Supplementary Information

Intra-Interlayer Competition: A Key Regulator for Sliding Ferroelectricity in Hydrogen-Functionalized Group-III Monochalcogenide Monolayer

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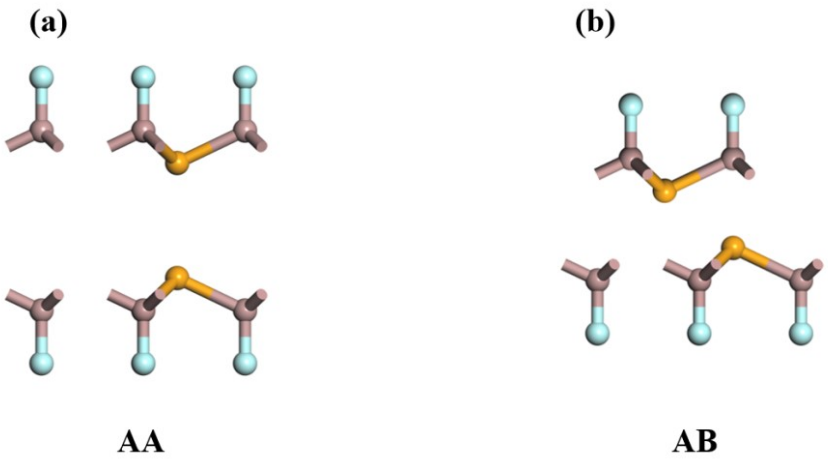

Fig. S1. Structures of GaSe(F@Ga). (a) AA stacking of GaSe(F@Ga), (b) AB stacking of GaSe(F@Ga).

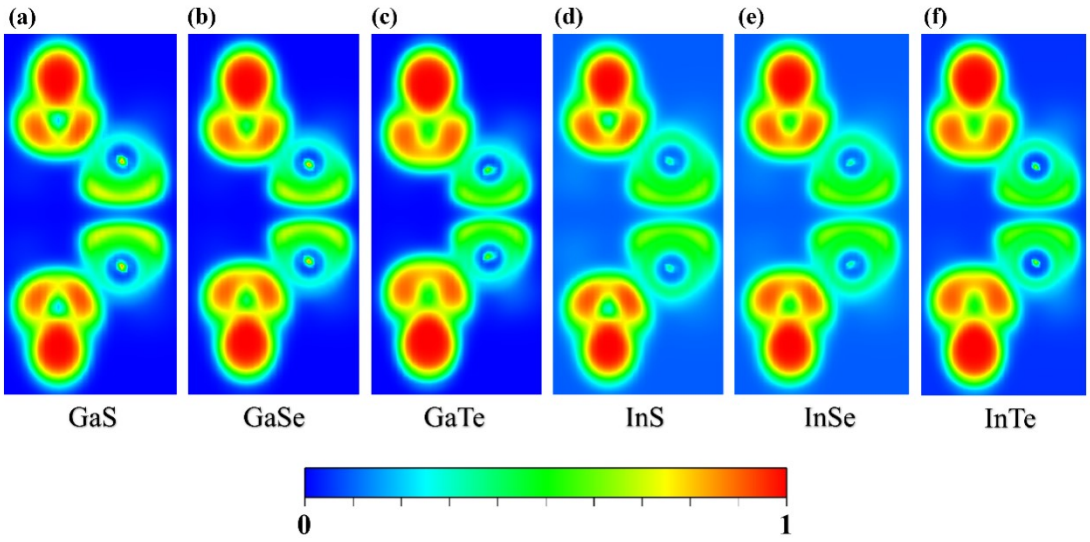

Fig. S2. ELF of *MX*(H@*X*). (a) ELF of GaS, (b) ELF of GaSe, (c) ELF of GaTe, (d) ELF of InS, (e) ELF of InSe and (f) ELF of InTe.

