Supplementary Information (SI) for Physical Chemistry Chemical Physics.

Supplementary Materials for Tunable valley splitting and magnetic anisotropy in two-dimensional buckled honeycomb lattice Mn_2X_2 (X = F, Cl, Br)

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TABLE S1: Calculated optimized lattice constants (a), nearest Mn–Mn distances (d), Mn–X–Mn angles (α), cohesive energy (ΔE_{coh}), and the energy differences of the FM and AFM2 states with respect to the AFM1 state (meV/unit cell).

Material	a (Å)	d (Å)	α (°)	ΔE_{coh} (eV/atom)	$\Delta E_{\mathrm{FM-AFM1}}$	$\Delta E_{\mathrm{AFM2-AFM1}}$
Mn_2F_2	3.07	2.64	91.7	-4.180	374.972	281.069
Mn_2Cl_2	3.37	2.66	84.9	-3.297	583.703	480.695
Mn_2Br_2	3.54	2.69	84.0	-2.957	621.503	536.694

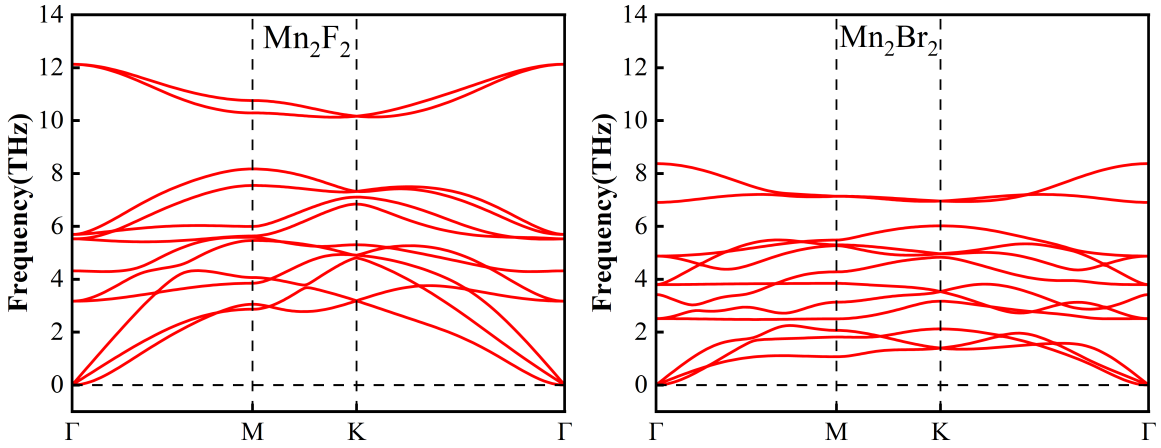

FIG. S1: Phonon spectra of Mn_2Br_2 and Mn_2F_2.

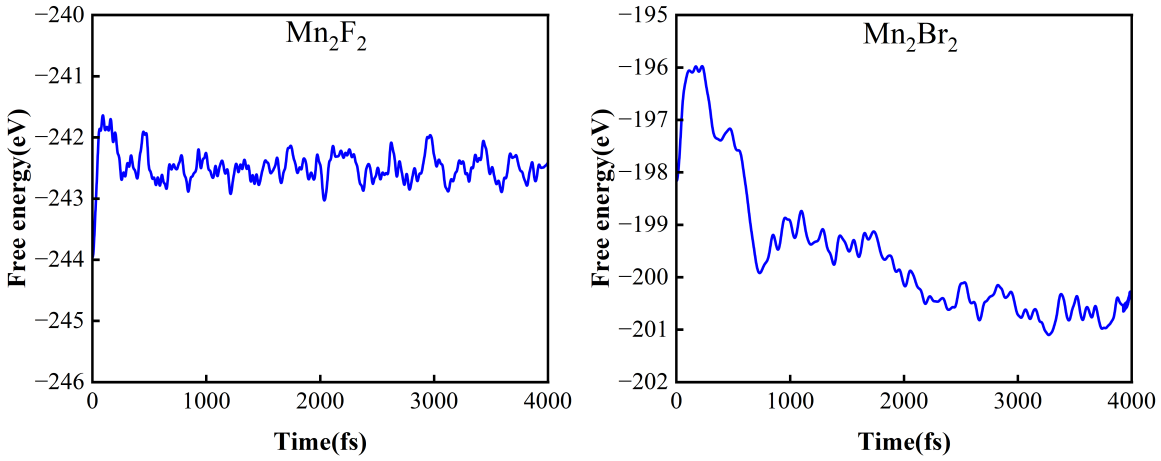

FIG. S2: AIMD simulations of Mn_2Br_2 and Mn_2F_2.

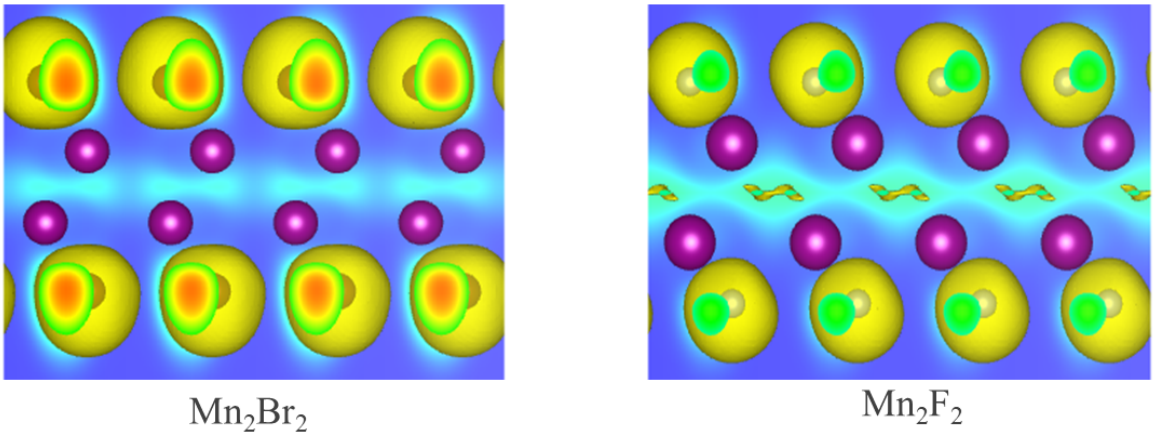

FIG. S3: ELF of Mn_2Br_2 and Mn_2F_2.

