Electronic Supplementary Material (ESI) for Nanoscale Advances.

Multiferroic Iron Arsenide Monolayer

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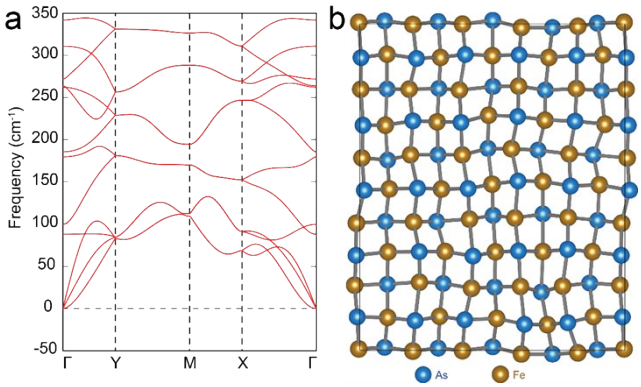

Fig. S1. (a) Calculated phonon spectra of the FeAs monolayer. (b) Snapshots of the FeAs monolayer at the end of 10 ps ab initio molecular dynamics simulations at 500 K.

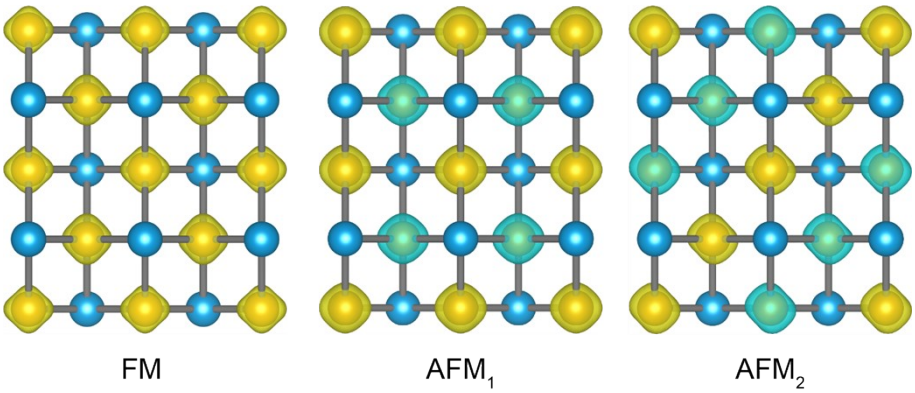

Fig. S2. Isosurface plots (0.67 e/Å^3) of spin density distribution.

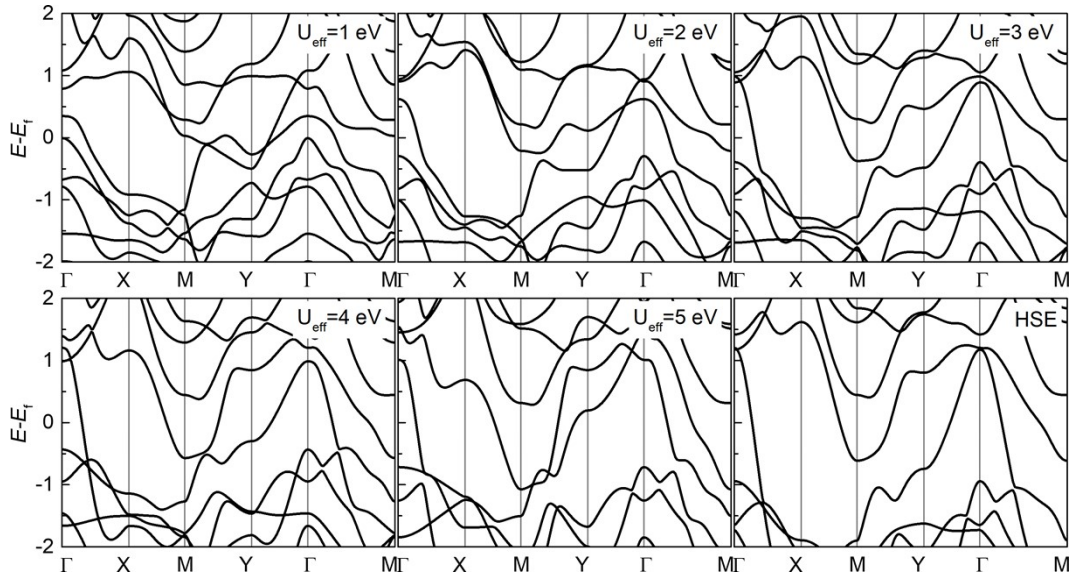

Fig. S3. Band structures of the FeAs monolayer in the AFM_1 state without the SOC calculated with the DFT+U method (the values of U_{eff} are provided) and HSE06 functional.

