

Supplementary information:

Strain-selected magnetic ordering in 1T' α -CrXY (X, Y = S, Se, Te) monolayers

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Table. S1 The calculated lattice parameters (a and b), nearest Cr-Cr distances (d_1 , d_2 and d_3), and Wyckoff sites of 1T' α -CrTeSe monolayer.

Lattice parameters (Å)	Nearest distances (Å)	Wyckoff sites
$a = 6.642$	$d_1 = 3.979$	Cr $2e(0.728, 1/4, 0.498)$
$b = 3.352$	$d_2 = 3.352$	Te $2e(0.064, 1/4, 0.440)$
	$d_3 = 3.470$	Se $2e(0.430, 1/4, 0.552)$

Table. S2 The calculated energies of four magnetic configurations (FM, sAFM, dAFM, zAFM) of 1T' α -CrTeSe monolayer, in which all values are referenced to the FM configuration.

sAFM (meV)	dAFM (meV)	zAFM (meV)
515.1	188.6	65.6

Table. S3 The calculated MAEs of the strain-free 1T' α -CrTeSe monolayer along the x (x'), y (y'), and z (z') axes, in which all values are referenced to x (x') axis.

E_y (μ eV)	E_z (μ eV)	$E_{y'}$ (μ eV)	$E_{z'}$ (μ eV)
1348	161	1708	504

Table. S4 The calculated MAEs of the 1T' α -CrTeSe monolayer under strains along the x , y , and z axes, in which all values are referenced to x axis.

ε (%)	E_y (μ eV)	E_z (μ eV)
-6	-735	-1008
-2	585	-159
-1	1352	-433
1	1285	77
0.2	1213	-188
0.4	1327	-57
0.6	1167	-212
0.8	1202	32
2	979	-120
6	1029	917

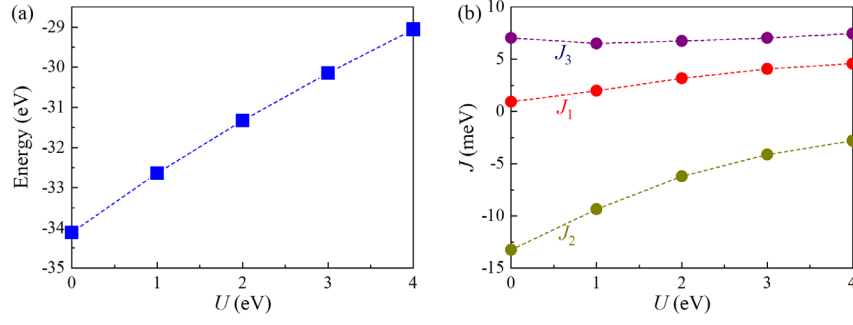


Fig. S1 The calculated (a) total energy and (b) exchange parameters (J) of 1T' α -CrTeSe monolayer as functions of U values.

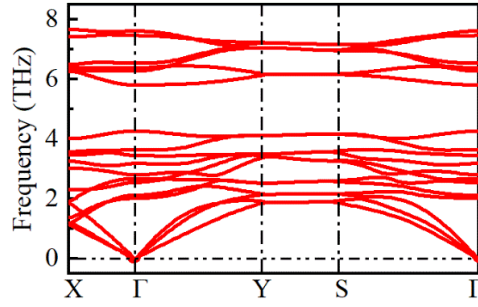


FIG. S2 Phonon spectrum of 1T' α -CrTeSe monolayer along the high-symmetric k points in Brillouin zone.

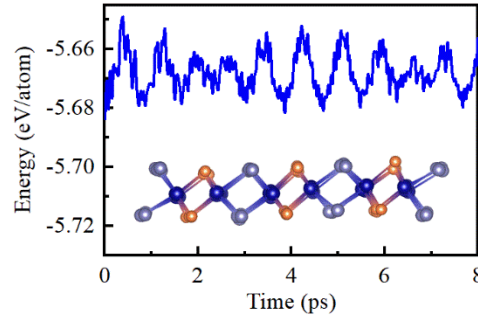


FIG. S3 AIMD simulation of 1T' α -CrTeSe monolayer at 300 K up to 8 ps. The final snapshots of these monolayers are inset.