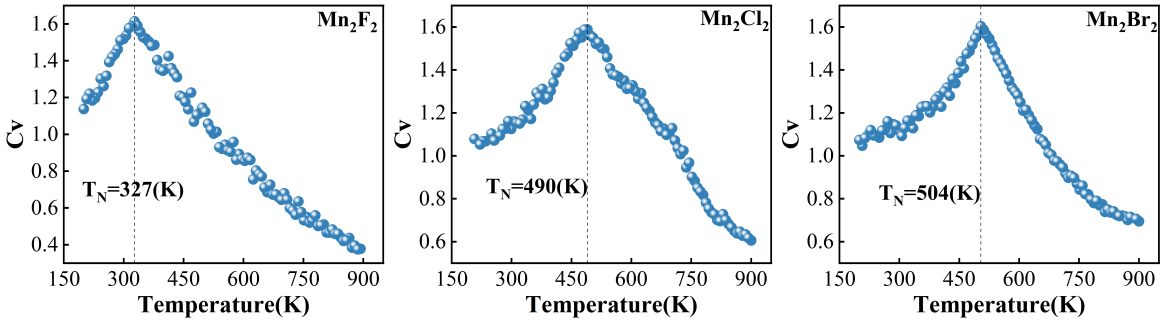

FIG. S4: T_N of monolayer Mn_2X_2 .

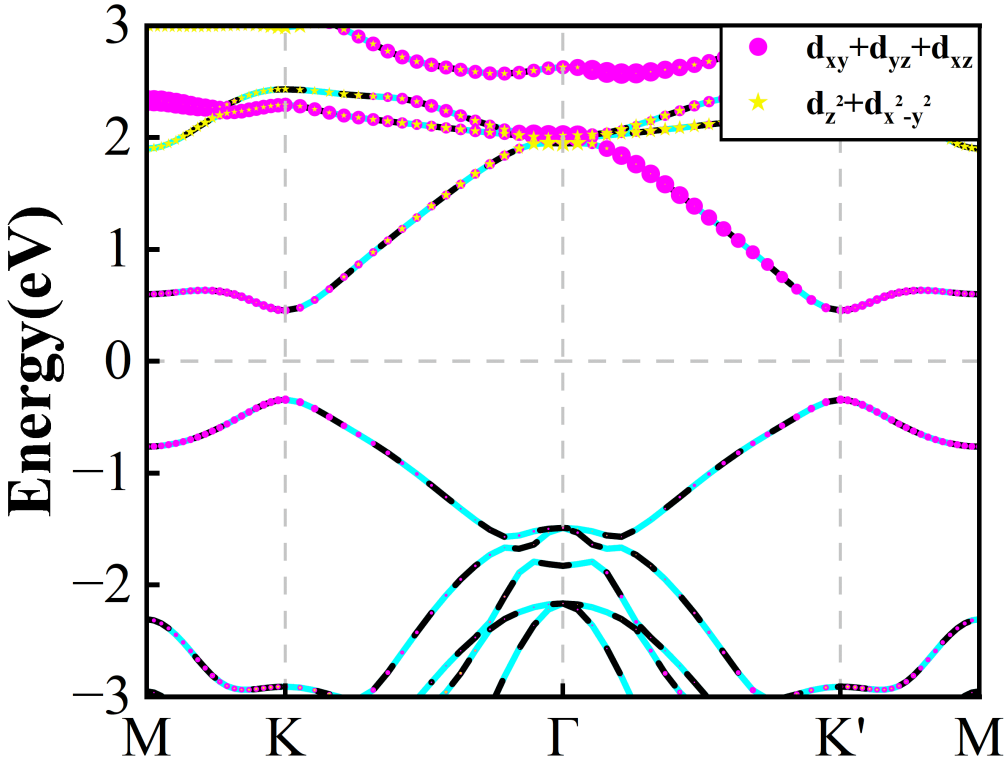

FIG. S5: PBAND of monolayer Mn_2Br_2 without SOC.

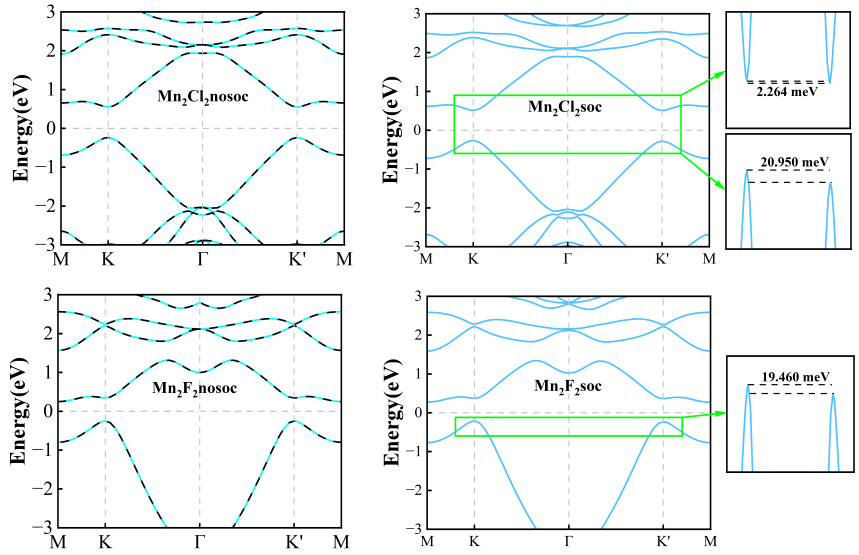

FIG. S6: Band structures of Mn_2Cl_2 and Mn_2F_2 with and without SOC.

