Electronic Supplementary Information for

Intrinsic valley polarization and manipulation by interfacial coupling in two-dimensional GdIBr/In_2Se_3 van der Waals multiferroic heterojunction

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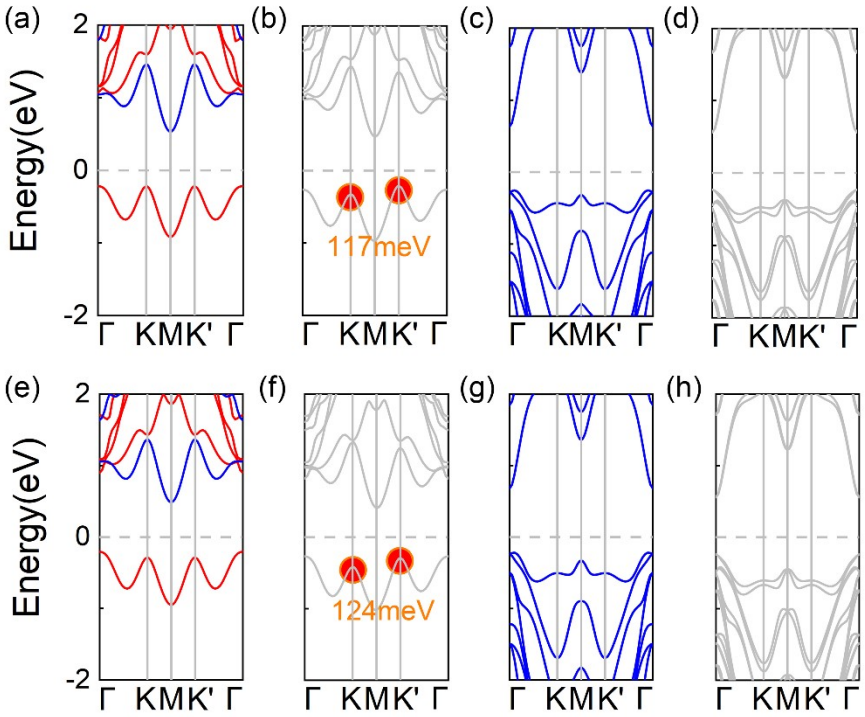

Fig. S1 The band structure of GdIBr (a) without and (b) with SOC. The band structure of In_2Se_3 (c) without and (d) with SOC. With a fixed lattice constant of 4.06 Å, the band structure of GdIBr is shown in (e) without and (f) with SOC. With a fixed lattice constant of 4.06 Å, the band structure of In_2Se_3 is shown in (g) without and (h) with SOC.

Table S1. In model-l,3,5,7↓ and model-2,4,6,8↑, the total energy (eV) of the GdIBr/In_2Se_3 heterojunction, and the energies of the ML GdIBr and In_2Se_3, respectively.

	E_{GdIBr/In_2Se_3}	E_{GdIBr}	$E_{In_2Se_3}$
model-1↓	–42.136161	–21.670765	–18.481858
model-2↑	–42.083700	/	/
model-3↓	–42.101599	/	/
model-4↑	–42.064838	/	/
model-5↓	–42.127538	/	/
model-6↑	–42.076853	/	/
model-7↓	–42.130557		
model-8↑	–42.079054		

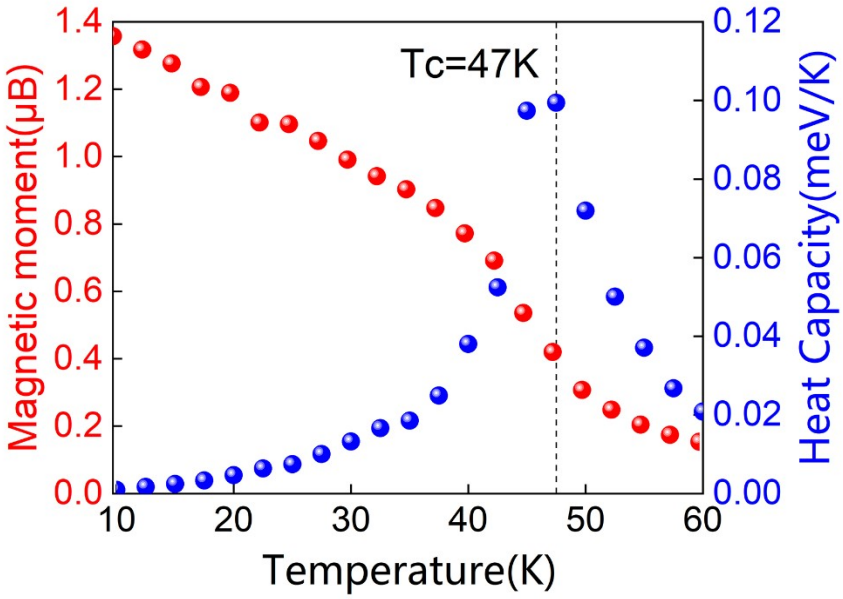

Fig. S2 The magnetic moment and heat capacity as functions of temperature for ML CrI_3.

