Supplementary Material for

Strain-Driven Skyrmion-Bimeron Switching in Topological Magnetic Monolayer CrSeBr

Junhuang Yang‖, Kaiying Dou‖, Xinru Li*, Ying Dai*, Baibiao Huang, and Yandong Ma*
School of Physics, State Key Laboratory of Crystal Materials, Shandong University, Shandanan Str. 27, Jinan 250100, China

E-mail: daiy60@sdu.edu.cn (Y.D.); lixr@sdu.edu.cn (X.L.); yandong.ma@sdu.edu.cn (Y.M.)

Note 1: Phonon spectra and AIMD simulations.

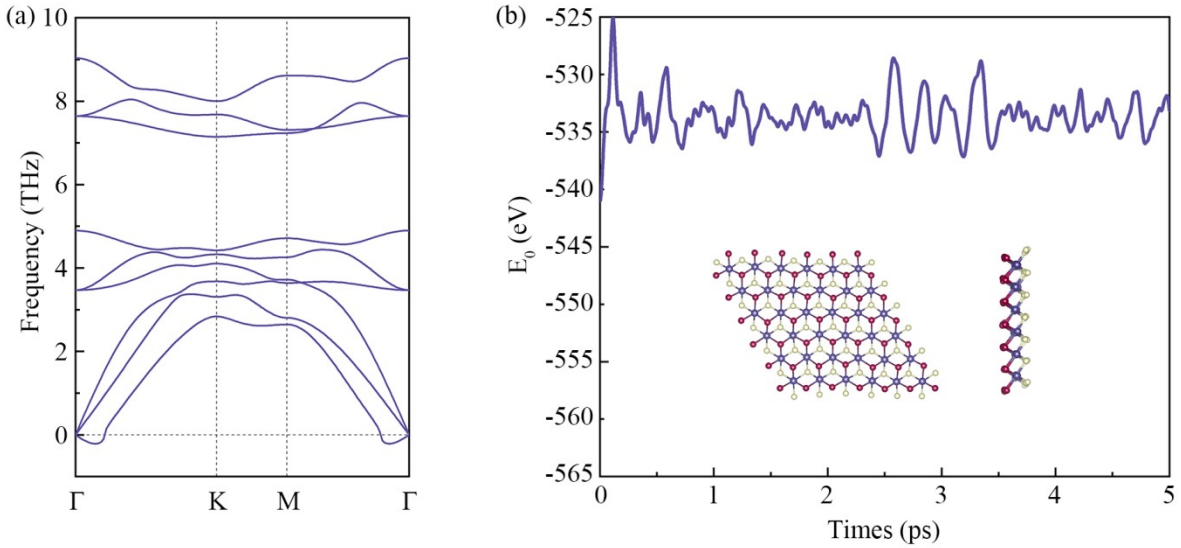

Figure S1 (a) Phonon spectra and (b) variation of total energy with time during the AIMD simulations for monolayer CrSeBr. Insert in (b) is snapshot of the structure for monolayer CrSeBr taken from the end of AIMD simulations (300 K and 5 ps).

Note 2: Calculation details of magnetic parameters in spin Hamiltonian.

To calculate the magnetic parameters J and K we consider the following four spin configurations as shown in **Fig S2(a)**. The energies of these different spin configurations can be written as follows:

$$E_1 = 2E_0 - 6J|S|^2$$

$$E_2 = 2E_0 + 6J|S|^2$$

$$E_3 = E_0 - 3J|S|^2$$

$$E_4 = E_0 - 3J|S|^2 - K|S|^2$$

According to Moriya's rule, DMI between the nearest Cr atoms can be written as $D_{ij} = d_{||}(u_{ij} \times z) + d_{\perp} z$, here $d_{\perp}$ can be ignored. To obtain the in-plane component $d_{||}$, we consider two configurations [clockwise (CW) and anticlockwise (ACW)], as shown in **Fig S2(b)**. The total energies can be written as follows:

$$E_{CW} = E_0 - \frac{3}{2} d_{||} |S|^2 \times 4$$

$$E_{ACW} = E_0 + \frac{3}{2} d_{||} |S|^2 \times 4$$

Therefore, the in-plane component $d_{||}$ can be obtained by $d_{||} = (E_{ACW} - E_{CW})/12$.