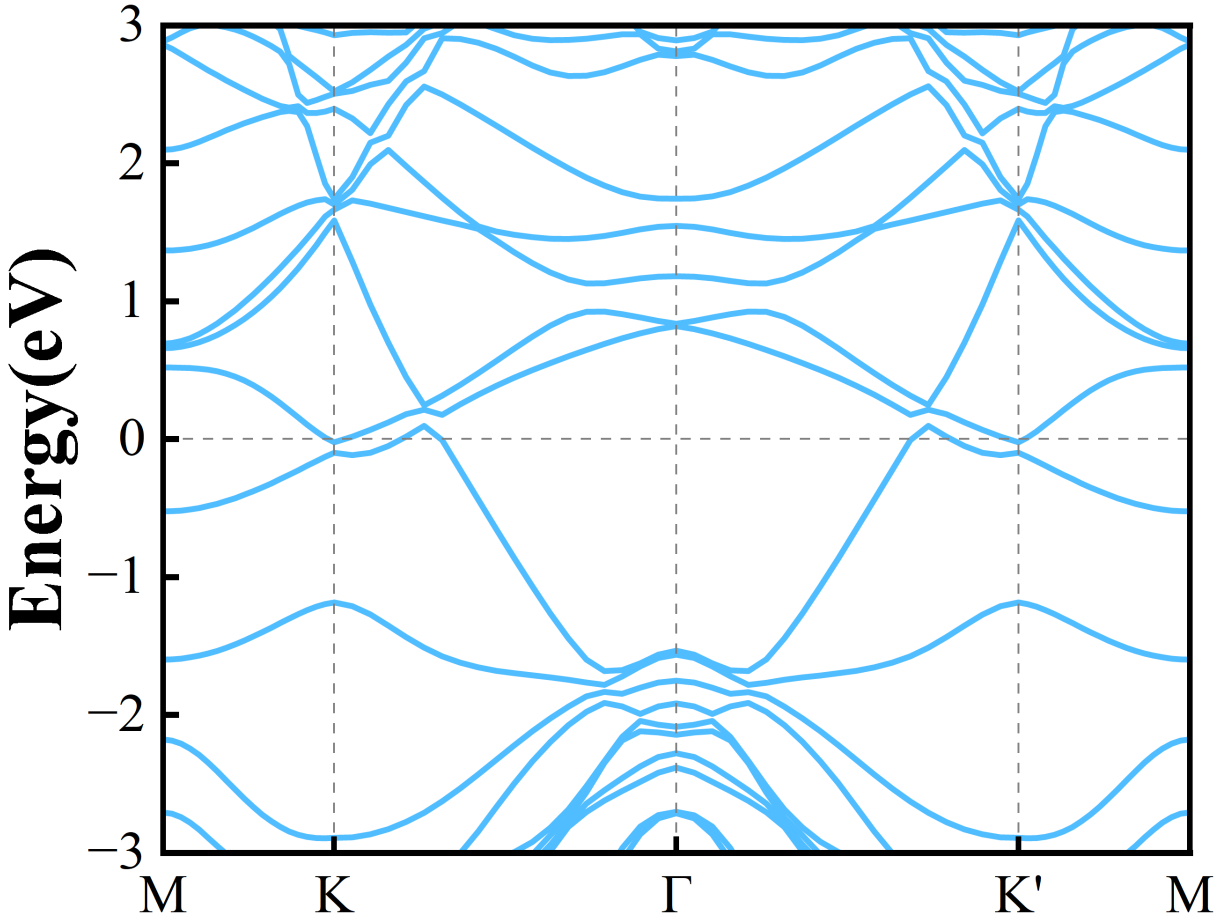

FIG. S7: Band structure of Mn_2Br_2 in the FM state with SOC.

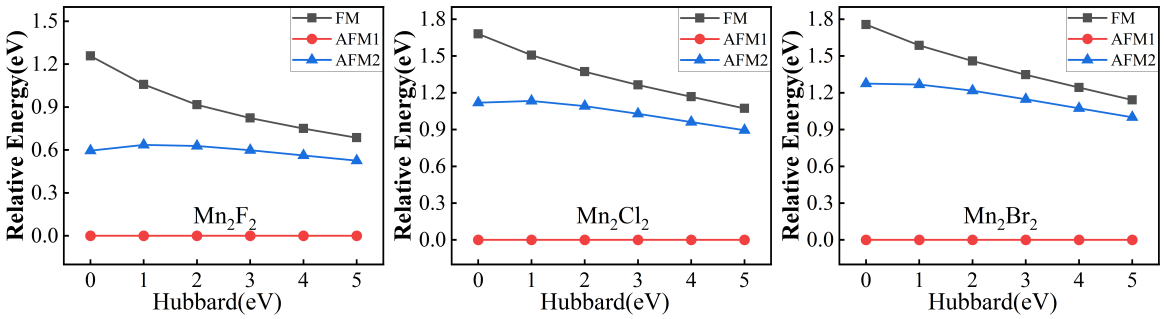

FIG. S8: Total energies of monolayer Mn_2X_2 for different magnetic configurations as a function of the Hubbard U, with the energies measured relative to the AFM1 state.