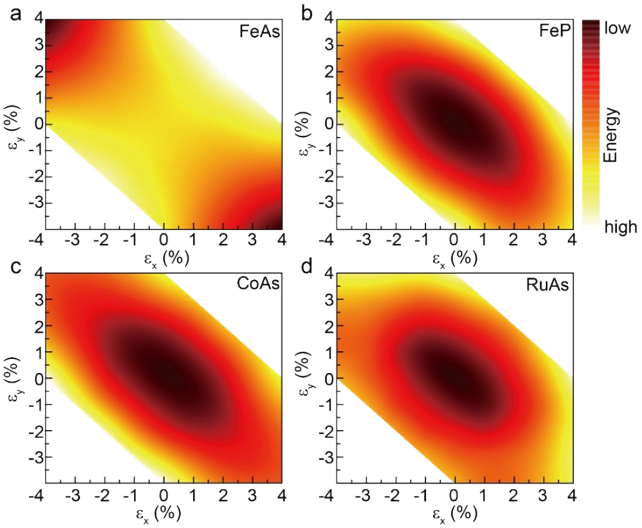

Fig. S4. The relationship between external strain (ε) and total energy of the FeAs (a), FeP (b), CoAs (c) and RuAs (d) monolayers. When $\varepsilon_x=\varepsilon_y=0$, the crystal structure of these materials is square.

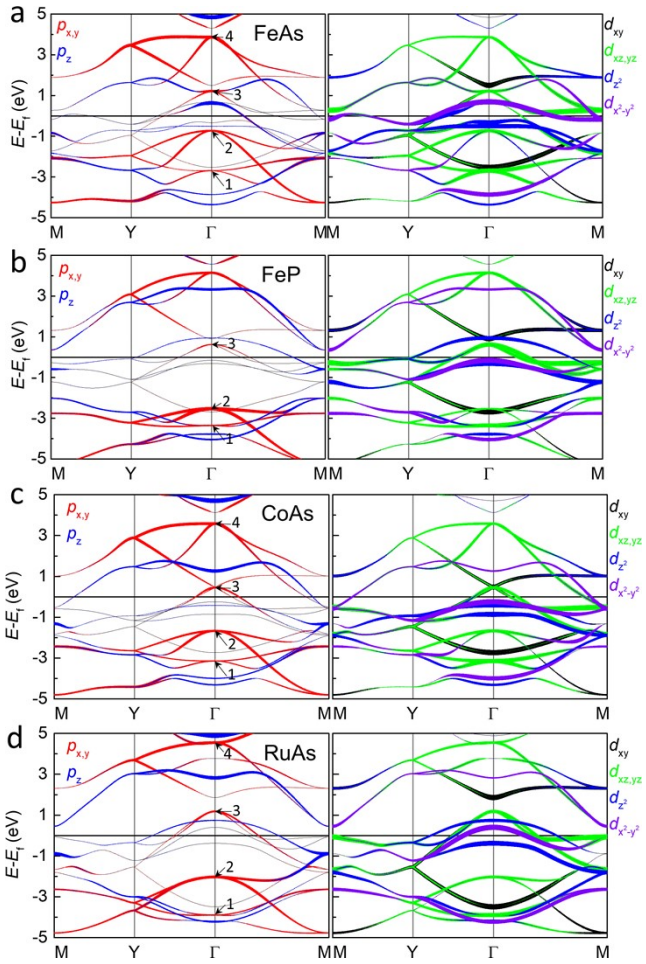

Fig. S5. The band structure without spin polarize of the FeAs (a), FeP (b), CoAs (c) and RuAs (d) monolayer. The contribution of $p_{x,y}$ and p_z orbitals to the Bloch states plotted in left panel are denoted by red and blue, respectively, while the contribution of d_{xy}, $d_{xz,yz}$, d_{z2} and d_{x2-y2} orbitals to the Bloch states plotted in the right panel are denoted by black, green, blue and violet, respectively. The four double degenerated states formed by $d_{xz,yz}$ and $p_{x,y}$ orbitals are denoted by 1, 2, 3 and 4.