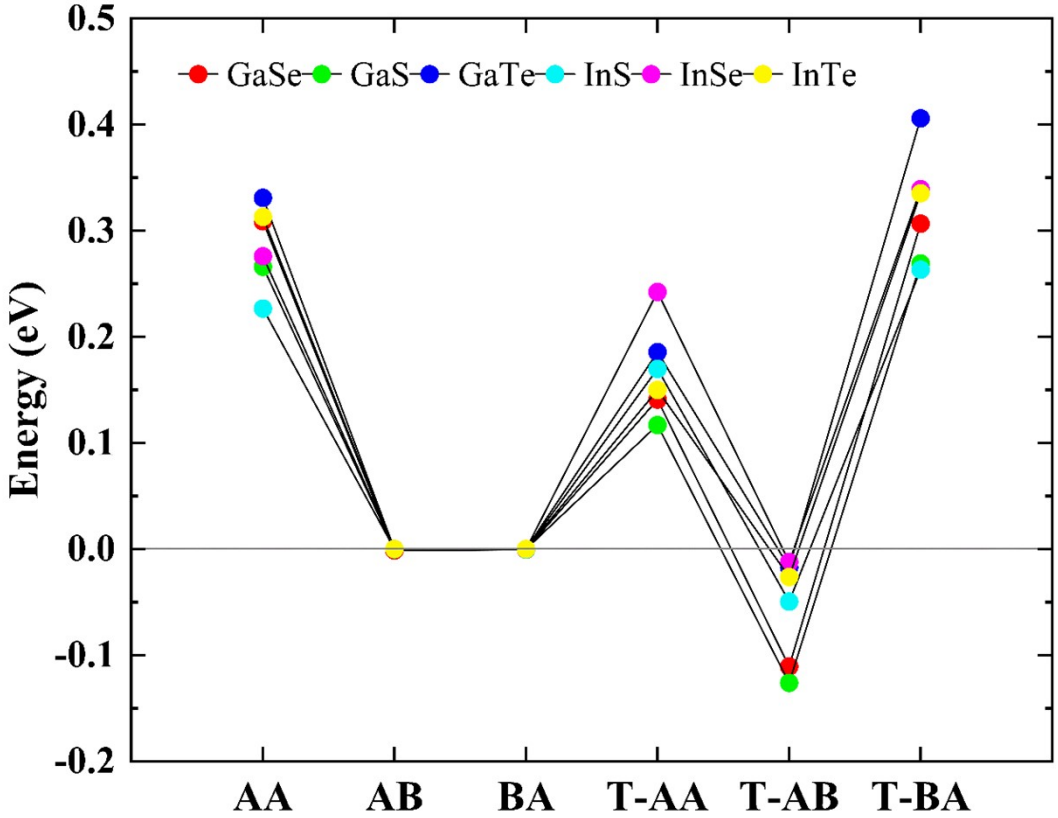

Fig. S3. Energy comparison of different stacking configurations in *MX*(H@*X*).

