ anions in CrI$_3$\textsuperscript{7,13,16,46−49}, suggesting that CrS$_X$ may have a significant magnetic proximity effect on other vdW functional materials.

**Large-gap QAHE in CrSBr/Bi$_2$Se$_3$/CrSBr vdW heterostructure**

We first examine the effect of CrS$_X$ MLs on magnetizing topological surface states (TSSs) of 3D topological insulators for the realization of QAHE\textsuperscript{8}. We construct an vdW heterostructure with two CrSBr MLs and a six quintuple-layer Bi$_2$Se$_3$ (BS) thin film (CrSBr/BS/CrSBr). The calculated binding energies of different stacking configurations suggest that Cr$^{3+}$ cations of CrSBr prefer to align with Bi and the $S^2^\text{−}$ side contacts BS thin film (Fig. 3a and more details in Supplementary Fig. 4). Our ab initio molecular dynamics simulations show that this CrSBr/BS/CrSBr heterostructure is thermodynamically stable (Supplementary Fig. 5). The induced spin polarization on BS penetrates through the film (Fig. 3b), with a strength twice as large as that in CrI$_3$/BS/CrI$_3$\textsuperscript{13}. Figure 3c shows the band structure of CrSBr/BS/CrSBr when the magnetizations of top and bottom CrSBr MLs are ferromagnetically ordered. As a result of the strong magnetization in CrSBr, the system has a large band gap up to 19 meV at the $\Gamma$ point, indicating its efficient magnetic proximity effect on the TSSs of BS thin film. By examining the spin components of the four bands near the Fermi level (Fig. 3d), we find that the two bands below (above) the Fermi level have the same spin-down (spin-up) components. These features clearly suggest the TSSs of the BS thin film are strongly magnetized by CrSBr.

To investigate the topological property of CrSBr/BS/CrSBr, we fit its band structure using an effective four-band model. With the bases of $\{ |s, \uparrow\rangle, |s, \downarrow\rangle, |b, \uparrow\rangle, |b, \downarrow\rangle \}$, the model Hamiltonian of inversion symmetric heterostructures is written as\textsuperscript{50}

$$H(k_x, k_y) = i k^2 + \begin{bmatrix} v_F (k_y \sigma_x - k_x \sigma_y) & M_k \sigma_0 \\ M_k \sigma_0 & -v_F (k_y \sigma_x - k_x \sigma_y) \end{bmatrix} + \begin{bmatrix} \Delta \sigma_z & 0 \\ 0 & \Delta \sigma_z \end{bmatrix}.$$  \hspace{1cm} (2)
and conduction bands of MoTe2 are not much affected by CrSBr, proposed in ref. 13, CrSBr/BS/CrSBr is a Chern insulator with Chern numbers 1.0 and 10.4 meV, respectively. According to the general rule that a high temperature can be achieved in CrSBr/BS/CrSBr, (19 meV, corresponding to 220 K), it is conceivable that QAHE with the FM semiconductor CrSBr ML and the large nontrivial band gap of MoTe2 has a nonzero anomalous Hall conductivity, $\sigma_{xy}$, when Fermi level lies between the valence band maxima of K+ and K valleys (Fig. 4d). Taking together the anomalous Hall conductivity and sizable valley splitting, it is conceivable that a spin- and valley-polarized Hall current can be generated in CrSBr/MoTe2 when applying an in-plane electric field, because of the strong SOC of Te and the symmetry reduction. This is a very attractive feature for diverse applications as discussed in the literatures for the studies of other magnetic systems.

To quantitatively determine the valley splitting in CrSBr/MoTe2, we adopt an energy scale, $\Delta^{\text{val}}_i = E_i^{(\text{val})} - E_i^{(\text{con})}$. Here, $\nu$ (c) denotes valence (conduction) bands; $K_\pm$ are distinguished by index $\tau = \pm$. According to this definition, $\Delta^{\text{val}}_{\text{crsbr}}$, $\Delta^{\text{val}}_{\text{mo}}, \Delta^{\text{con}}_{\text{crsbr}}$, and $\Delta^{\text{con}}_{\text{mo}}$ are estimated as $-37.9, -8.8, 8.6$, and $8.7$ meV, respectively. We see that $\Delta^{\text{val}}_{\text{crsbr}}$ in CrSBr/MoTe2 is sizable, corresponding to the valley splitting generated by a magnetic field of $379$ T. It is worth noting that this value is much larger than the counterparts in CrI3/WSe2 and CrI3/MoTe2. More remarkably, the smallest energy for the band edge vertical optical transition without spin flip in two valleys reaches a giant value of $46.5$ meV. Thanks to the time reversal symmetry breaking and the nonvanishing Berry curvature, CrSBr/MoTe2 has a nonzero anomalous Hall conductivity, $\sigma_{xy}$, when Fermi level lies between the valence band maxima of K+ and K valleys (Fig. 4d). Taking together the anomalous Hall conductivity and sizable valley splitting, it is conceivable that a spin- and valley-polarized Hall current can be generated in CrSBr/MoTe2 when applying an in-plane electric field, thus providing applications in valleytronics.

