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Supplementary Information for "Electronic properties, skyrmions and bimerons in Janus CrXY (X, Y = S, Se, Te, Cl, Br, I, and X $\neq$ Y) monolayers"

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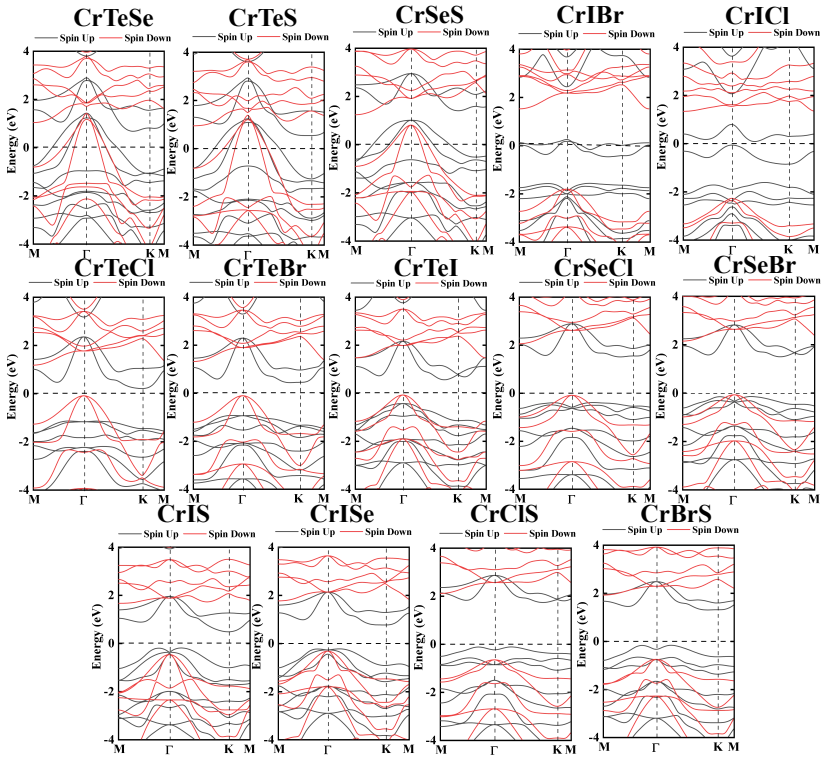

Fig. S1 Band structures of CrXY monolayers with PBE+U.

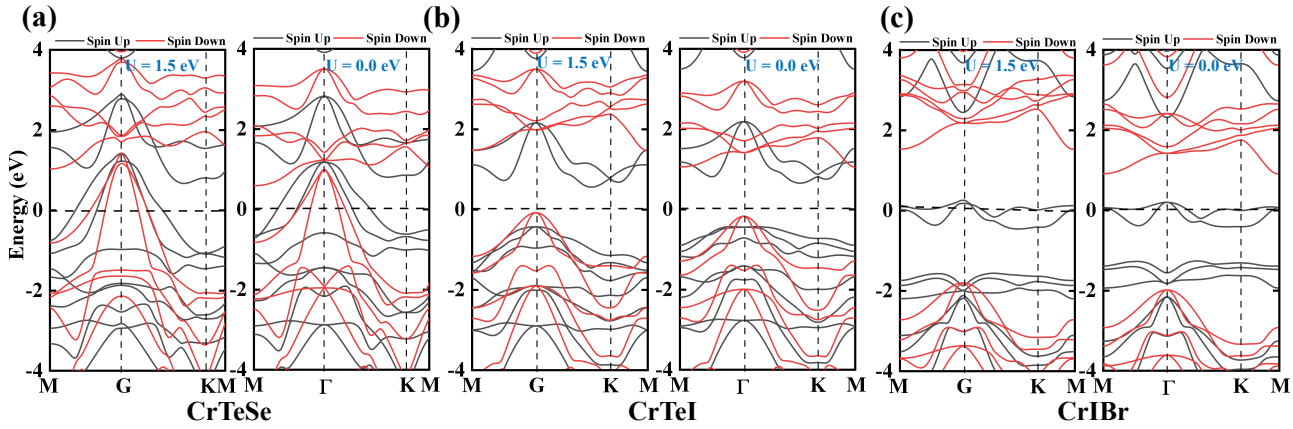

Fig. S2 Band structures by using PBE with U = 1.5 and 0.0 eV of (a) CrTeSe, (b) CrTeI and (c) CrIBr, respectively.

