Supplementary Information

Tunable Magnetic Anisotropy and Phase Transitions in 2D Janus Transition-Metal Chalcogenide Halides

Siyu Han,[a] Jiaqi Wang,[a] Tong Zhao,[a] Jian Zhou,[a] Naihua Miao*[a] and Zhimei Sun*[a]

1.School of Materials Science and Engineering, Beihang University, Beijing 100191,P. R. China.

Corresponding authors:

E-mail: zmsun@buaa.edu.cn. (Z. Sun)

E-mail: nhmiao@buaa.edu.cn. (N. Miao)

TABLE S1. Energy difference between FM and AFM states, $\Delta E = E_{FM} - E_{AFM}$ (meV/TM) for MSBr and Janus M_2S_2BrX (M = Cr/V, X = F, Cl, I) monolayers with/without SOC.

	With SSC	W/o SSC
CrSBr	-40.85	-42.82
Cr_2S_2BrF	-30.67	-32.88
Cr_2S_2BrCl	-39.56	-41.74
Cr_2S_2BrI	-41.59	-44.20
VSBr	-26.81	-31.45
V_2S_2BrF	-26.12	-33.00
V_2S_2BrCl	-27.68	-32.86
V_2S_2BrI	-26.08	-30.72

TABLE S2. Hubbard U parameter test for VSBr: Magnetic state and lattice constant evolution U and J_H in eV.

U (eV)	J_H (eV)	E_{FM}-E_{AFM} (meV/TM)	a (Å)	b (Å)
1.36	0.39	-23.84	3.49	4.91
2.36	0.39	-24.95	3.53	4.99
3.36	0.39	-26.81	3.55	5.02
4.36	0.39	-27.55	3.57	5.05
5.36	0.39	-27.47	3.59	5.09

TABLE S3. Hubbard *U* parameter test for CrSBr: Magnetic state and lattice constant evolution *U* and J_H in eV.

U (eV)	J_H (eV)	E_{FM}-E_{AFM} (meV/TM)	*a* (Å)	*b* (Å)
2.03	0.96	-42.26	3.55	4.78
3.03	0.96	-46.55	3.56	4.80
4.03	0.96	-42.82	3.57	4.83
5.03	0.96	-50.58	3.59	4.85
6.03	0.96	-40.48	3.60	4.88

TABLE S4. Exchange parameters J for 2D CrSBr in different references.

	J_1	J_2	J_3	U (eV)	J (eV)
this paper	2.25	3.15	1.61	4.03	0.96
$calc_1$[1]	1.66	3.09	1.52	4.03	0.96
$calc_2$[2]	4.29	3.23	3.04	4.03	0.96
$calc_3$[3]	3.22	4.11	2.78	4.00	1.00
$calc_4$[4]	5.05	5.54	1.87	3.00	/
expt[5]	1.90	3.38	1.67		

