Supporting Information

Reversible switching of half-metallicity and magnetic order in 2D FeI_2/Al_2Te_3 van der Waals multiferroics tailored by ferroelectric polarization

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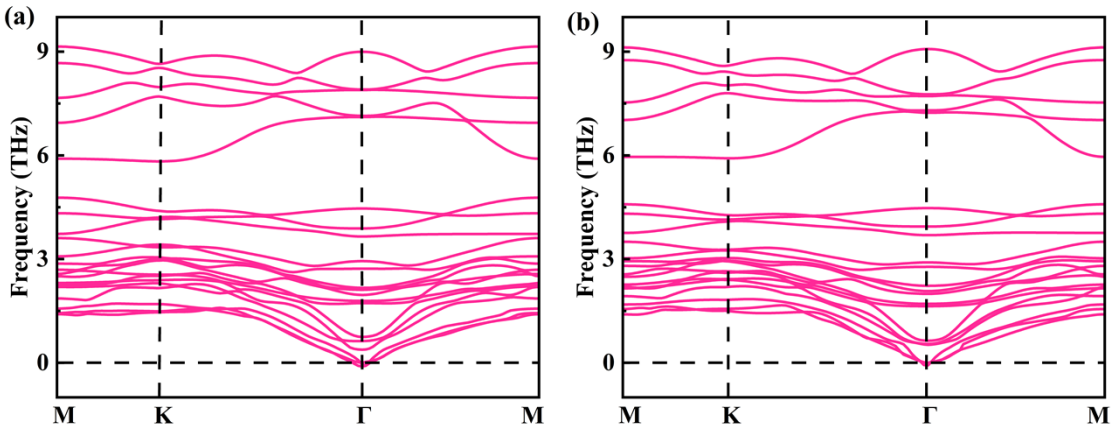

Figure S1. Phonon spectrums of heterostructures (a) type III P↑ and (b) type VI P↓.

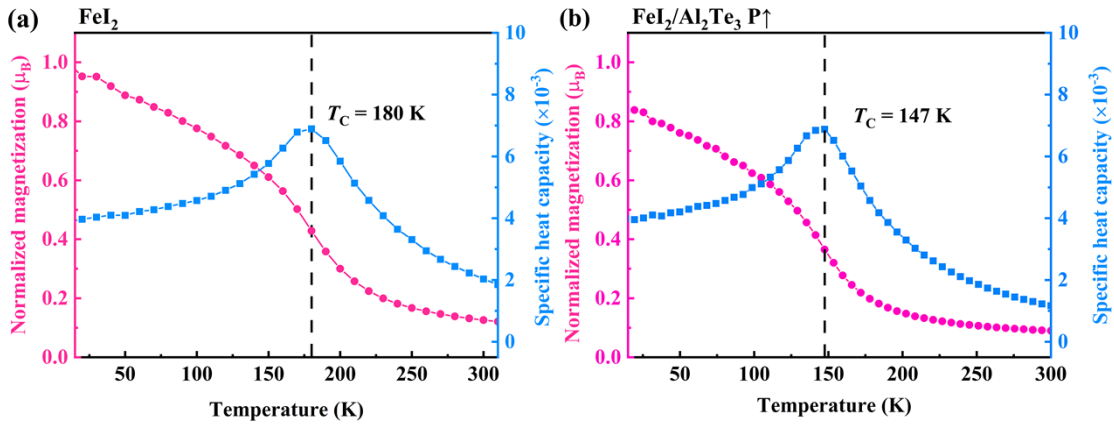

Figure S2. Temperature dependence of the normalized magnetization and specific heat capacity for: (a) the isolated FeI_2 monolayer, (b) the FeI_2/Al_2Te_3 heterostructure under P↑ polarization.