For the Heisenberg exchange coefficients J, we can obtain the values by the following four equations:

$$E_1 = 18J_1 + 18J_2 + 18J_3 + E_0, \tag{S1}$$

$$E_2 = -6J_1 - 6J_2 + 18J_3 + E_0, \tag{S2}$$

$$E_3 = -6J_1 + 2J_2 + 2J_3 + E_0, \tag{S3}$$

$$E_4 = -2J_1 - 6J_2 + 2J_3 + E_0. \tag{S4}$$

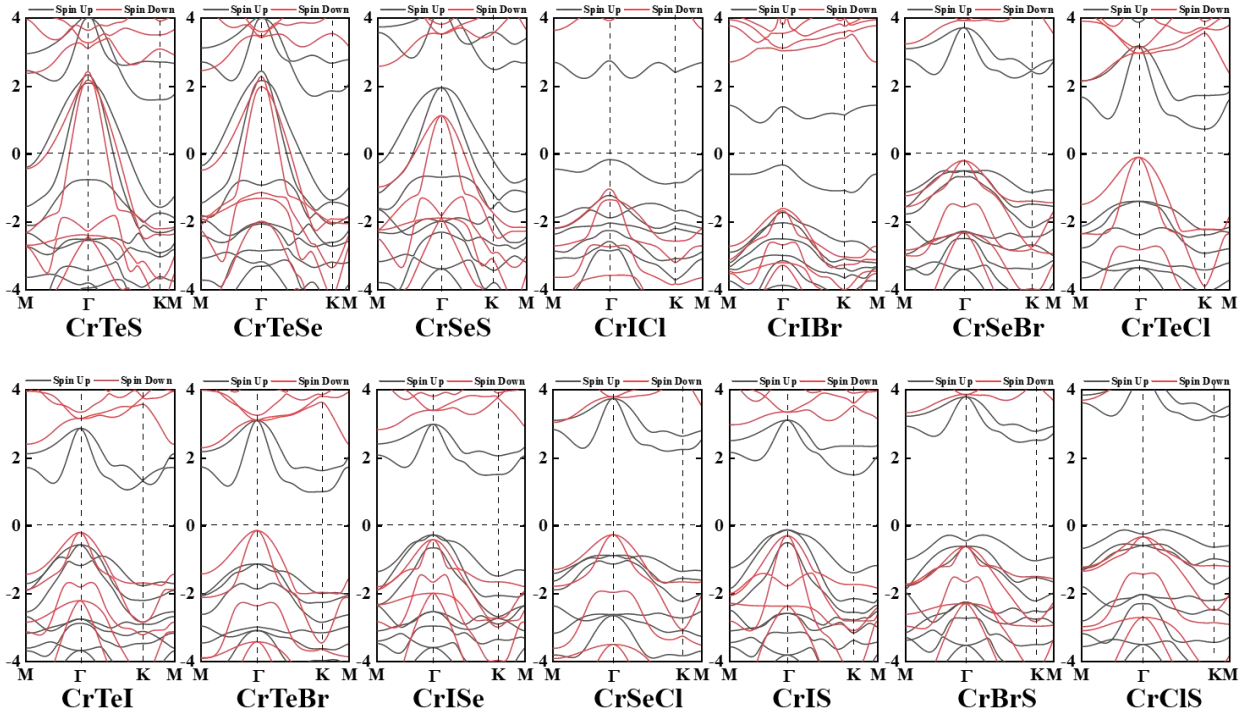

Fig. S3 Band structures of CrXY monolayers with HSE06.

Here, E_1, E_2, E_3, E_4 are the energies with four different spin configurations and E_0 is the spin-independent energy. The J_1, J_2, J_3 are the Heisenberg exchange coefficients of the first, second and third nearest neighbors as shown in Fig.S6 and the values are shown in Table S5.