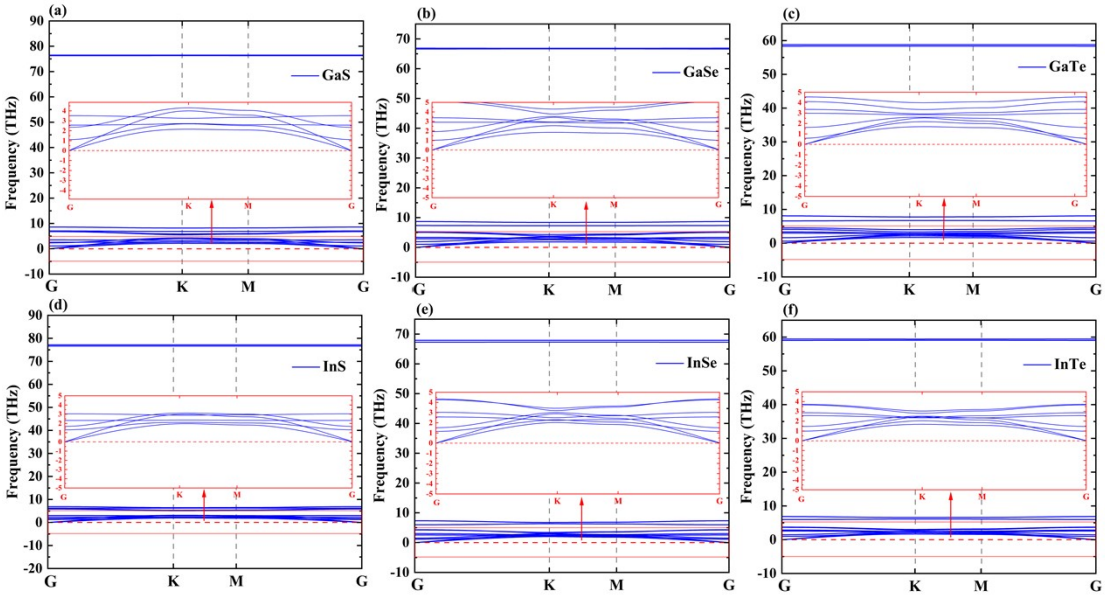

Fig. S4. Phonon spectra of AB stacking *MX*(H@*X*) and the embedded images are the partial enlarged views of the areas marked by the red boxes. (a) GaS, (b) GaSe, (c) GaTe, (d) InS, (e) InSe and (f) InTe.

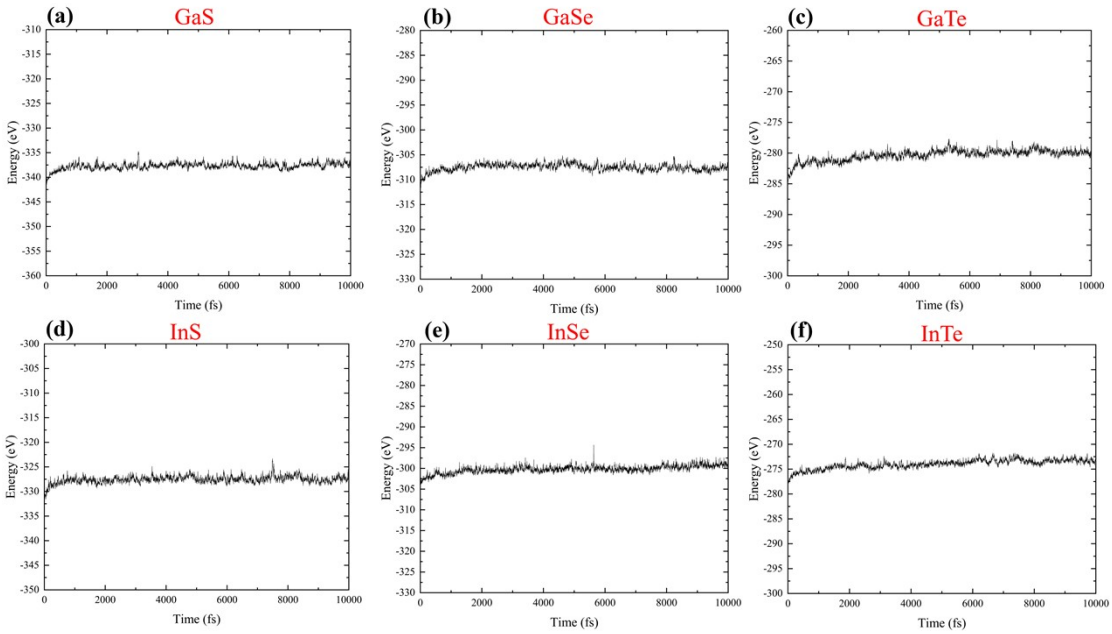

Fig. S5. Energy fluctuation of AB stacking *MX*(H@*X*) during 10 ps AIMD simulations at 300 K. (a) GaS, (b) GaSe, (c) GaTe, (d) InS, (e) InSe and (f) InTe.