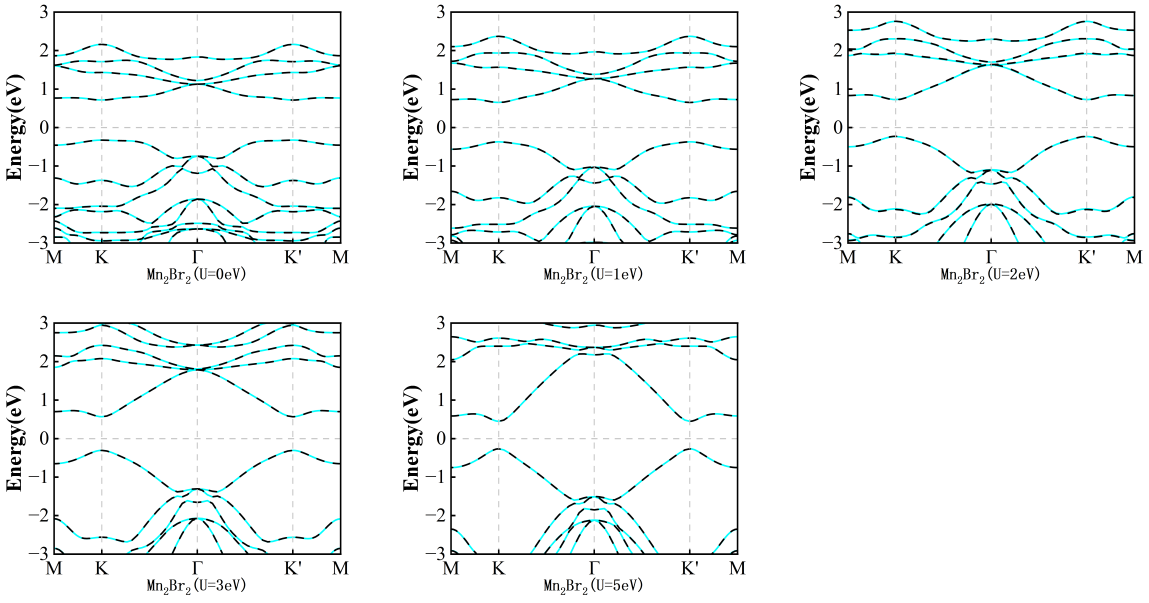

FIG. S9: Band structures of Mn_2Br_2 without SOC under different Hubbard U values.

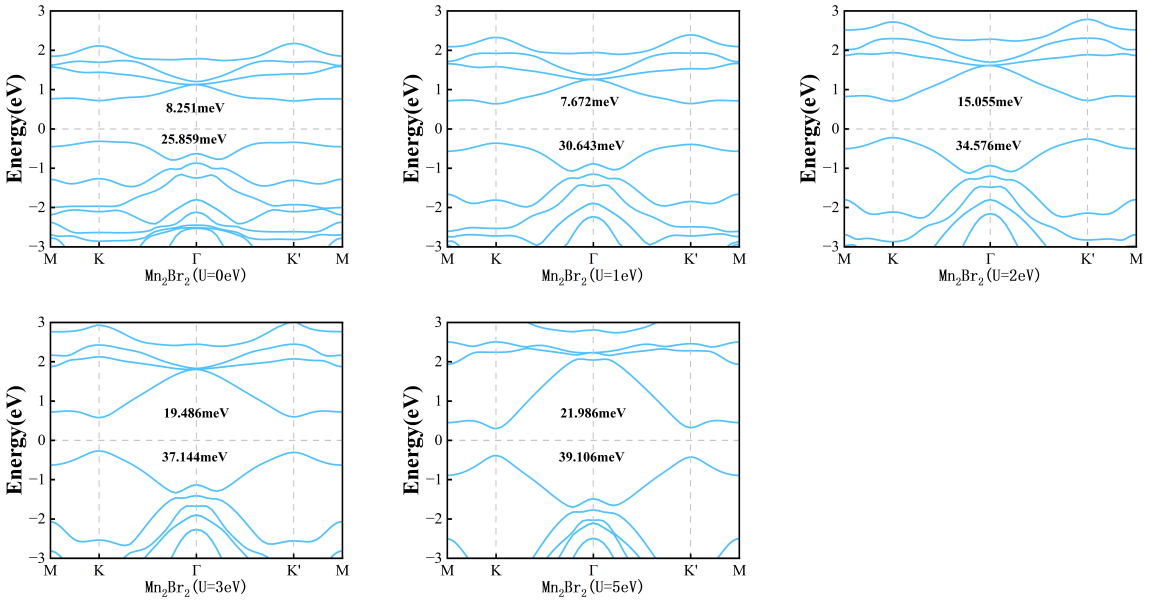

FIG. S10: Band structures of Mn_2Br_2 with SOC under different Hubbard U values.