We have calculated the single-ion anisotropy coefficient (K_{single}), MAE constant (K_i) and effective anisotropy (K_{eff}) of CrXY as shown in Table Rs4. K_{eff} can be expressed as:

$$K_{\mathrm{eff}} t_{\mathrm{eff}} = K_i - \frac{1}{2}\mu_0 M_s^2 t_{\mathrm{eff}}, \tag{S5}$$

where K_{eff} is the effective anisotropy per unit volume, t_{eff} is the thickness of the ferromagnetic layer, μ_0 is the magnetic constant, and M_s is the saturation magnetization per unit volume [1].

The formation energy (E_f) is defined by:

$$\Delta H_{\mathrm{f}} = E_{\mathrm{tot}} - \sum_{\mathrm{i}} \mu_{\mathrm{i}} x_{\mathrm{i}}, \tag{S6}$$

where E_{tot} is the DFT total energy of the compound, μ_i is the chemical potential of element i and x_i is the quantity of element i in the compound [2].

Table S1 The energies with respect to the Fermi level of crystal field splittings of the Cr d orbitals, the X (Y) p orbitals and pd hybridization.

	Energy (eV)									
CrXY	Cr-d (x^2-y^2, xy)	Cr-d z^2	Cr-d (xz, yz)	$pd\pi$ (x^2-y^2, xy)	$pd\sigma$ z^2	pd (xz, yz)	X-p z	X-p (x, y)	Y-p z	Y-p (x, y)
CrTeSe	−2.94	−2.54	−1.83	(2.89, 2.78)	−0.98	(1.42, 1.22)	−2.15	(1.16, 1.41)	−4.79	(−1.65, −1.50)
CrTeI	−2.92	−2.03	−1.92	2.15	−0.98	−0.45	−1.56	−0.1	−1.93	−4.4
CrIBr	−4.05	−5.59	−1.89	0.24	−1.89	0.24	−3.03	−1.82	−5.21	−3.41

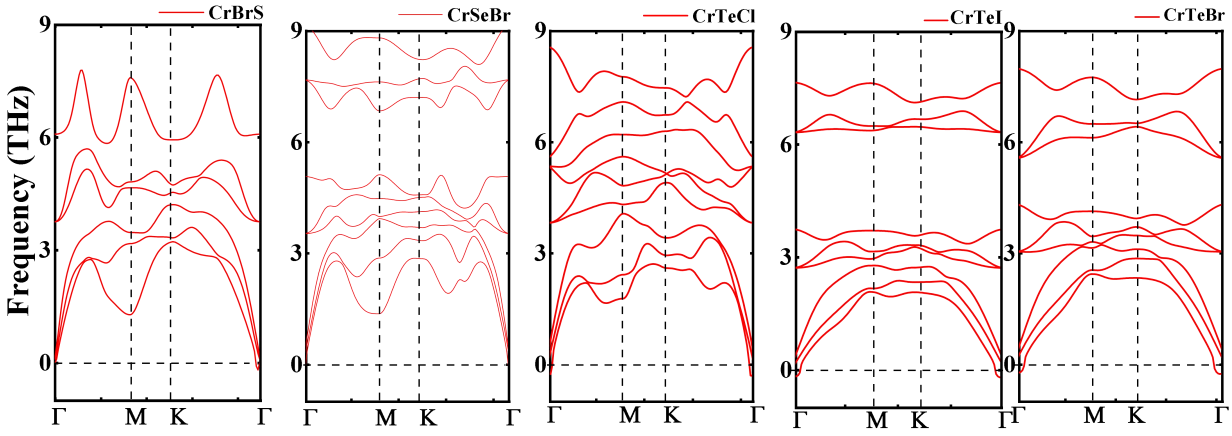

Fig. S4 Phonon spectra of Janus CrBrS, CrSeBr, CrTeCl, CrTeI and CrTeBr monolayers, respectively.

Table S2 Charge transfer of CrXY monolayers. Negative values indicate electron gain, and positive values represent electron loss.