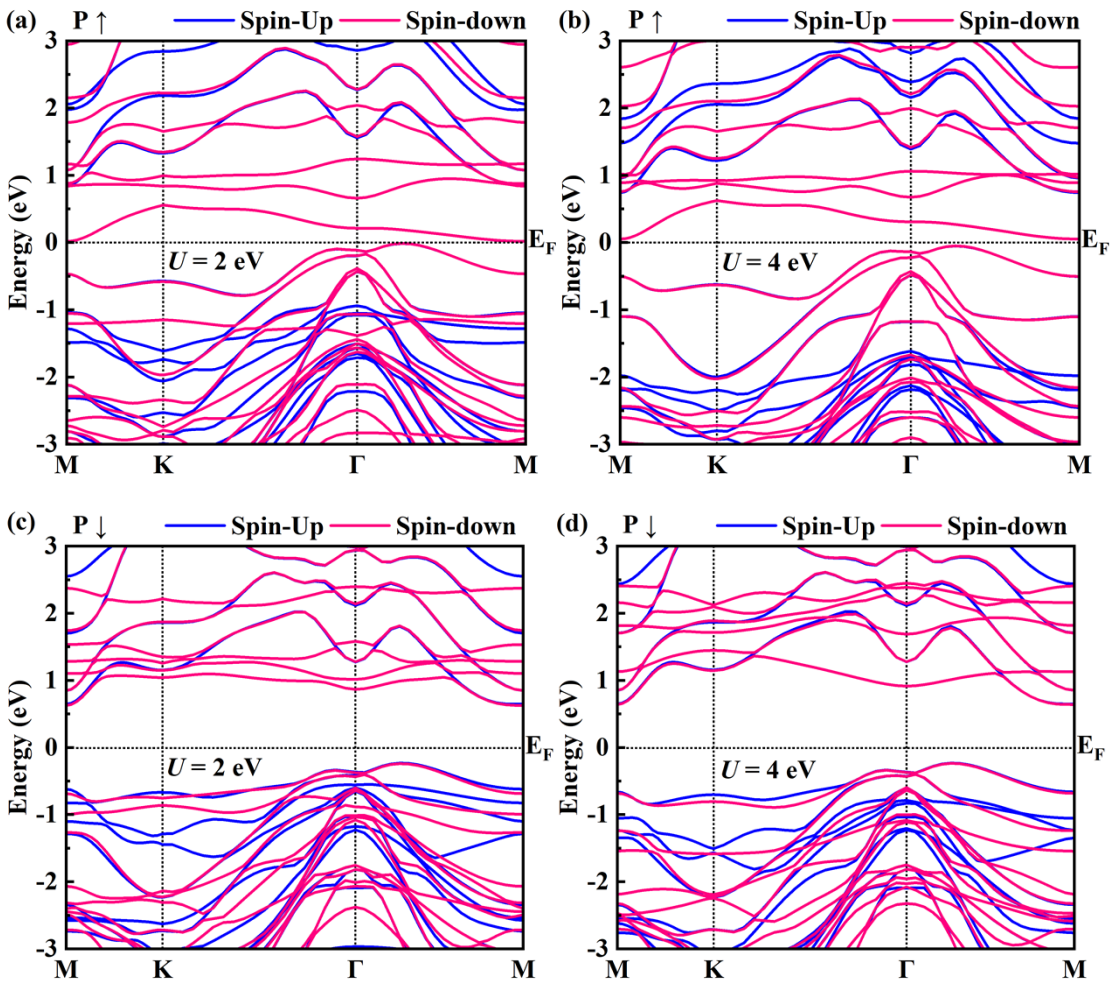

Figure S3. The comparisons of the spin-resolved band structures of (a)-(b) type III P↑ and (c)-(d) type VI P↓ under different *U* values.