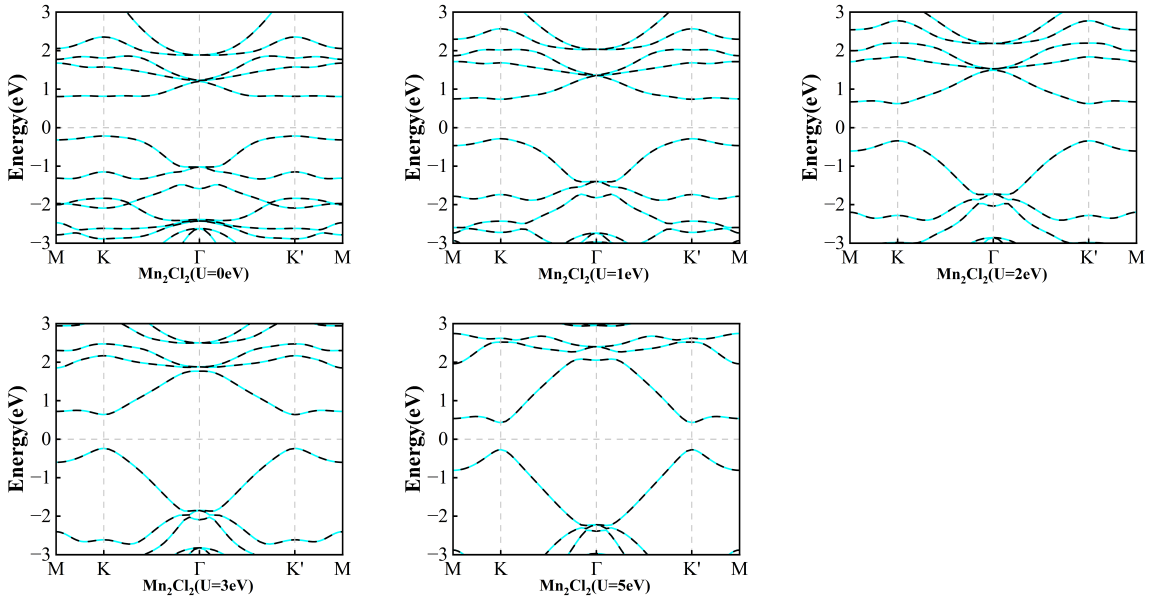

FIG. S11: Band structures of Mn_2Cl_2 without SOC under different Hubbard U values.

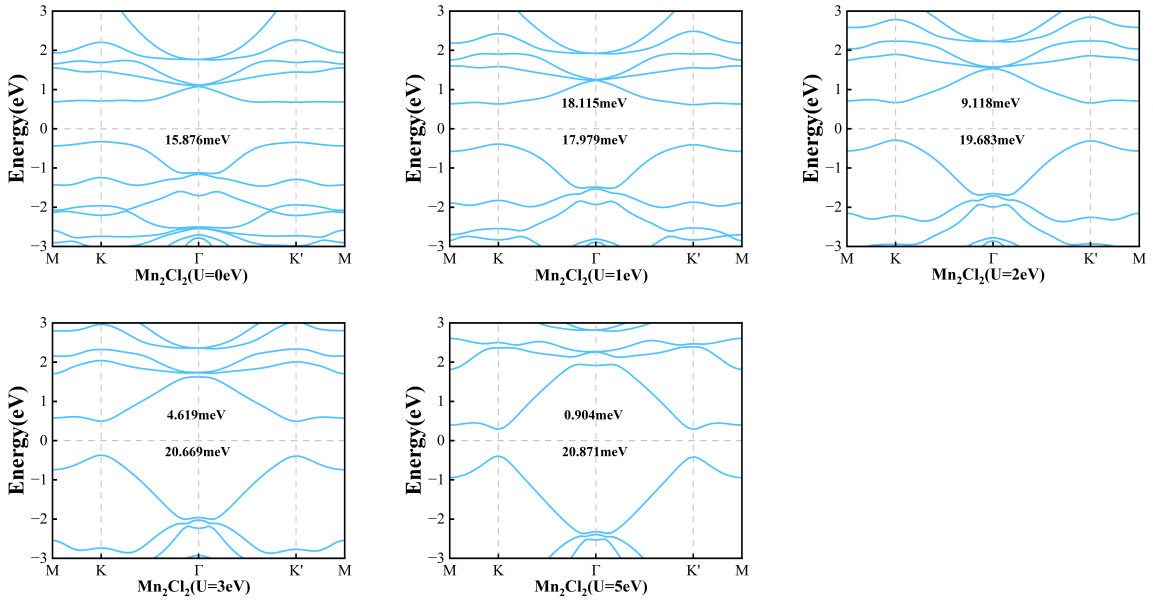

FIG. S12: Band structures of Mn_2Cl_2 with SOC under different Hubbard U values.

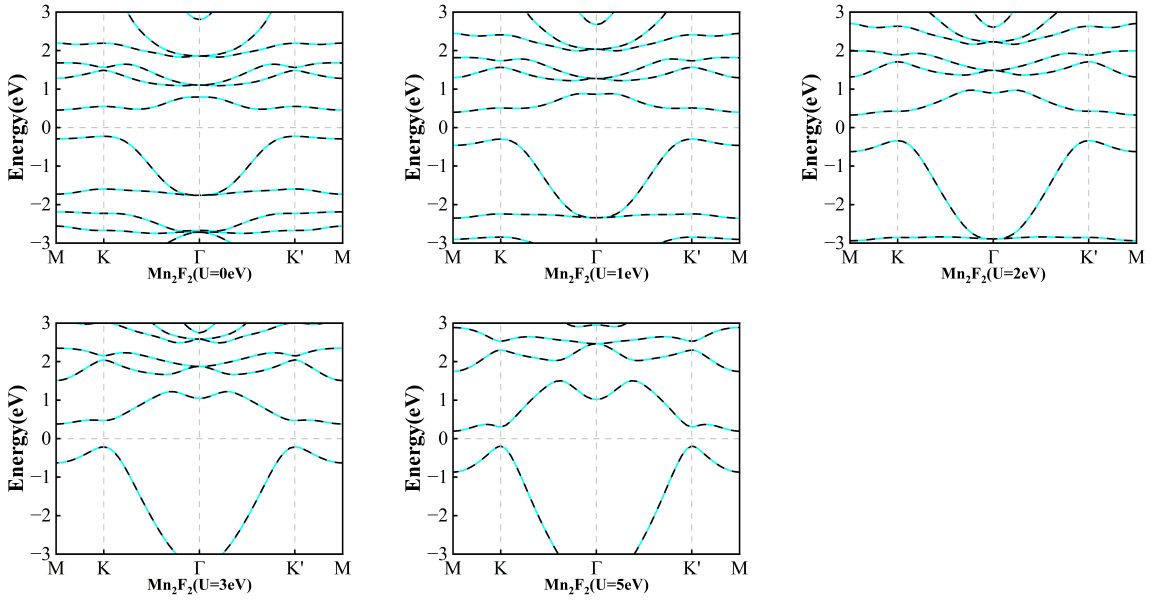

FIG. S13: Band structures of Mn_2F_2 without SOC under different Hubbard U values.