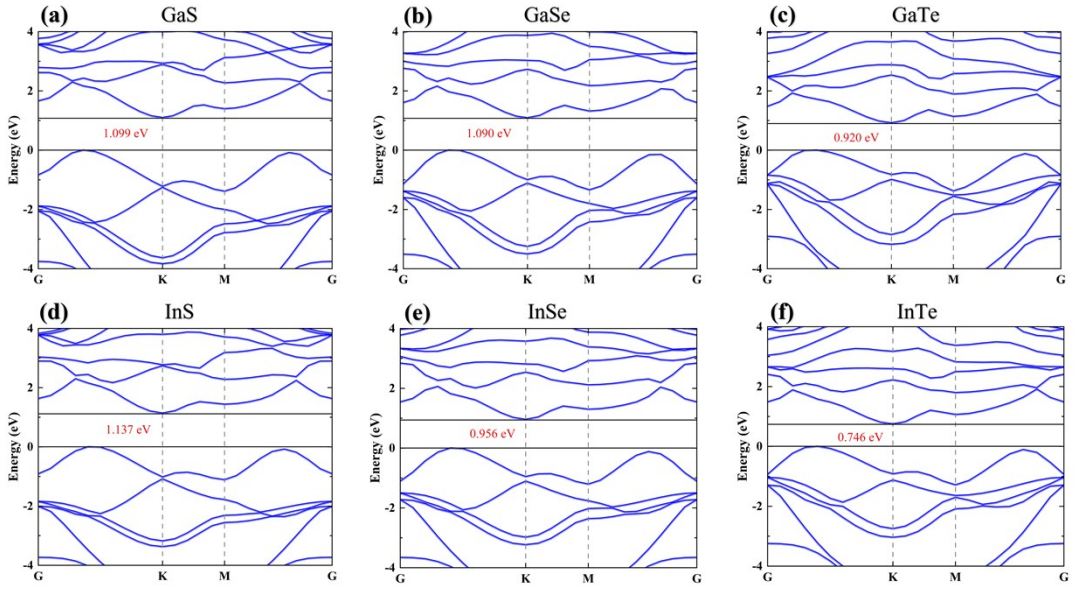

Fig. S6. Band structrues of AB stacking *MX*(H@*X*). (a) GaS, (b) GaSe, (c) GaTe, (d) InS, (e) InSe and (f) InTe.