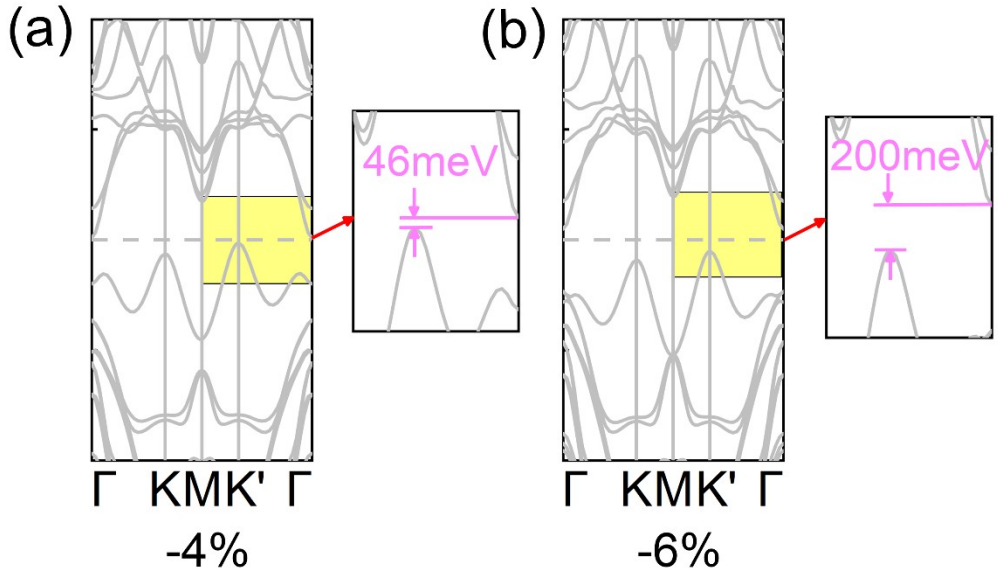

Fig. S3 Band structure and enlarged plots near the Fermi level when –4% and –6% compressive strains are applied to model-2↑.

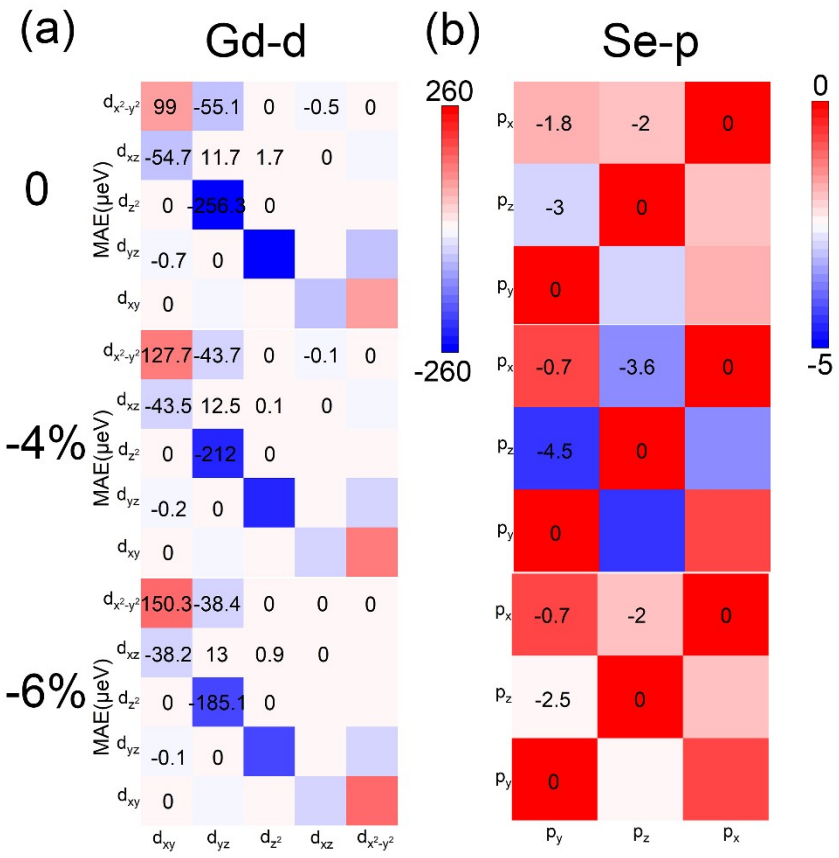

Fig. S4 The orbital resolved MAE of (a) Gd-*d* and (b) Se-*p* for model-2↑ under no strain, –4%, and –6% strain.

Table S2. The orbital resolved MAE of Gd-*d* and Se-*p*, the work function and bader charge of model-2↑ under no strain, –4%, and –6% strain.

	Gd-*d* (μeV)	Se-*p* (μeV)	Work function (eV)	Bader charge (eV)
0	–509.8	–13.6	4.005	0.022
–4%	–318.4	–17.6	4.009	0.012
–6%	–195.2	–10.4	4.010	0.010