Supporting Information

Room Temperature Ferromagnetism and Antiferromagnetism in Two-Dimensional Iron Arsenides

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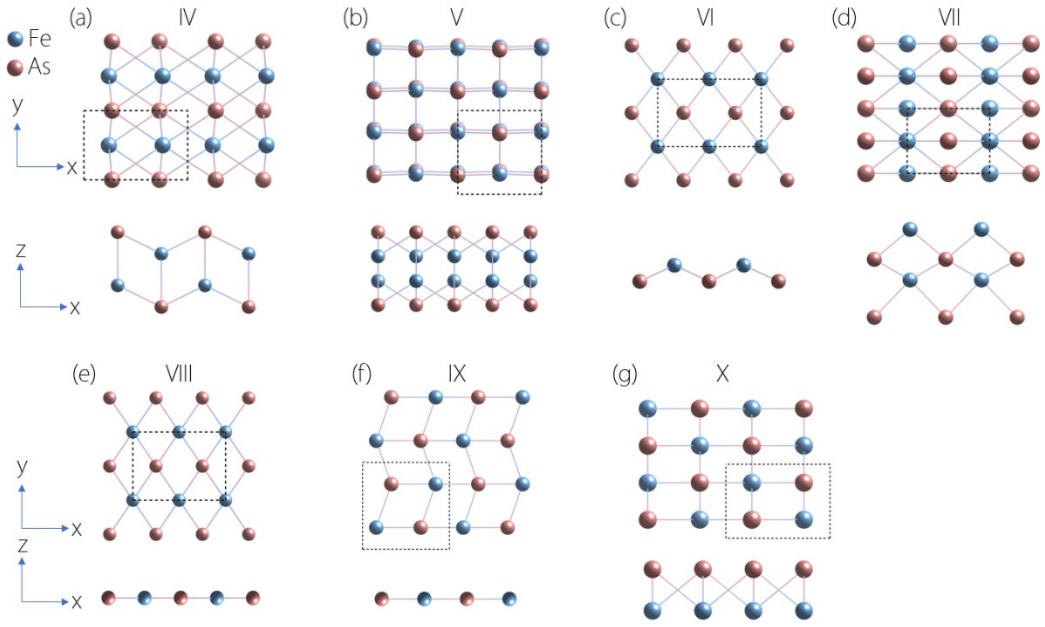

Figure S1 Structures of FeAs monolayers predicted by our structural search.

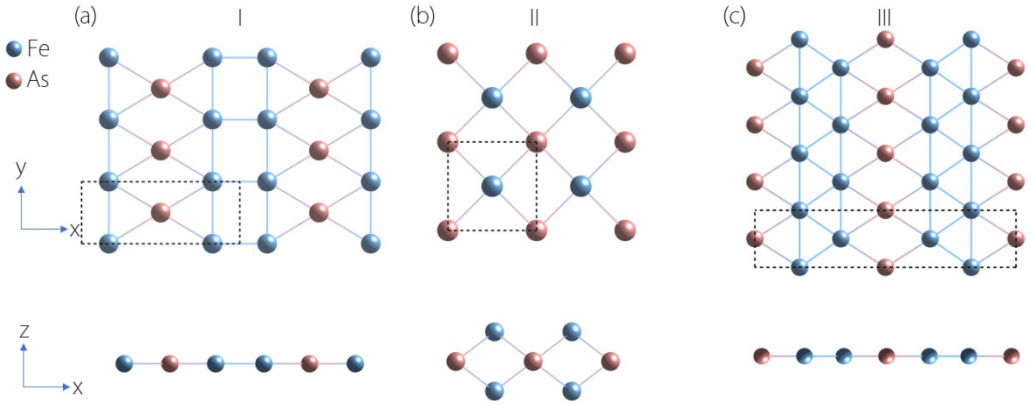

Figure S2 Structures of Fe_2As monolayers predicted by our structural search.

