

Supporting Information

Engineering the ligand states by surface functionalization: A new way to enhance the ferromagnetism of CrI₃

Table S1. 2×2 supercell of pristine monolayer CrI₃. The exchange energy (EX) and magnetic anisotropy energy (MA) that calculated by PBE and PBE+U with different values of U-J.

U-J (eV)	PBE	1.3	2.0	2.3	3.3
EX (meV)	157.2	186	201.2	208.0	231.2
MA (meV)	6.2	2.8	3.7	4.2	7.6

Table S2. 2×2 supercell of CrI₃ adsorbed with single F-atom (F@CrI₃)

U-J (eV)	PBE	1.0	2.0	3.0	4.0
EX (meV)	339.1	341.5	348.7	361.2	380.0
MA (meV)	7.6	7.5	8.5	9.7	11.2

Table S3. 2×2 supercell of FF@CrI₃

U-J (eV)	PBE	1.0	2.0	3.0	4.0
EX (meV)	3037.6	2994.4	2905.2	2801.2	2690.0
MA (meV)	9.6	22.8	39.6	61.6	85.6

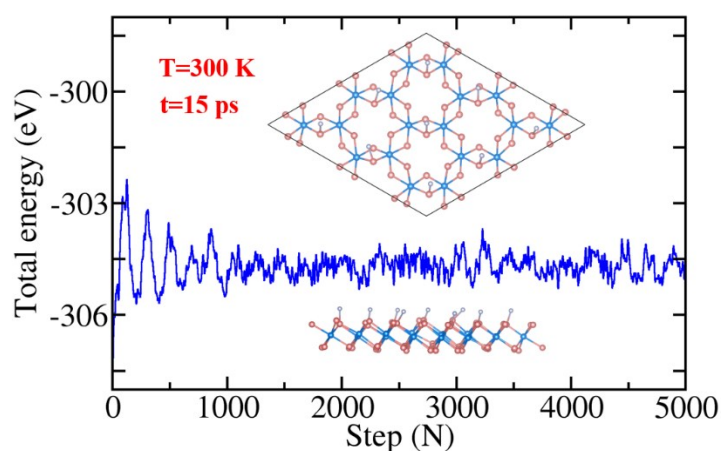


Figure S1. The molecular dynamical simulation result for CrI₃ adsorbed with 33.3% F-atom at T=300 K.

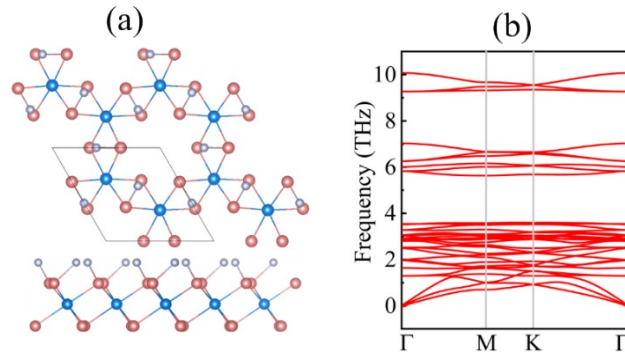


Figure S2. The atomic structure (a) and the phonon spectrum (b) of fully F-functionalized CrI₃

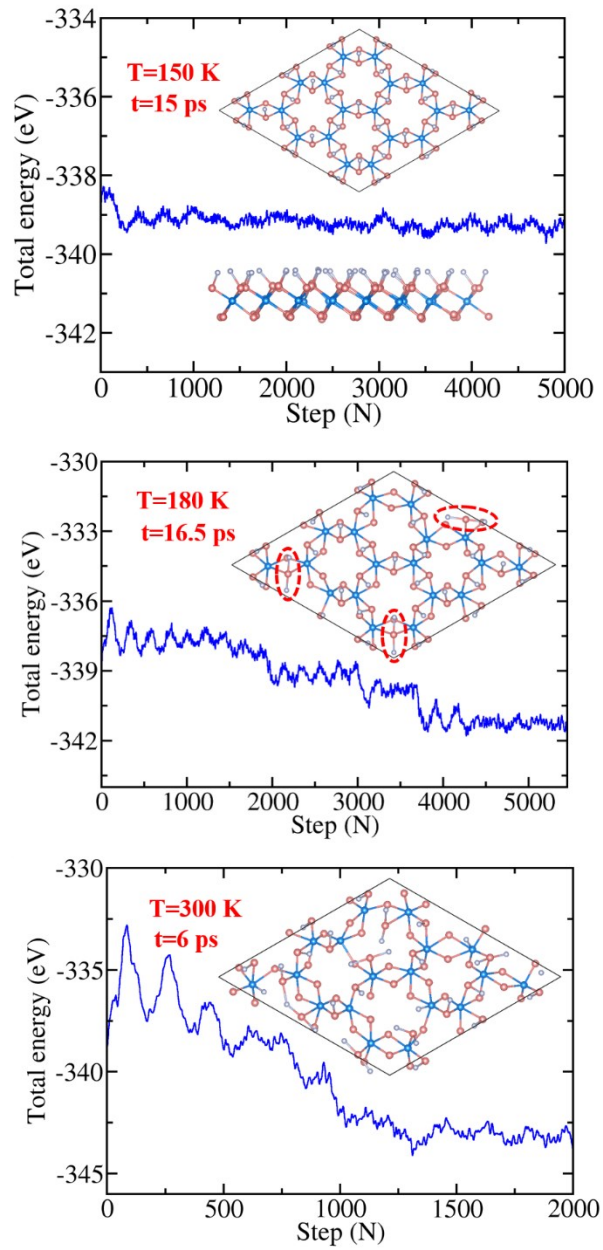


Figure S3. The molecular dynamical simulation results for FF@CrI₃ at different temperature.