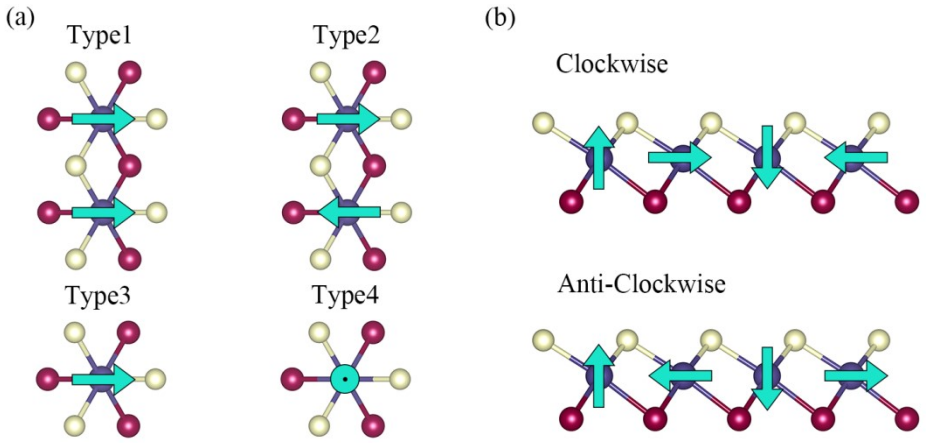

Figure S2 (a) Four spin configurations employed for calculating Heisenberg isotropic exchange and SIA parameters. (b) Two spin configurations used for calculating DMI parameters.

Note 3: Detailed spin patterns of monolayer CrSeBr.

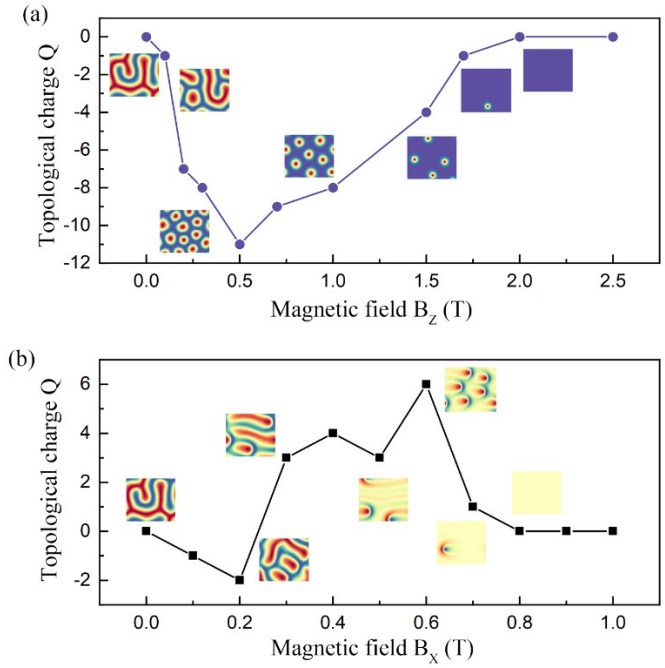

Figure S3 Evolutions of topological charge Q and spin patterns of monolayer CrSeBr under different external magnetic fields.

Table S1 Magnetic parameters of monolayer CrSeBr

Strain (%)	J(meV)	K (meV)	$d_{\|\|}$(meV)	m (μ_B)	$\|\kappa\|$
-5	21.435	-0.282	1.369	3.33	3.492
-4	23.645	-0.201	1.456	3.34	2.469
-3	25.563	-0.173	1.555	3.36	1.982
-2	27.224	-0.144	1.653	3.37	1.554
-1	28.619	-0.122	1.752	3.39	1.229
0	29.940	-0.084	1.859	3.40	0.789
1	31.002	-0.071	1.961	3.42	0.616

Note 4: Atomic-resolved localization of DMI associated SOC energy and the orbital-resolved MAE under different strains.