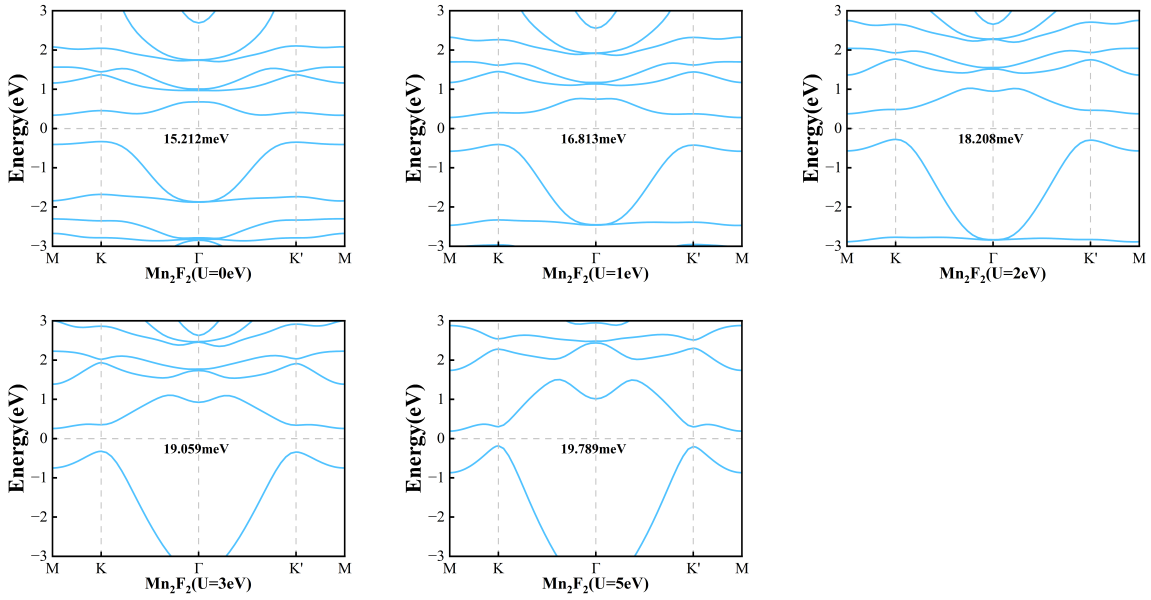

FIG. S14: Band structures of Mn_2F_2 with SOC under different Hubbard U values.

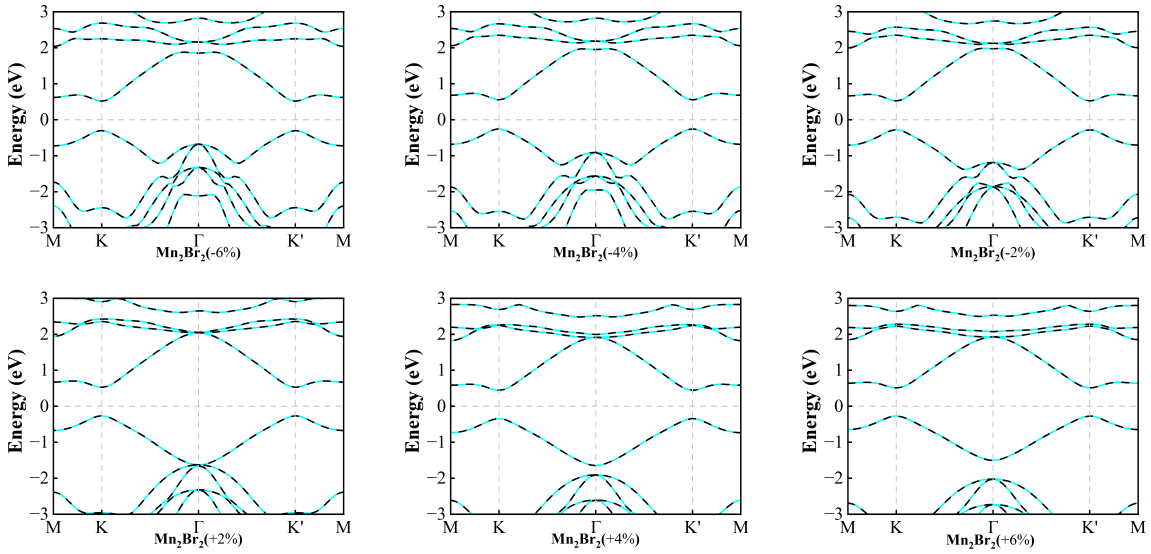

FIG. S15: Band structures of Mn_2Br_2 without SOC under different strain values.