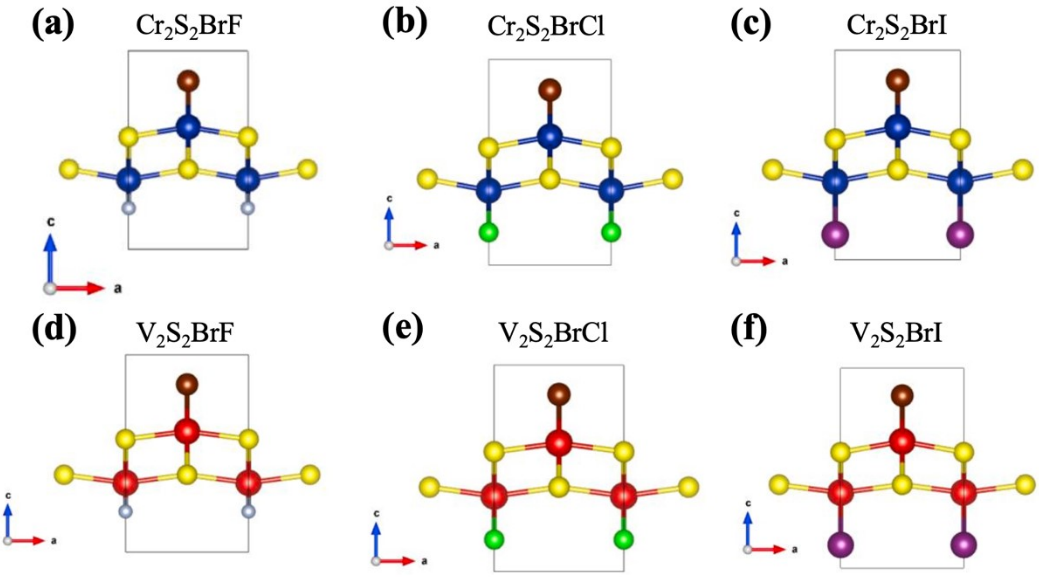

FIG. S1. Atomic structures of Janus M_2S_2BrX (M = Cr/V, X = F, Cl, I) monolayers.

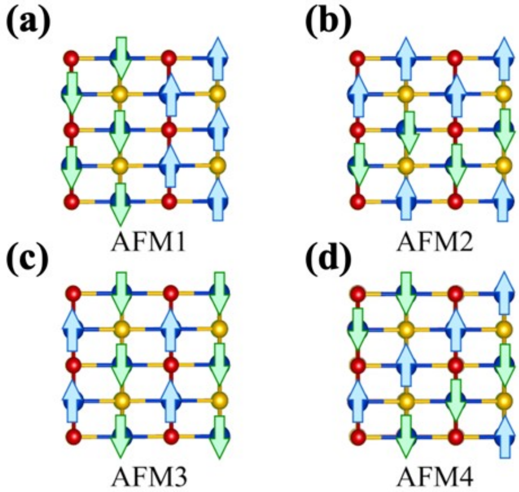

FIG. S2. Top views of AFM states of MSBr and M_2S_2BrX (M = Cr/V, X = F, Cl, I). Blue, yellow and red spheres denote M, S, and X atoms, respectively. Blue and green arrows represent the spin-up and spin-down moments, respectively.

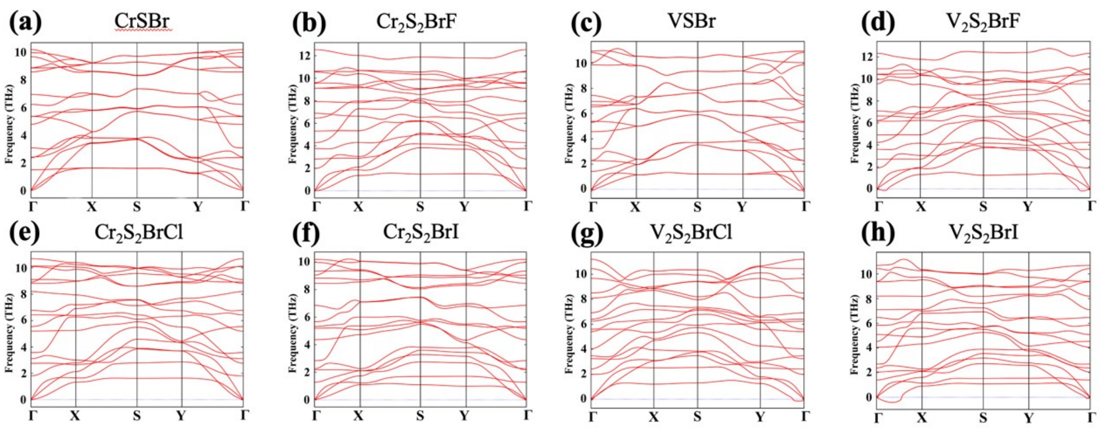

FIG. S3. Phonon dispersions for MSBr and M_2S_2BrX ((M = Cr/V, X = F, Cl, I)).