	CrSeS	CrSCl	CrSeCl	CrTeS	CrTeSe	CrSeBr	CrBrS	CrTeCl	CrTeBr	CrICl	CrIS	CrISe	CrIBr	CrTeI
Cr	−1.16	−1.34	−1.22	−1.09	−0.96	−1.15	−1.28	−1.06	−1.01	−1.14	−1.20	−1.08	−1.08	−0.93
X	0.47	0.62	0.61	0.35	0.37	0.62	0.53	0.41	0.44	0.48	0.41	0.43	0.48	0.47
Y	0.69	0.72	0.61	0.74	0.59	0.53	0.75	0.65	0.57	0.66	0.79	0.65	0.6	0.46

Table S3 Formation energies ΔH_f (eV) of CrXY.

	CrSeS	CrSCl	CrSeCl	CrTeS	CrTeSe	CrSeBr	CrBrS	CrTeCl	CrTeBr	CrICl	CrIS	CrISe	CrIBr	CrTeI
ΔH_f	−1.47	−1.87	−1.58	−1.26	−0.96	−1.40	−1.66	−1.26	−1.10	−1.22	−1.44	−1.21	−0.97	−0.95

Table S4 Angles θ_1 and θ_2, the paths of superexchange interactions r_1, r_2, r_3, r_4 and the paths of direct interactions r_5 of nearest Cr atoms are shown in Fig.S5. The unit of θ and r are Å and degree, respectively.

	CrSeS	CrSeCl	CrSeBr	CrTeS	CrTeSe	CrTeCl	CrTeBr	CrTeI	CrClS	CrBrS	CrIS	CrISe	CrICl	CrIBr
θ_1	91.76	89.38	86.37	95.7	91.43	93.13	90.00	87.29	93.89	95.94	99.30	96.08	94.19	87.10
θ_2	81.61	90.65	92.70	75.62	78.38	86.66	88.66	91.86	86.92	83.97	81.67	83.90	88.11	93.95
r_1	2.32	2.50	2.63	2.33	2.46	2.53	2.66	2.82	2.34	2.63	2.40	2.52	2.43	2.68
r_2	2.32	2.50	2.63	2.31	2.45	2.53	2.66	2.82	2.34	2.63	2.40	2.52	3.09	2.67
r_3	2.54	2.47	2.49	2.80	2.78	2.68	2.69	2.71	2.49	2.62	2.79	2.80	3.09	2.84
r_4	2.54	2.47	2.49	2.81	2.79	2.68	2.69	2.71	2.49	2.62	2.79	2.80	2.74	2.84
r_5	3.33	3.52	3.60	3.44	3.62	2.67	3.75	3.89	3.42	3.50	3.65	3.75	4.06	3.91

Table S5 Optimized lattice constants a, electronic structure E.S., magnetic ground state G.S., magnetic anisotropic energy MAE, Heisenberg exchange coefficients J_1, J_2 and J_3, Curie temperature T_C and the nonlinear magnetic exchange energy DMI in CrXY monolayers.

	a (Å)	E.S.	Gap (eV)	G.S.	MAE (meV)	T_C (K)	J_1 (meV)	J_2 (meV)	J_3 (meV)	DMI (meV)
CrSeS	3.33	M	–	FM	−1.26	65.2	0.63	−4.93	−0.38	0.39
CrSeCl	3.52	S	1.60	FM	−0.16	198	−18.61	−1.19	2.44	−0.64
CrSeBr	3.60	S	1.57	FM	−0.08	247	−21.90	−1.08	2.07	−1.20
CrTeS	3.44	M	–	FM	0.95	137	−19.85	0.93	2.84	0.63
CrTeSe	3.52	M	–	FM	0.75	49.1	−9.12	0.34	1.63	2.03
CrTeCl	3.67	S	0.30	FM	−2.19	104	−11.38	−2.21	7.41	−1.07
CrTeBr	3.75	S	0.55	FM	−1.76	60.2	−16.80	−1.88	6.26	−1.81
CrTeI	3.89	S	0.64	FM	−0.84	216	−22.79	−1.53	4.81	−3.10
CrSCl	3.42	S	1.85	FM	0.05	252	−20.21	−0.93	0.96	0.07
CrBrS	3.50	S	1.47	FM	0.06	371	−22.00	−0.84	0.75	0.38
CrIS	3.65	S	0.67	FM	0.07	252	−20.02	−0.60	0.56	0.71
CrISe	3.75	S	1.00	FM	0.11	287	−23.59	−0.89	1.64	1.96
CrICl	3.92	HM	–	AFM	0.16	–	4.71	−1.21	0.16	0.39
CrIBr	3.91	HM	–	FM	0.78	90.3	−10.86	8.95	−6.31	−0.36