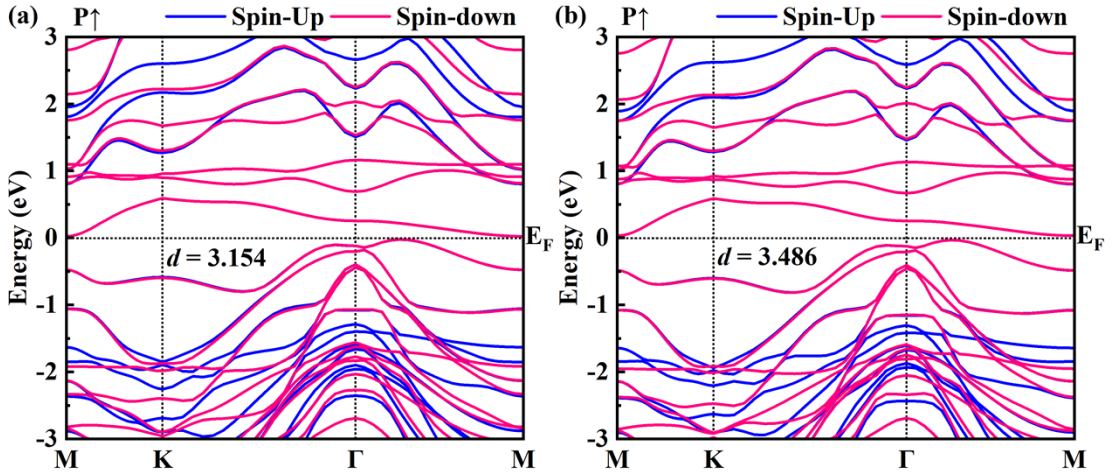

Figure S4. Band structures of type III P↑ calculated at U = 3 eV for different interlayer spacings: (a) d = 3.154, (b) d = 3.486.

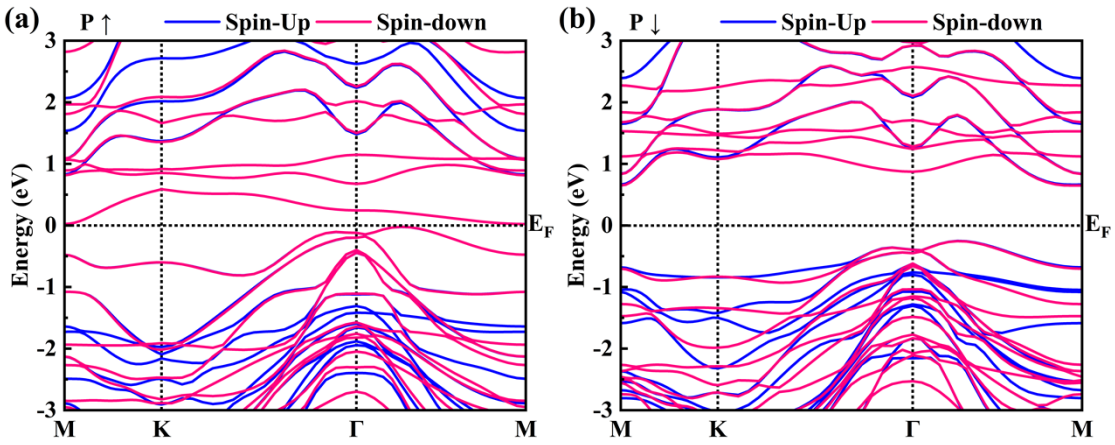

Figure S5. Band structures of (a) type I P↑ and (b) type IV P↓ calculated at U = 3 eV.