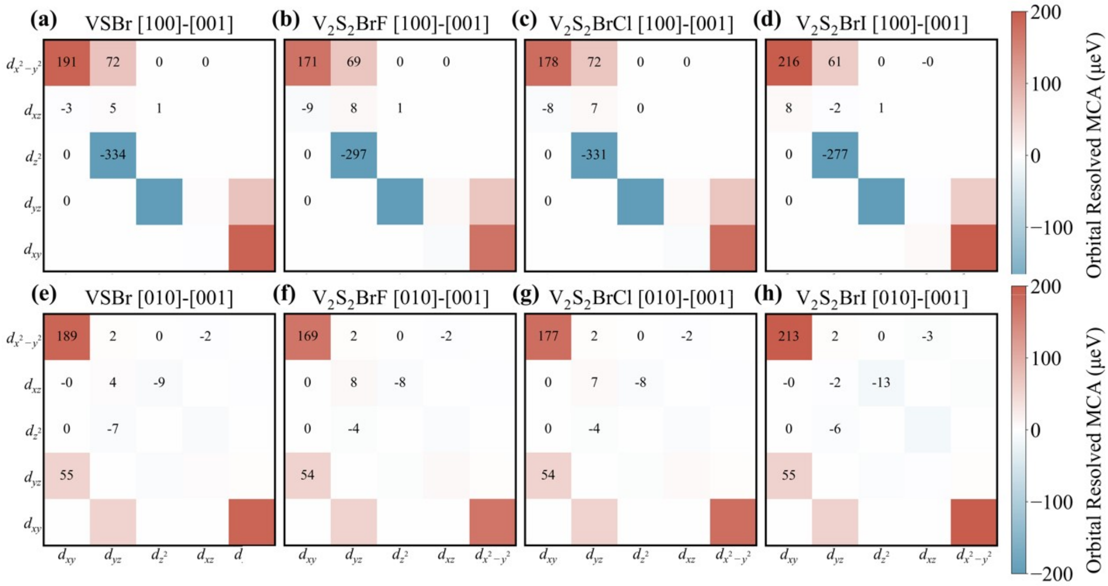

FIG. S4. Orbital-resolved SOC-MA contributions between V 3*d* orbitals in VSBr and Janus V_2S_2BrX (X = F, Cl, I) monolayers. Panels (a–d) show the SOC-MA contributions along the [100] directions, while panels (e–h) correspond to the [010] direction, with [001] taken as the reference axis. Positive (negative) values indicate orbital pairs favoring the out-of-plane (in-plane) magnetization direction.