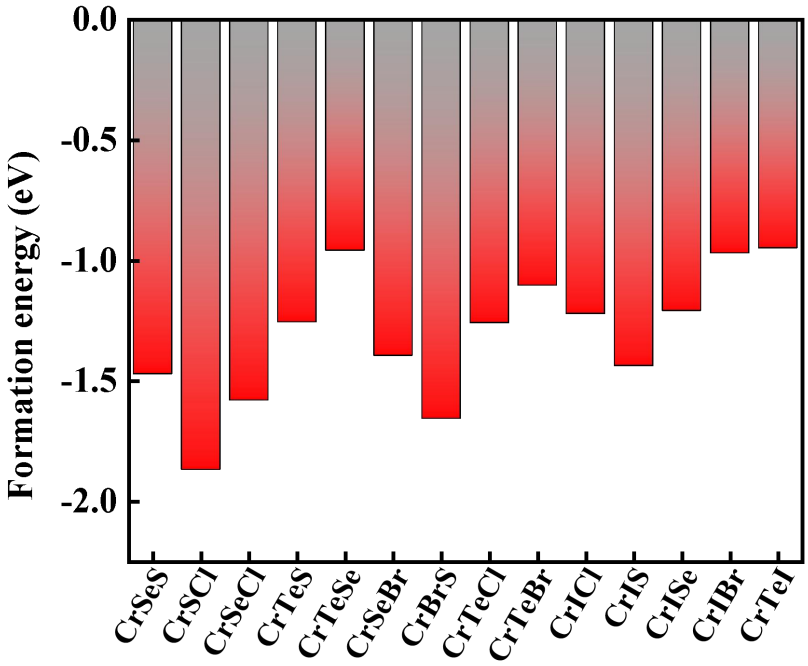

Fig. S5 Formation energies with respect to that of $CrTe_2$ monolayer as functions of various CrXY monolayers.

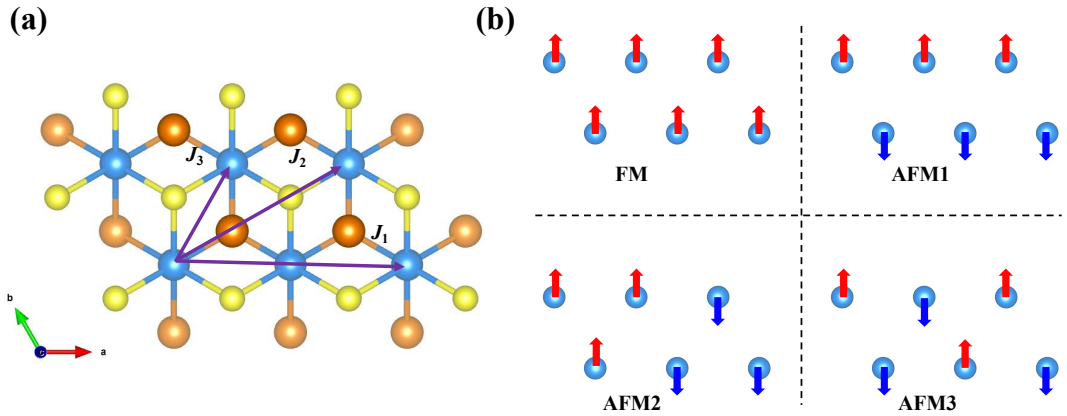

Fig. S6 (a) Heisenberg exchange coefficients J_1, J_2 and J_3; (b) the ferromagnetic and three antiferromagnetic configurations used to extract the Heisenberg coupling coefficients.