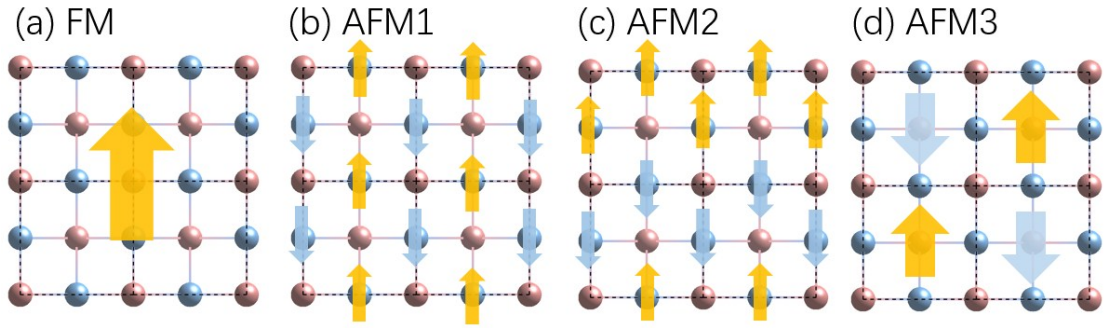

Figure S3 (a-d) Different magnetic configurations for the FeAs-III monolayer. The arrows indicate the spin directions on the Fe atoms. AFM1 is found to be the ground state.

We adopted a 2 x 2 supercell and compared the different magnetic configurations for FeAs-III on the PBE+U level. As shown in Fig. S3, AFM1 has the lowest energy (set as the reference), which is lower than that of the FM (+1.606 eV per cell), AFM2 (+0.581 eV per cell) and AFM3 (+0.149 eV per cell).

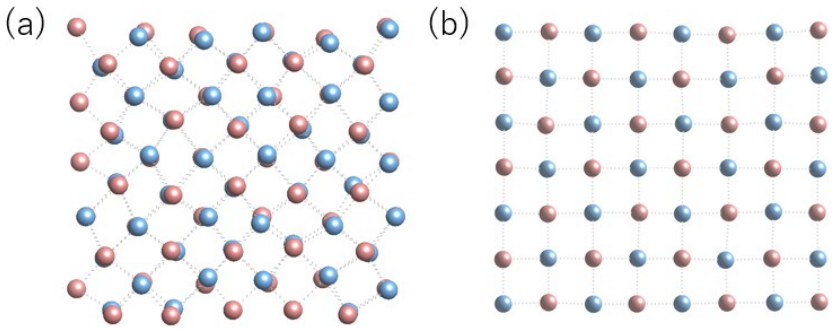

Figure S4 Snapshots of (a) FeAs-I and (b) FeAs-III monolayer after 5 ps AIMD simulation at 300 K.

The thermal stability of FeAs monolayers at room temperature were examined by performing spin-polarized ab initio molecular dynamics simulations (AIMD). In this calculation, the 3 x 3 supercells were constructed and a Nose-Hoover thermostat at 300 K was employed. After heating up and maintained at the targeted temperature for 5 ps with a time step of 1 fs, we find FeAs-I and III monolayer can retain their structures, indicating they are thermally stable at room temperature.

To test if these FeAs monolayers can sustain themselves (i.e., to form a freestanding membrane), we adopt the standard approach [Nano Lett. 8, 2442 (2008)] by calculating their individual in-plane stiffness constant $C = (1/A_0)(\partial^2 E_S/\partial \varepsilon^2)|_{\varepsilon=0}$, where A_0 is the equilibrium area, E_S is the strain energy, and ε is the applied uniaxial strain. We find C $\approx$1.15 eV/Å^2 for FeAs-I and 0.49 eV/Å^2 for FeAs-III. Consider the deformation of a freestanding FeAs flake under gravity. From the elastic theory, by balancing gravity and 2D strain energy, we find the ratio between the out-of-plane deformation and the lateral scale for a 10^4 μm^2 flake is as small as 10^3~10^{-4} for both monolayers. This suggests that they are strong enough to form freestanding 2D structures even without support of a substrate.