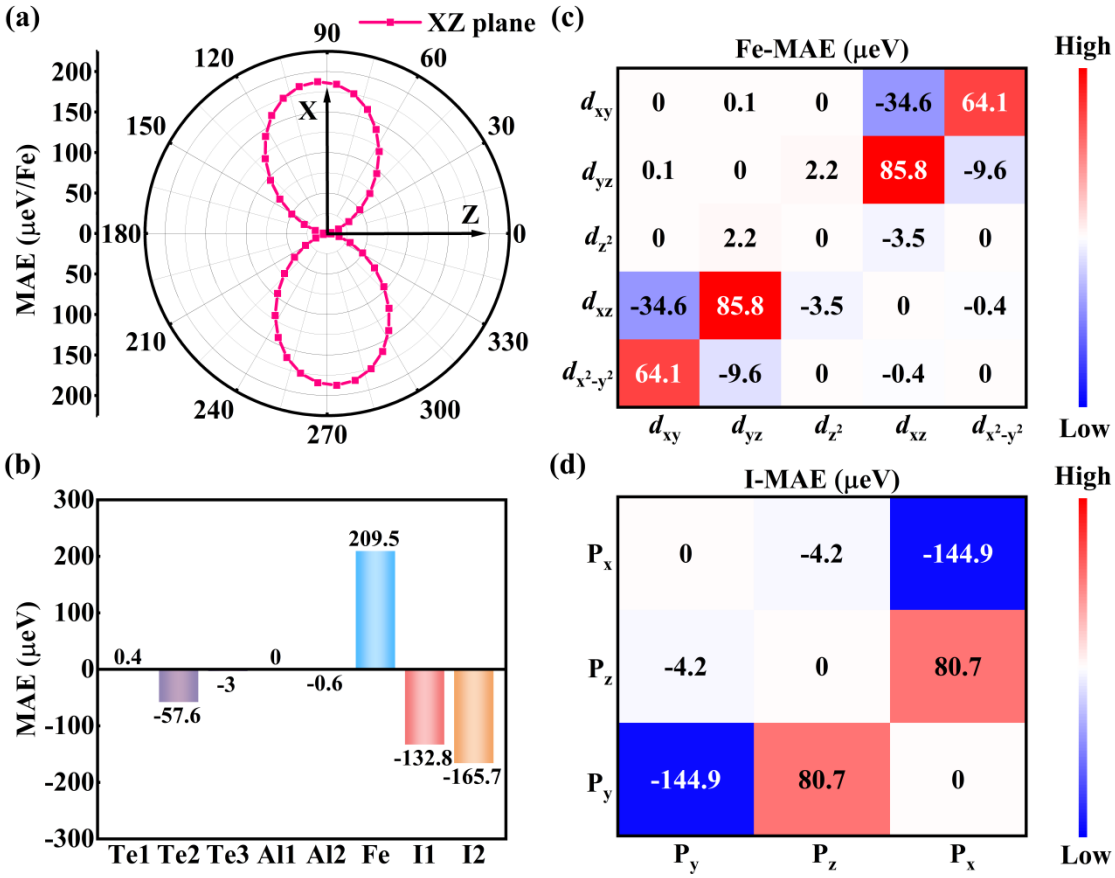

Figure S6. (a) Evolution of the MAE for the type I P↑ in the FeI_2/Al_2Te_3 heterostructure, within the *xz*-plane. (b) Atoms-resolved MAE for the type I P↑ in the FeI_2/Al_2Te_3 heterostructure. Orbital-resolved MAE of (c) Fe atoms and (d) I2 atoms for the type I P↑ in the FeI_2/Al_2Te_3 heterostructure.

Table S1. The $d_{Fe\text{-}Fe}$ and θ represent the Fe-Fe distance and Fe-I-Fe angle, the energy differences ($\Delta E = E_{AFM} - E_{FM}$) between the FM and AFM configurations, the magnetic moment of Fe is represented by M_{Fe}, and MAE represents the magnetocrystalline anisotropy energy of the type I P↑ and type IV P↓ in the FeI_2/Al_2Te_3 heterostructures.

	$d_{Fe\text{-}Fe}$ (Å)	θ (deg)	ΔE (eV)	M_{Fe} (μ_B)	MAE (μeV/Fe)
Type I P↑	4.08	91.9	0.067	3.535	209.5
Type IV P↓	4.08	91.5	−0.039	3.575	257.7