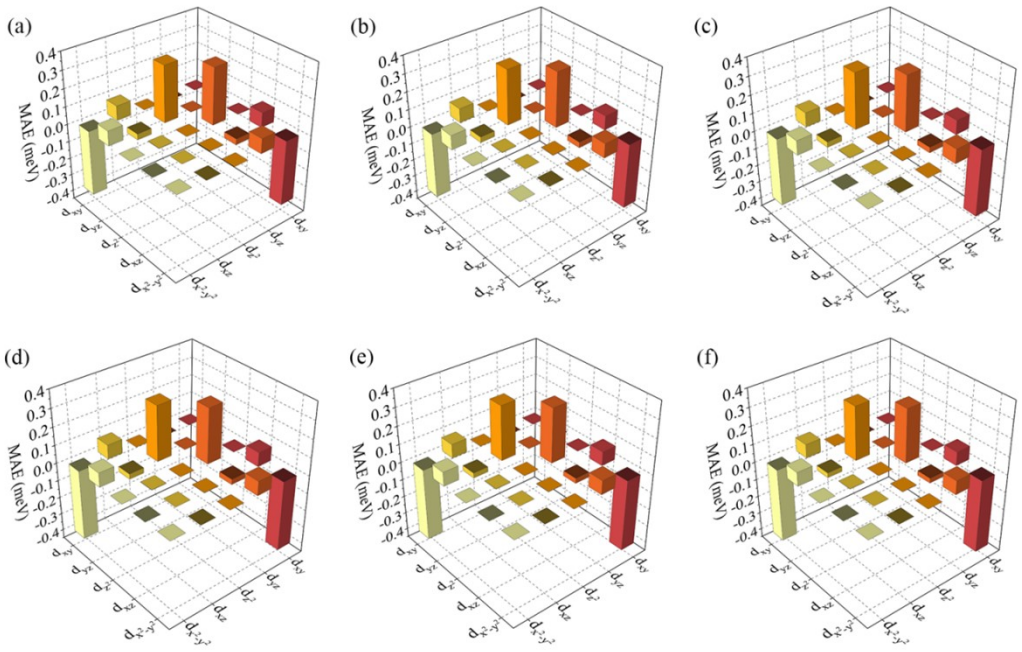

Figure S4 MAE contributions from the 3*d* orbitals of Cr atoms hybridization under (a)0%, (b)-1%, (c)-2%, (d)-3%, (e)-4%, (f)-5% strain.

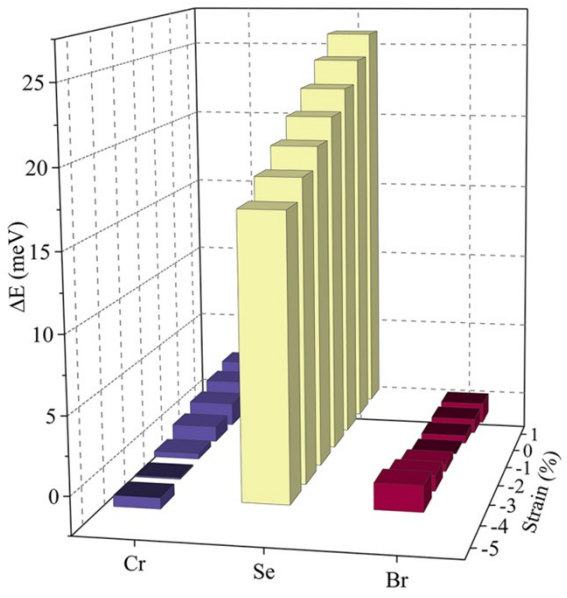

Figure S5 Atomic-resolved SOC energy for monolayer CrSeBr as a function of the strain.