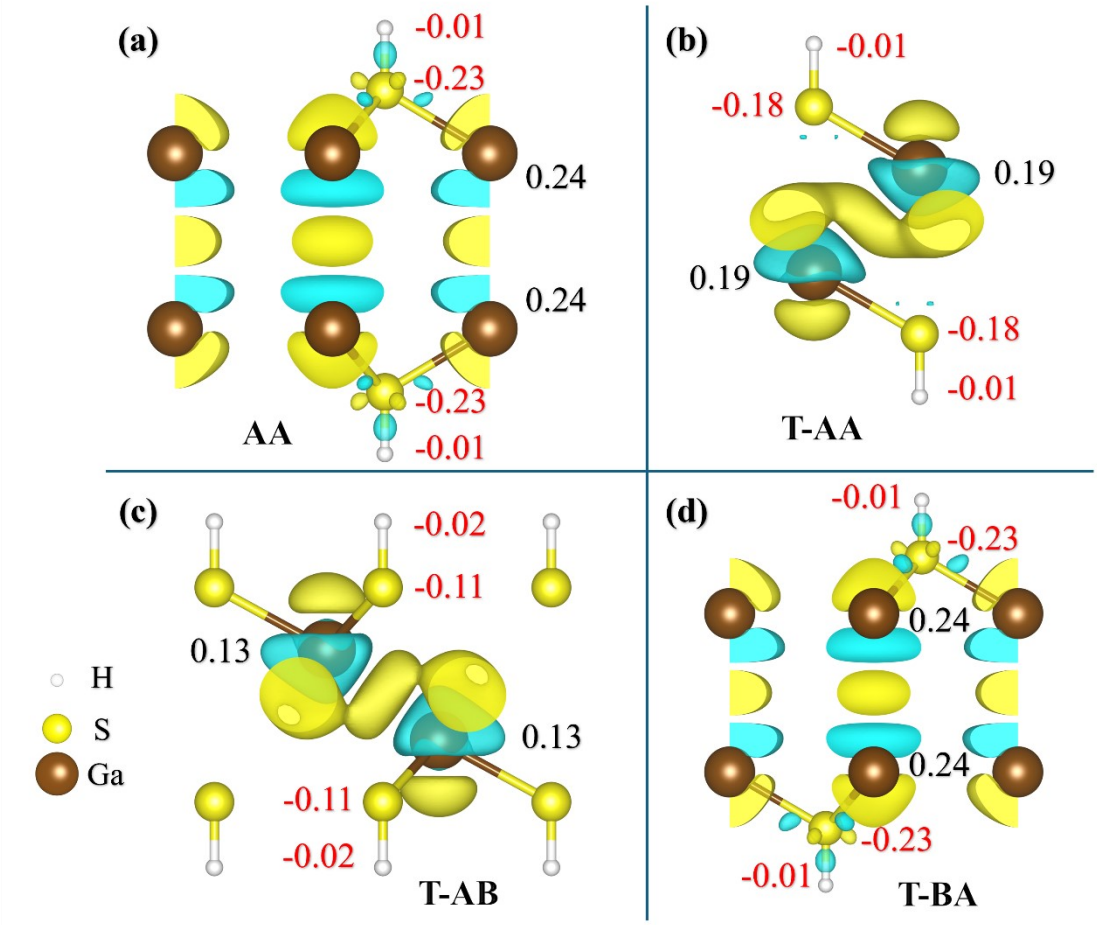

Fig. S7. Differential charge density plot and Mulliken charge analysis for non-ferroelectric stacking configurations in GaS(H@S). (a) AA stack, (b) T-AA stack, (c) T-AB stack and (d) T-BA stack.

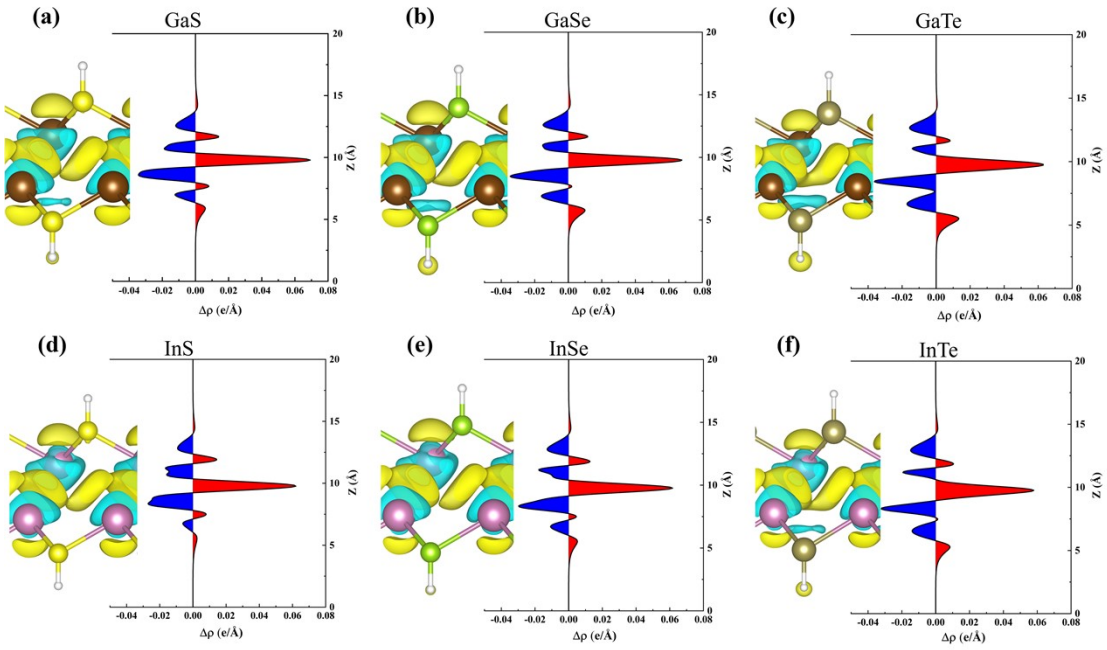

Fig. S8. Differential charge density plot and 1-D planar-averaged differential charge density along the z-direction of AB stacking *MX*(H@*X*). (a) GaS, (b) GaSe, (c) GaTe, (d) InS, (e) InSe and (f) InTe.