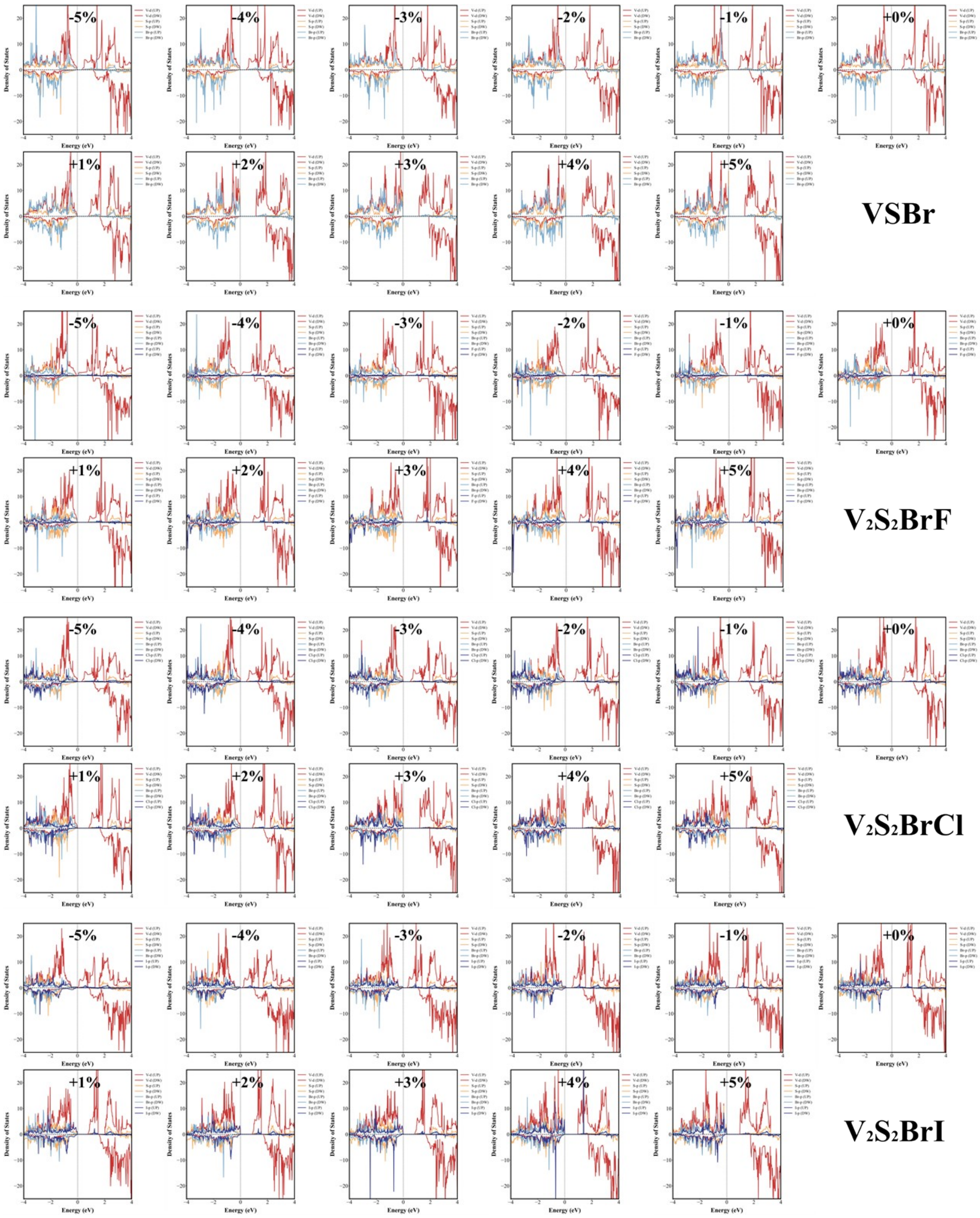

FIG. S5 Spin-polarized PDOS for strained VSBr and Janus V_2S_2BrX (X = F, Cl, I) monolayers under biaxial strains ranging from –5% to +5%. Positive and negative values correspond to spin-up and spin-down channels, respectively. The PDOS includes contributions from V-*d* (red), S-*p* (orange), Br-*p* (light blue), and X-*p* (dark blue) orbitals.

References

[1] C. Wang, X. Zhou, L. Zhou, N.-H. Tong, Z.-Y. Lu and W. Ji, Sci. Bull. **64**, 293 (2019).

[2] B. Wang, Y. Wu, Y. Bai, P. Shi, G. Zhang, Y. Zhang and C. Liu, Nanoscale **15**, 13402 (2023).

[3] K. Yang, G. Wang, L. Liu, D. Lu and H. Wu, Phys. Rev. B **104**, 144416 (2021).

[4] H. Wang, J. Qi and X. Qian, Appl. Phys. Lett. **117**, 083102 (2020).

[5] A. Scheie, M. Ziebel, D. G. Chica, Y. J. Bae, X. Wang, A. I. Kolesnikov, X. Zhu and X. Roy, Adv. Sci. **9**, 2202467 (2022).