**DISCUSSION**

Finally, we find that CrTeX ($X = \text{Cl, Br, I}$) MLs can host magnetic skyrmions in an external magnetic field, because of the strong SOC of Te and the symmetry reduction. This is a very attractive feature for diverse applications as discussed in the literatures for the studies of other magnetic systems.

To characterize the presence of magnetic skyrmions in MC simulations, we calculate the topological charge $Q$ which is defined as:

$$Q = \frac{1}{4\pi} \int \mathbf{m} \cdot \left( \frac{\partial \mathbf{m}}{\partial x} \times \frac{\partial \mathbf{m}}{\partial y} \right) \, dx \, dy.$$
In Eq. (3), \( \mathbf{m} \) is a normalized magnetization vector; \( x \) and \( y \) are in plane coordinates. On a discrete spin lattice, Eq. (3) is evaluated by summing over the solid angle \( \Omega \) of three spins according to the Berg formula. Supplementary Fig. 8 shows the topological charge \( Q \) of CrTeI as a function of temperature (T) and out-of-plane external magnetic field (B). Through examining the spin textures under different \( T \) and \( B \), we find that the red area with large negative value of \( Q \) corresponds to the formation of magnetic skyrmion lattices in CrTeI. Because of the strong DMI, magnetic skyrmion lattices may exist in a large \( T-B \) parameter space, with \( T \) up to 80 K and \( B \) from 1 to 8 T. CrTeI and CrTeBr also form magnetic skyrmion lattices but in a smaller \( T-B \) region (Supplementary Fig. 9). Hence, we recommend CrTeML as the most promising 2D platform for the realization of magnetic skyrmions.

In summary, based on systematical first-principles studies on vdW Janus ML CrXY (\( Y = S, Se, Te; X = Cl, Br, I \)), we find that CrSX (\( X = Cl, Br, I \)) are useful FM semiconductors with high Curie temperatures up to 176 K and large induced magnetic moments on the ligand \( S^{2-} \) anions. Remarkably, the long-sought QAHE with a large gap of 19 meV and a sizable valley splitting of 37.9 meV are achieved through the magnetic proximity effect in vdW heterostructures CrSBr/BrS/CrSBr and MoTe2/CrSBr, respectively. Furthermore, CrTeX (\( X = Cl, Br, I \)) may host magnetic skyrmion states under external magnetic fields. Our work unveils the promising multifunctionalities of 2D vdW Janus magnet Cr-based dichalcogenide halides and reveals their potential for diverse applications in topotronic and valleytronics devices.

**METHODS**

**First-principles calculations**

Our first-principles calculations based on the density functional theory (DFT) are performed using the Vienna Ab initio Simulation Package with the generalized gradient approximation. Core-valence interactions are described by projector-augmented wave pseudopotentials. We utilize an energy cutoff of 350 eV for the plane-wave expansion and fully relax lattice constants and atomic positions until the force acting on each atom is smaller than 0.01 eV Å\(^{-1}\). To take into consideration the strong correlation effect among Cr 3d electrons, we adopt \( U = 3.0 \) eV and \( J_{H} = 0.9 \) eV\(^{-1}\). As discussed in Supplementary Note 8, we obtain similar results when different \( U \) values are employed. In building vdW heterostructures, we use an inplane lattice constant \( a_{0} = 4.16 \) Å of the relaxed bulk BS for CrSBr/BS/CrSBr and \( d_{AB} = 3.55 \) Å of MoTe2 (MT) ML for MoTe2/CrSBr. When relaxing the first Brillouin zone is sampled by \( 12 \times 12 \times 1 \), \( 6 \times 6 \times 1 \), and \( 12 \times 12 \times 1 \) centred Monkhorst-Pack \( k \) meshes for CrXY, CrSBr/BS/CrSBr, and MoTe2/CrSBr, respectively. We add a vacuum space of 12 Å between slabs along the normal axis to eliminate the spurious interactions. To obtain the accurate magnetic anisotropy energies of CrXY that arise from SIA, a very dense \( \Gamma \)-centered Monkhorst-Pack \( k \) mesh of \( 24 \times 24 \times 1 \) is used to sample the first Brillouin zone and the total energy convergence criterion is set to be \( 10^{-7} \) eV. Magnetic anisotropy energies are determined by computing the total energy difference with magnetic moments of Cr\(^{3+} \) ions being parallel and perpendicular to the plane of the CrXY ML. When calculating magnetic anisotropy energies, SOC is explicitly included in self-consistent loops. To correctly describe the weak interaction across the vdW gap in these heterostructures, we employ the nonlocal vdW functional (optB86b-vdW)\(^{63,64}\). Berry curvatures and chiral edge states are calculated by the wannier90\(^{65}\) and WannierTools\(^{66}\).

**DATA AVAILABILITY**

All data used in this study are available from the corresponding author upon reasonable request.

**CODE AVAILABILITY**

The central codes used in this paper are VASP, Wannier90, and WannierTools. The Detailed information related to the license and user guide for these codes are available at https://www.vasp.at, http://www.wannier.org and http://www.wanniertools.com.
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AUTHOR CONTRIBUTIONS
Y.H. and R.W. conceived the whole project. Y.H. and L.Q. carried out the DFT calculations. F.X. performed the MC simulations. All authors made contribution to the final version of the manuscript.

COMPETING INTERESTS
The authors declare no competing interests.

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