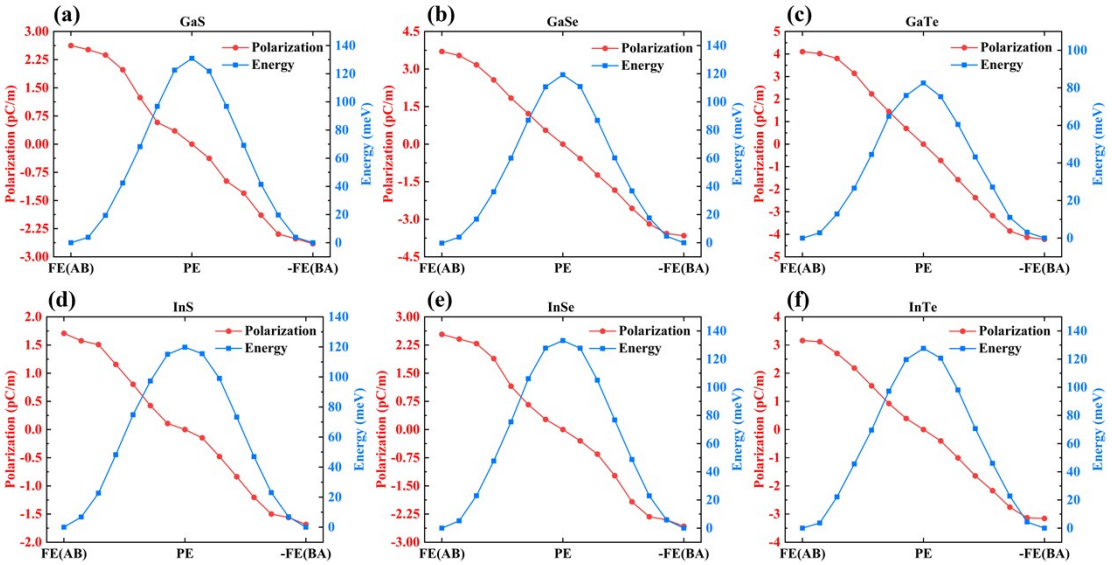

Fig. S9. Polarization intensity and energy change during the transition from the FE state to the -FE state for (a) GaS, (b) GaSe, (c) GaTe, (d) InS, (e) InSe and (f) InTe.

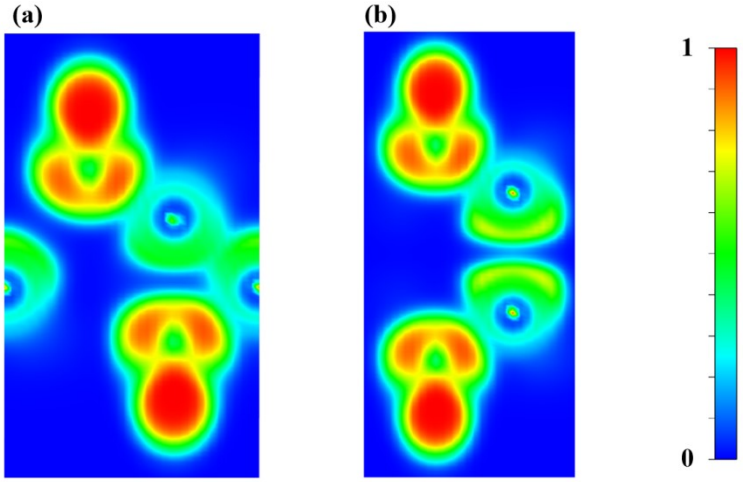

Fig. S10. ELF of GaSe(H@*X*). (a) ELF of AB configuration, (b) ELF of AA configuration.