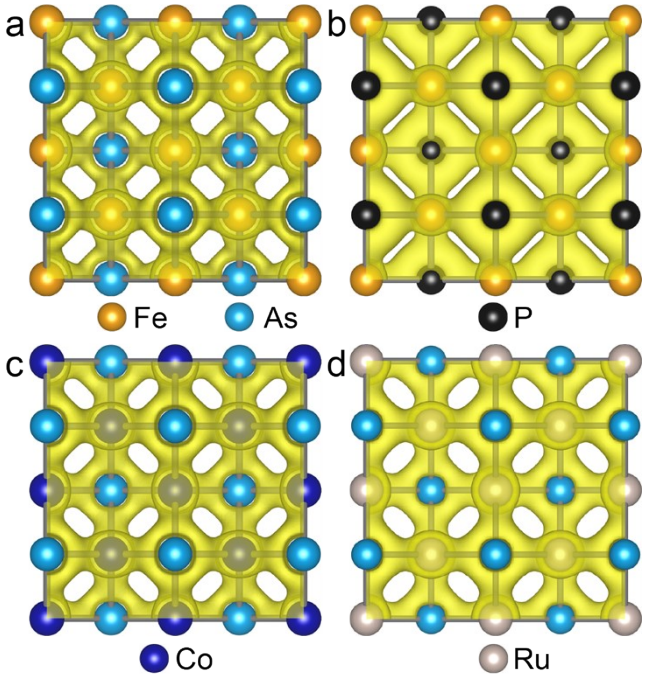

Fig. S6. Isosurface plots of the partial charge density to state 2 in FeAs (a), FeP (b), CoAs (c) and RuAs (d) monolayers with an isosurface value of 0.054 e/Å^3.

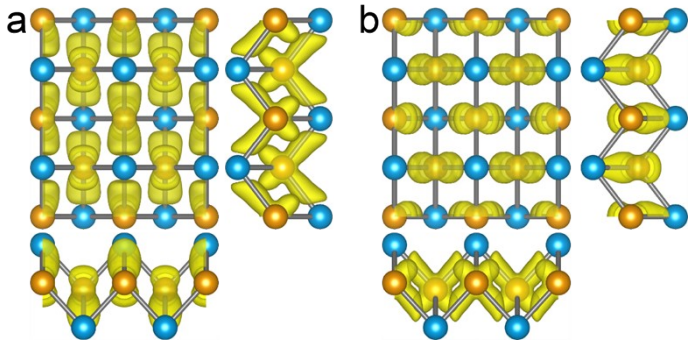

Fig. S7. Isosurface plots of the partial charge density to state 1 along *y*-axis direction (a) and *x*-axis direction (b) in FeAs monolayer with an isosurface value of 0.054 e/Å^3.

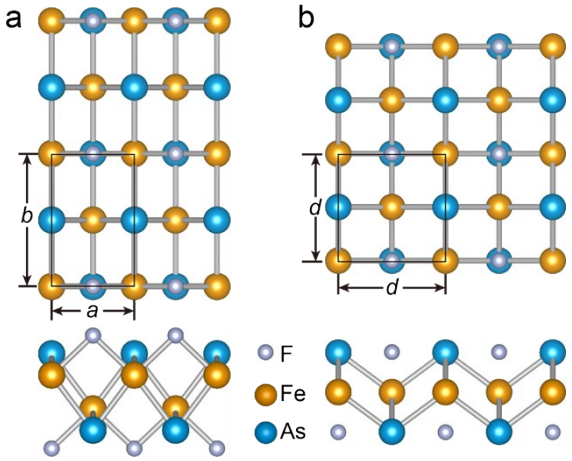

Fig. S8. Top and side views of the FeAsF monolayer at the initial state (a) and transition state (b), where the unite cell is denoted by the black rectangle. The origin, blue and grey balls represent Fe, As and F atoms, respectively. The lattice constant values of *a*,

b and *d* are 2.93, 4.71 and 3.91 Å, respectively.

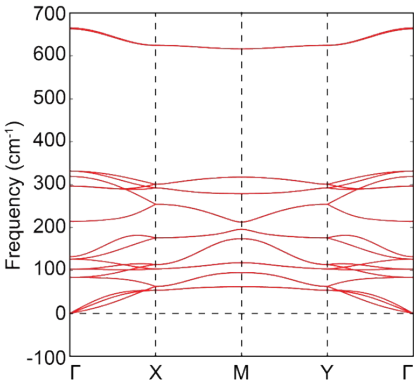

Fig. S9. Calculated phonon spectra of the fluorinated FeAs monolayer.

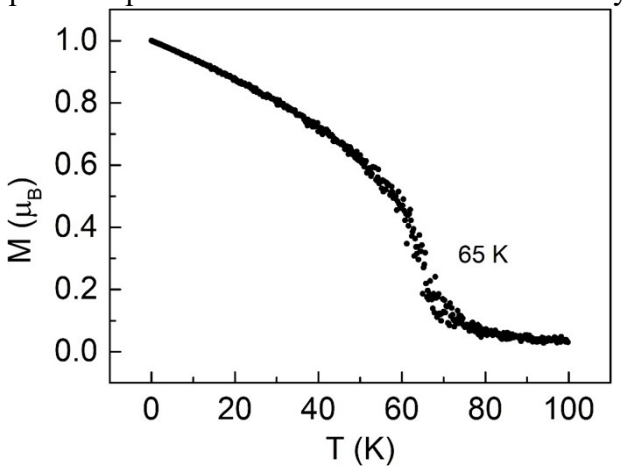

Fig. S10 Magnetic moments in the FeAsF monolayer as functions of temperature obtained from Monte Carlo simulations.