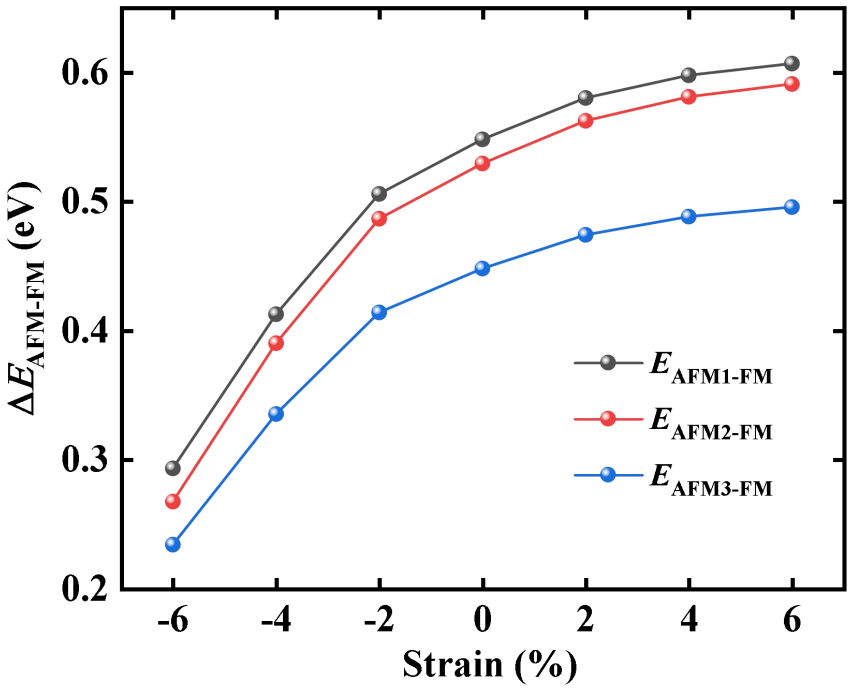

Fig. S7 Energy difference (ΔE) of AFM1, AFM2 and AFM3 spin configurations with respect to FM configuration as functions of strains in CrBrS.

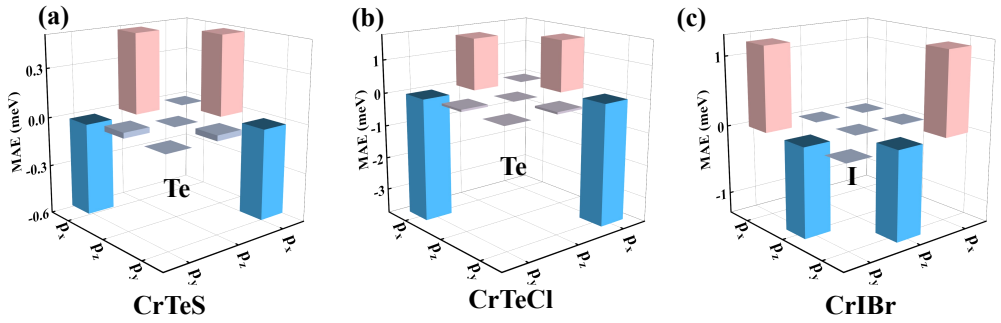

Fig. S8 Te atomic orbitals contribution to MAE in (a) CrTeS and (b) CrTeCl; (c) I atomic orbitals contribution to MAE in CrIBr.

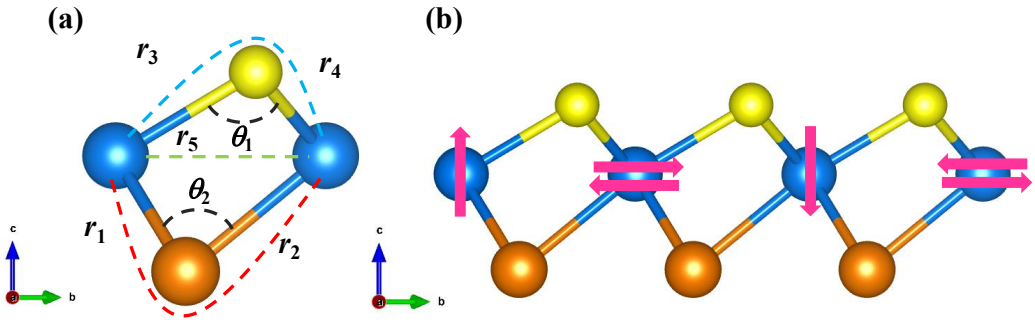

Fig. S9 (a) Schematic of θ_1, θ_2, paths of superexchange interactions r_1, r_2, r_3, r_4 and paths of direct interactions r_5; (b) clockwise and anticlockwise spin configurations to extract the DMI.