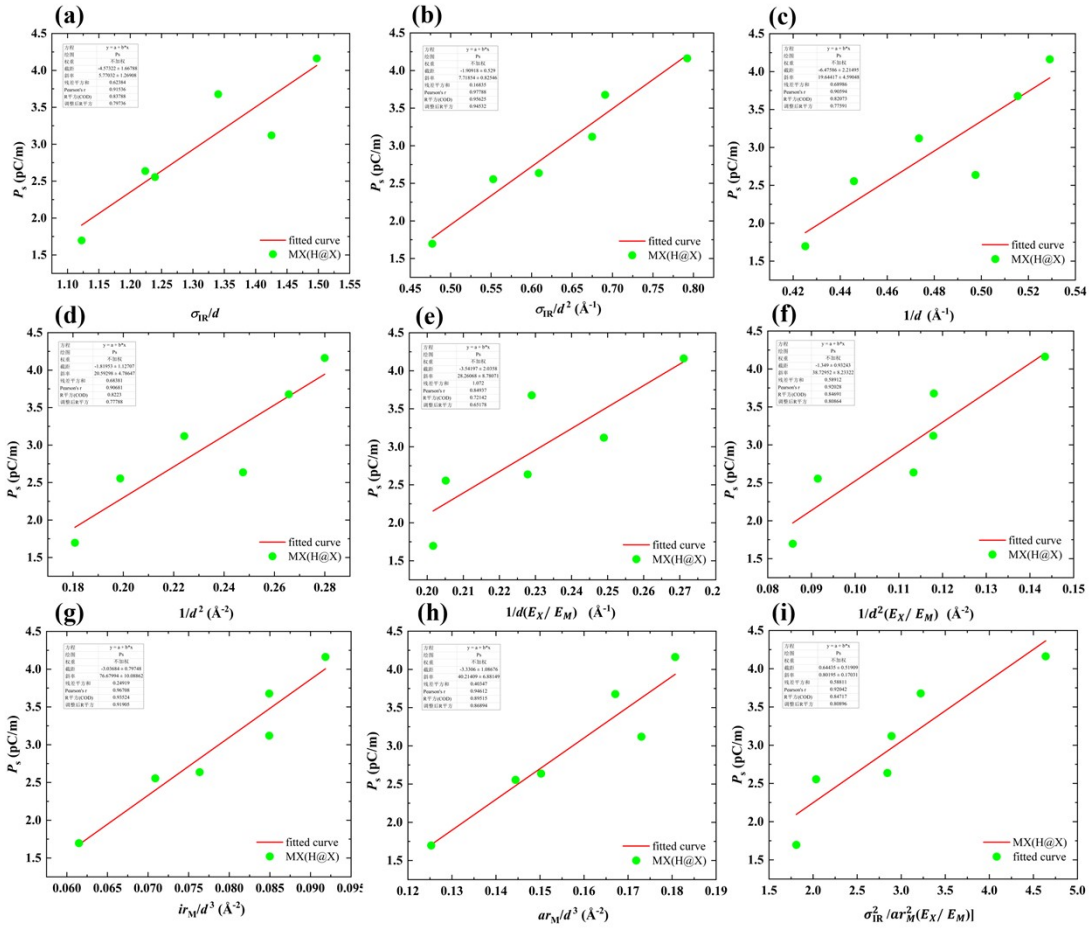

Fig. S11. Fitting relationship between spontaneous polarization P_s and descriptors in *MX* (H@*X*). ir_M is the ionic radius of *M* atom and ar_M is the atomic radius of *M* atom. (a) Relationship of σ_{IR}/d versus P_s. (b) Relationship of σ_{IR}/d^2 versus P_s. (c) Relationship of $1/d$ versus P_s. (d) Relationship of $1/d^2$ versus P_s. (e) Relationship of $1/d(E_X/E_M)$ versus P_s. (f) Relationship of $1/d^2(E_X/E_M)$ versus P_s. (g) Relationship of ir_M/d^3 versus P_s. (h) Relationship of ar_M/d^3 versus P_s. (i) Relationship of $\sigma_{IR}^2/[\ ar_M^2(E_X/E_M)\ ]$ versus P_s.