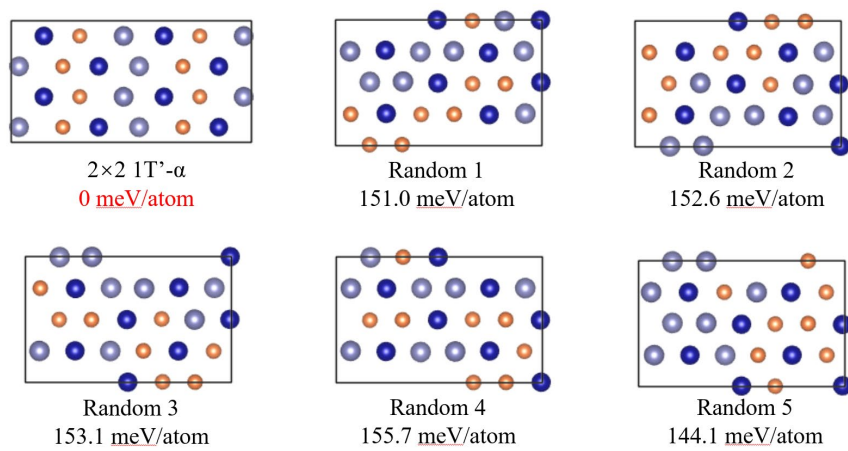


FIG. S4 The optimized structures of 1T' α -CrTeSe monolayer and several random patterns. The energies relative to 1T' α phase are inset.

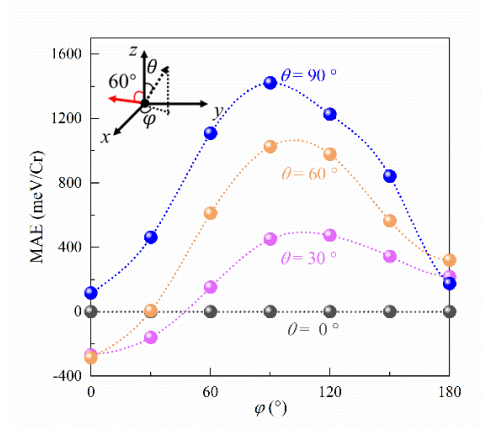


FIG. S5 MAEs of 1T' α -CrTeSe monolayer with the spin moments parallelly oriented along different directions. The spin rotation angle (θ/φ) of 0 is along the z/x axis.

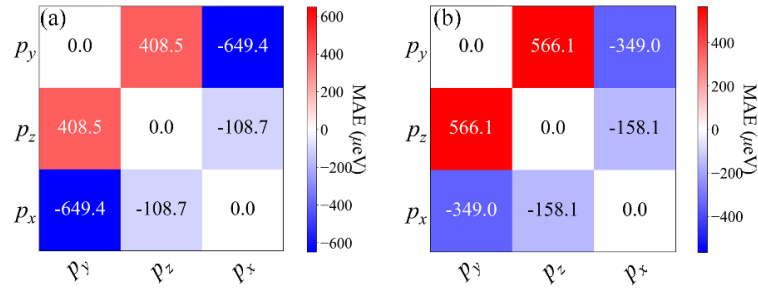


FIG. S6 Orbital-resolved MAEs of 1T' α -CrTeSe monolayer.

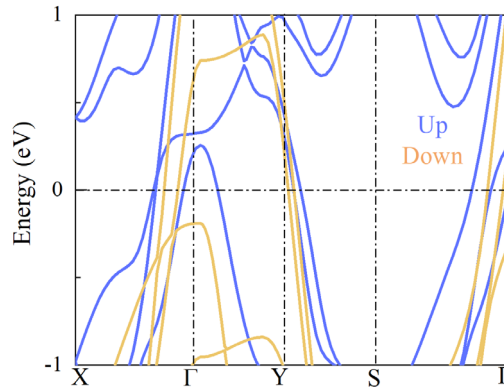


Fig. S7 Spin-polarization band structures of 1T' α -CrTeSe monolayer, in which the blue and yellow lines represent the spin-up and spin-down states, respectively.