Table S6 Single-ion anisotropy coefficient (K_{single}), MAE constant (K_i) and effective anisotropy (K_{eff}) of CrXY.

CrXY	K_{single} (meV)	K_i (mJ/m^2)	$0.5\mu_0 M_s^2 t_{\text{eff}}$ (mJ/m^2)	K_{eff} (J/cm^3)
CrSeS	−1.26	−2.11	0.22	−15.62
CrSCl	0.05	0.08	0.31	−1.70
CrSeCl	−0.16	−0.24	0.27	−3.55
CrTeS	0.95	1.49	0.23	7.83
CrTeSe	0.75	1.13	0.20	5.58
CrSeBr	−0.08	−0.12	0.24	−2.40
CrBrS	0.06	0.09	0.27	−1.26
CrTeCl	−2.19	−3.00	0.23	−21.38
CrTeBr	−1.76	−2.31	0.20	−16.11
CrICl	0.16	0.19	0.25	−0.40
CrIS	0.07	0.10	0.22	−0.87
CrISe	0.11	0.15	0.20	−0.32
CrIBr	0.78	0.95	0.26	4.29
CrTeI	−0.84	−1.023	0.17	−7.40

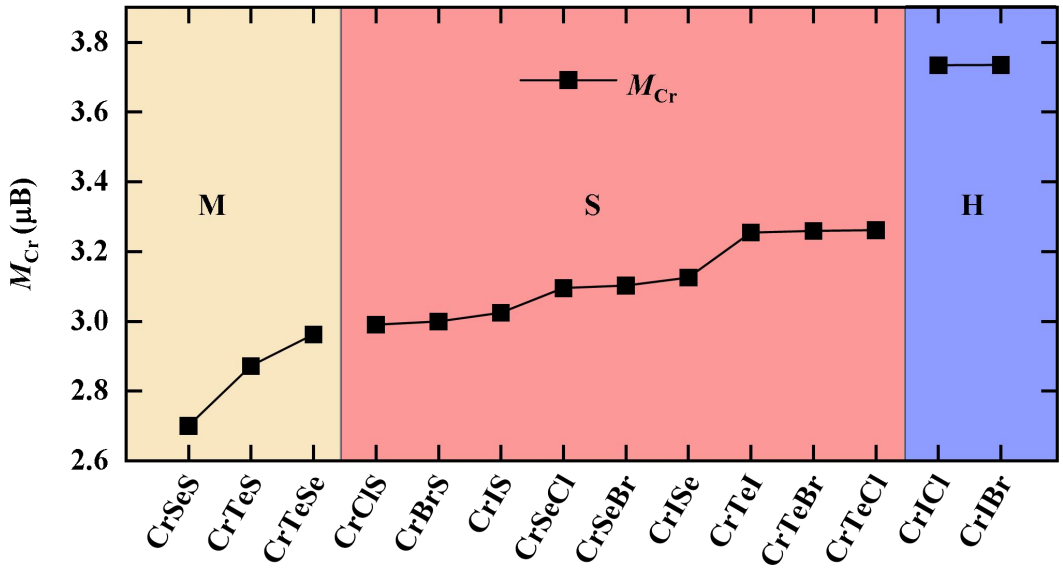

Fig. S10 Magnetic moments of Cr atoms in CrXY monolayers, where M, S, and H represent metal, semiconductor and half-metal, respectively. We note a significant correlation between the magnetic moment of Cr atoms and the electronegativity of the nonmagnetic atoms