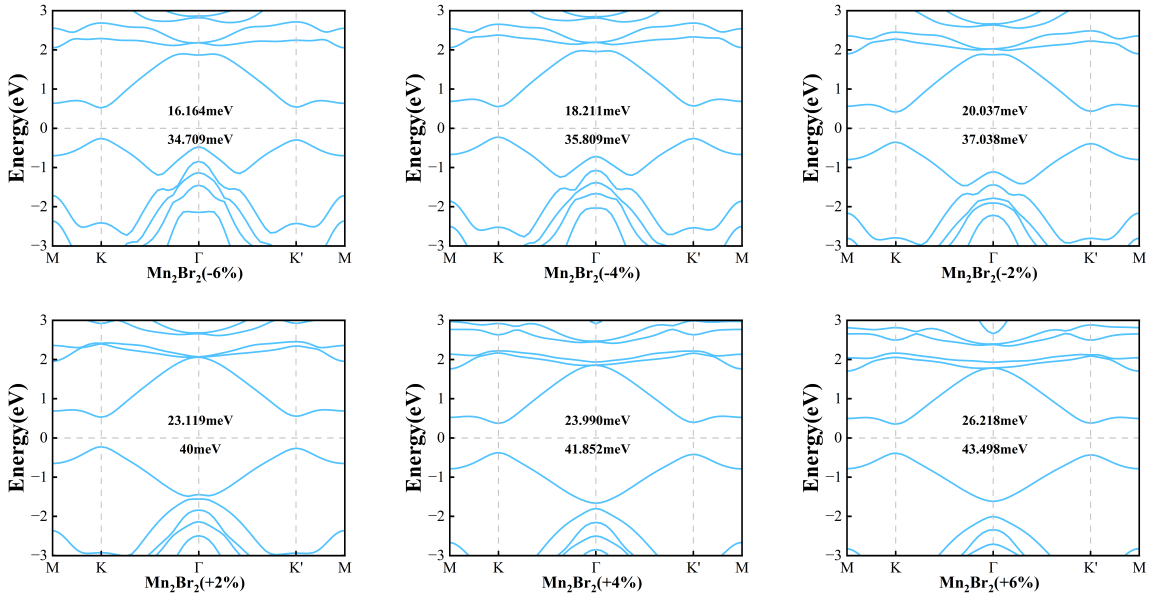

FIG. S16: Band structures of Mn_2Br_2 with SOC under different strain values.

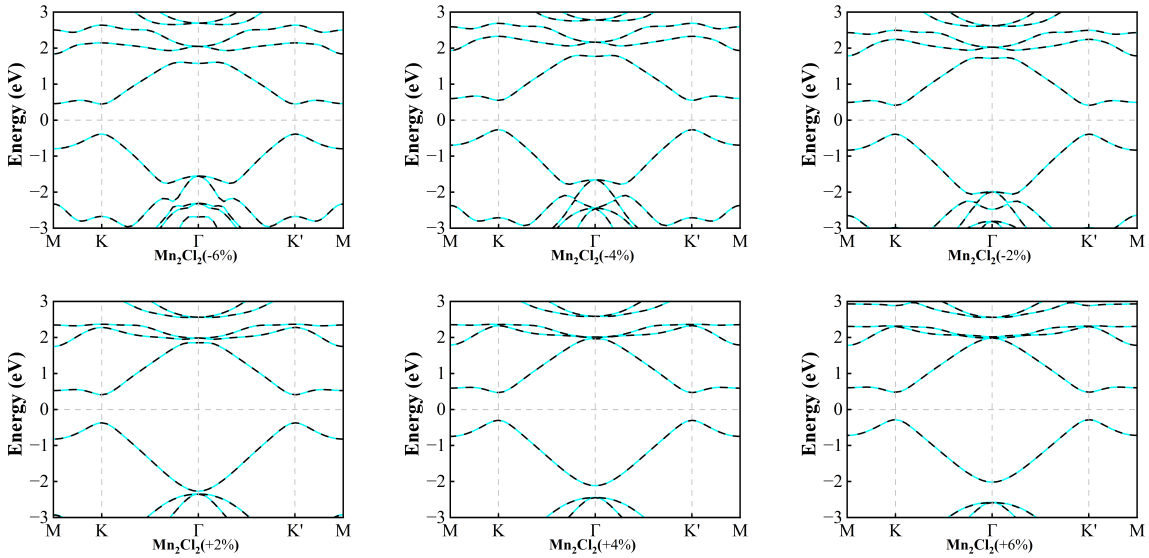

FIG. S17: Band structures of Mn_2Cl_2 without SOC under different strain values.

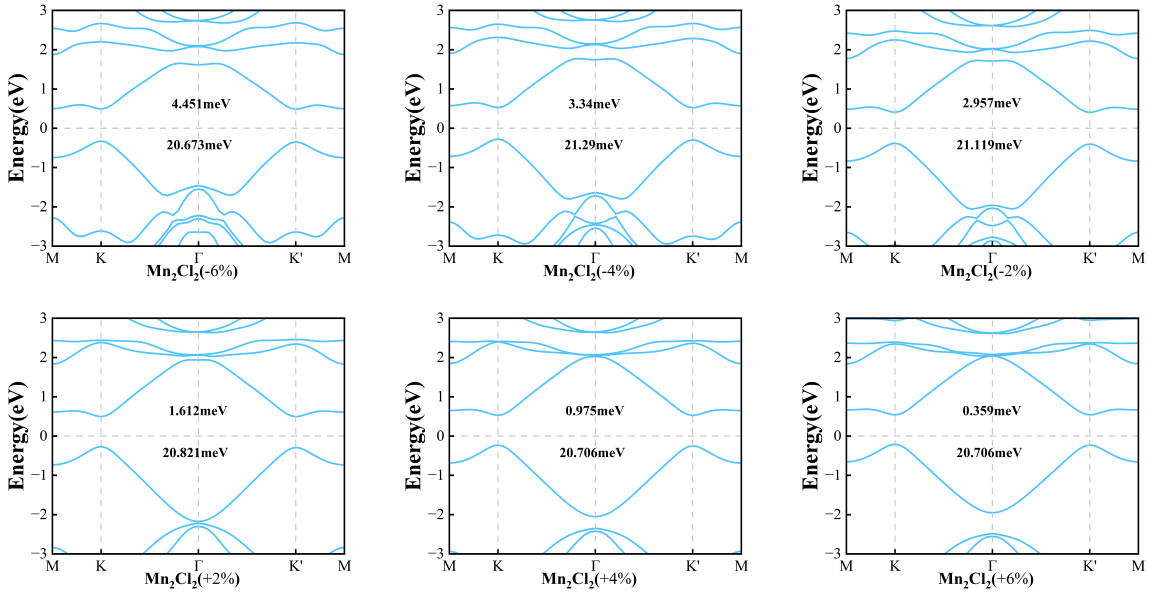

FIG. S18: Band structures of Mn_2Cl_2 with SOC under different strain values.