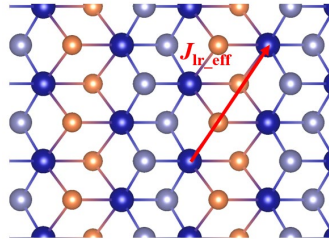


Fig. S8 Sketch of the effective long-range exchange parameter (J_{lr_eff}) in 1T' α -CrTeSe monolayer.

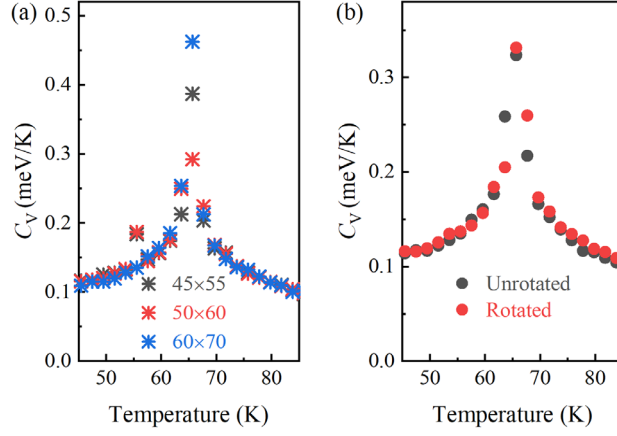


FIG. S9 MC simulations of the specific heats of 1T'α-CrTeSe monolayer by using (a) different supercells and (b) MAEs as functions of temperatures.

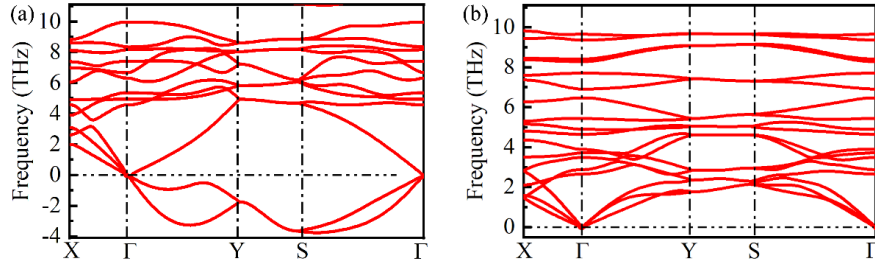


FIG. S10 Phonon spectra of (a) 1T'α-CrSO and (b) 1T'α-CrSeS monolayers along the high-symmetric k points in Brillouin zone.

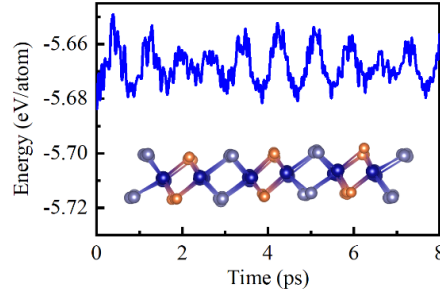


FIG. S11 AIMD simulation of 1T'α-CrSeS monolayer at 300 K up to 8 ps. The final snapshots of these monolayers are inset.

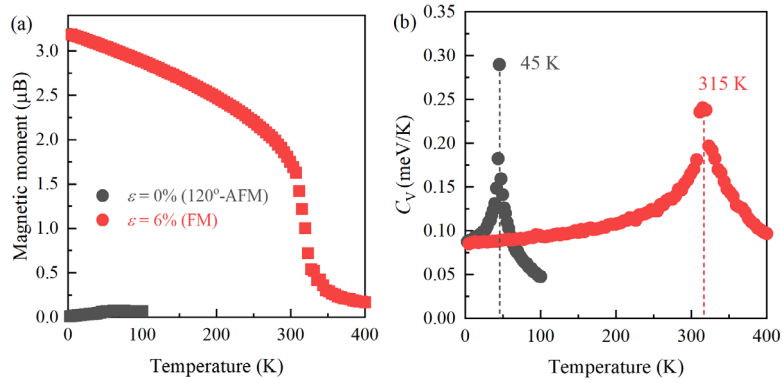


FIG. S12 MC simulations of the (a) magnetic moments and (b) specific heats of 1T'α-CrSeS monolayer with and without in-plane tensile biaxial strain. The lines and symbols with different colors represent different strain environments.