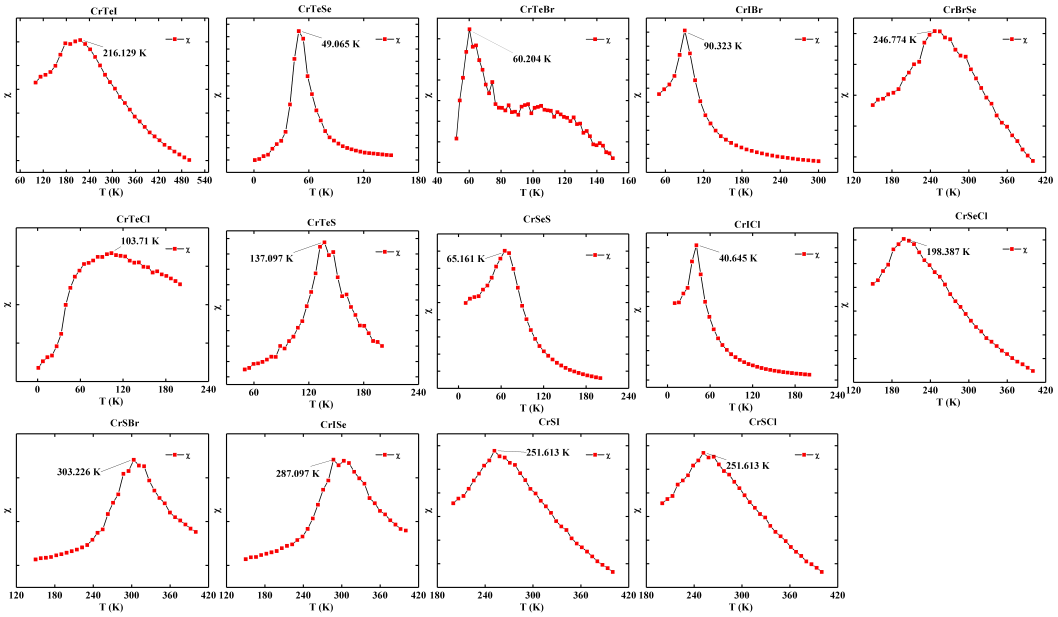

Fig. S11 Magnetic susceptibility as a function of temperature and the curve exhibits a peak at T_C, which corresponds to the critical temperature.

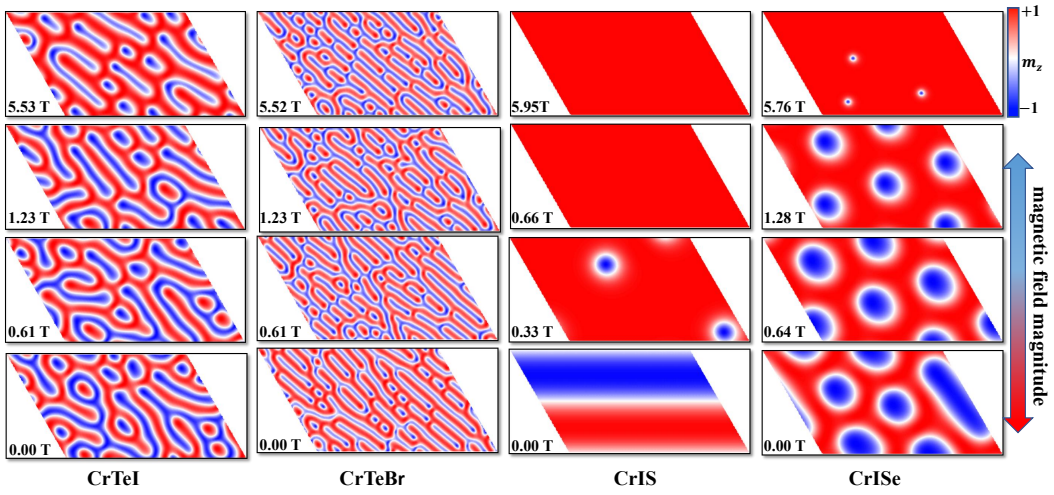

Fig. S12 Evolutions of spin textures in CrTeI, CrTeBr, CrIS and CrISe with changing magnetic fields at 0 K. In the simulations, a 60×60×1 supercell is adopted.

Notes and references

1 S. Jiang, S. Nazir and K. Yang, *Phys. Rev. B*, 2020, **101**, 134405.

2 S. Kirklin, J. E. Saal, B. Meredig, A. Thompson, J. W. Doak, M. Aykol, S. Rühl and C. Wolverton, *NPJ Comput. Mater.*, 2015, **1**, 1–15.