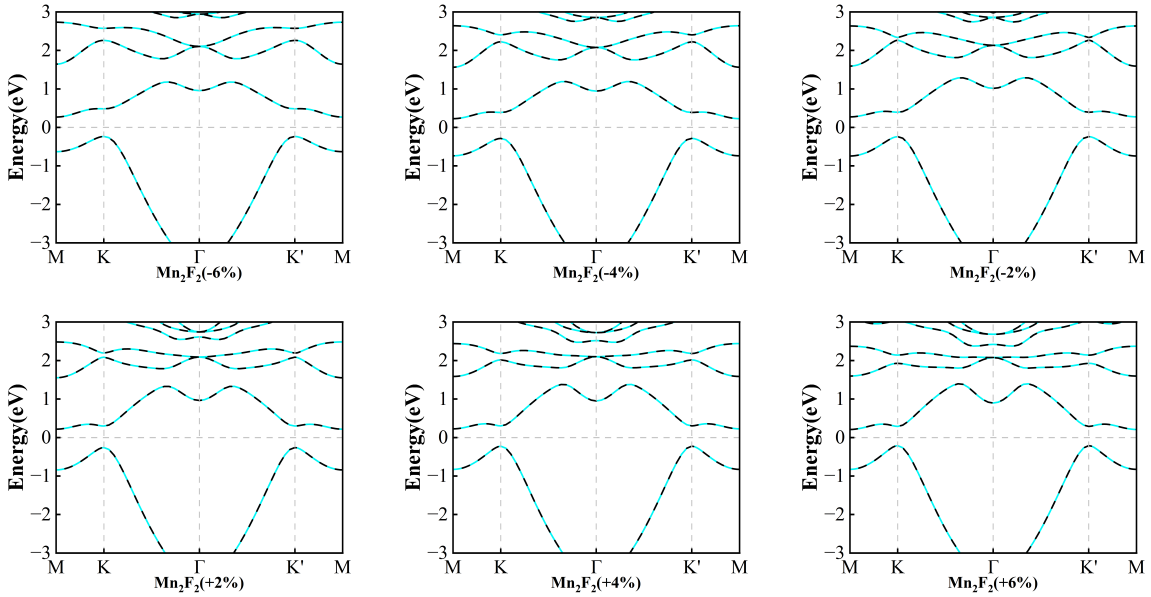

FIG. S19: Band structures of Mn_2F_2 without SOC under different strain values.

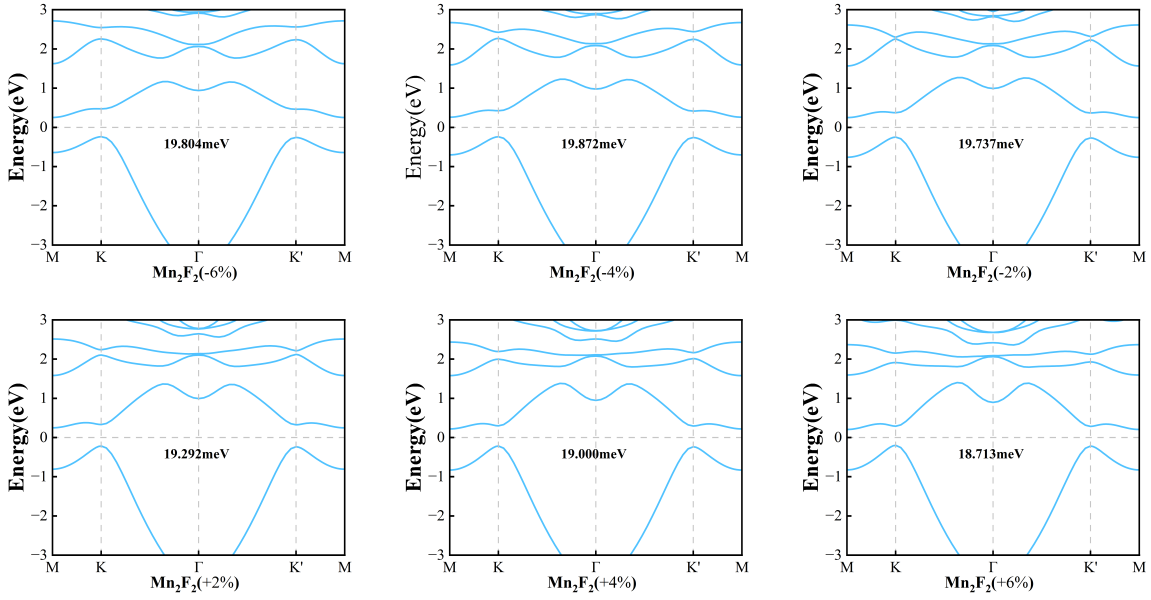

FIG. S20: Band structures of Mn_2F_2 with SOC under different strain values.

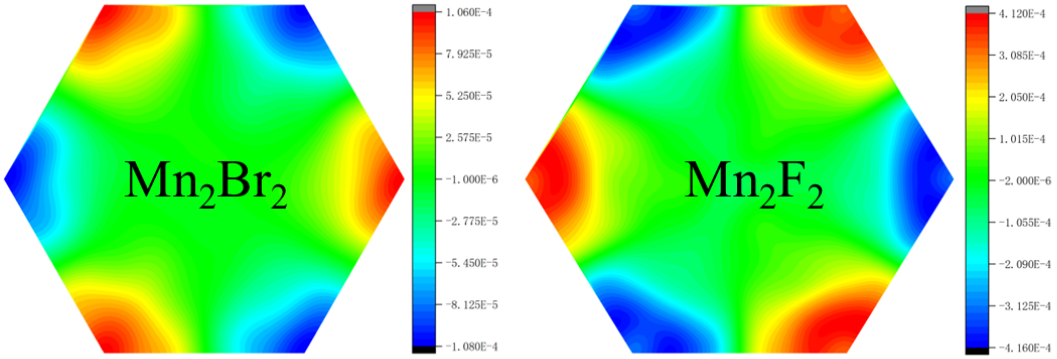

FIG. S21: Berry curvatures of monolayer Mn_2Br_2 and Mn_2F_2.