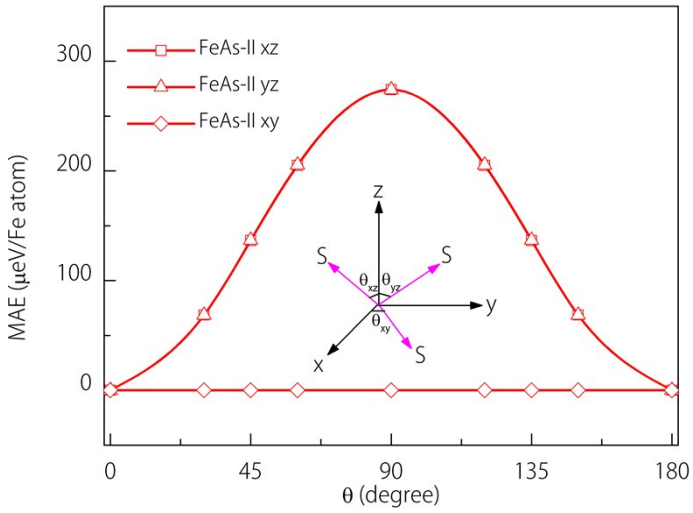

Figure S5 Angular dependence of MAE for single-layer FeAs-II.

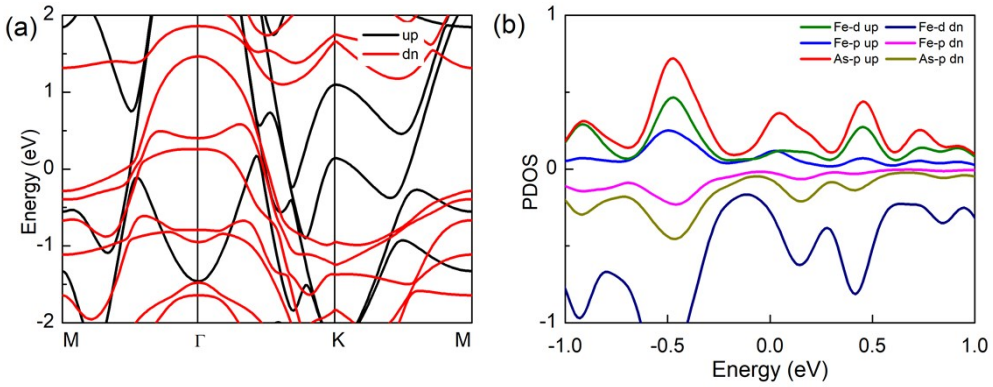

Figure S6 (a) Band structure and (b) PDOS of FeAs-II without SOC.

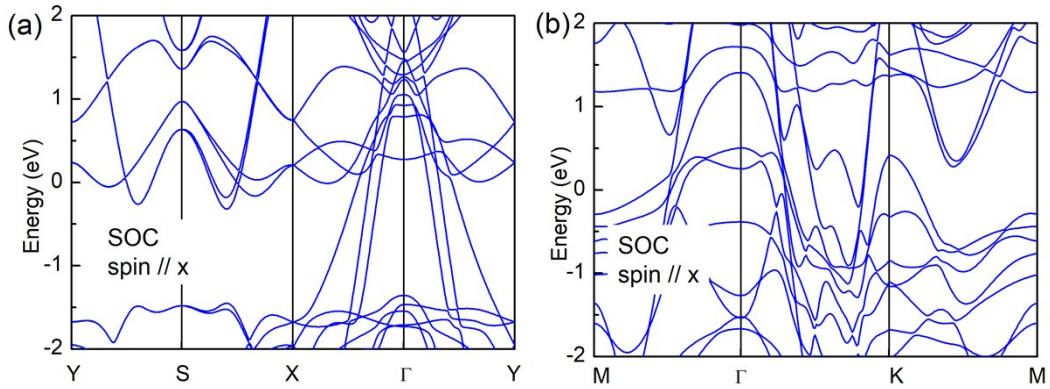

Figure S7 Band structures of (a) FeAs-I and (b) FeAs-II monolayers with SOC.

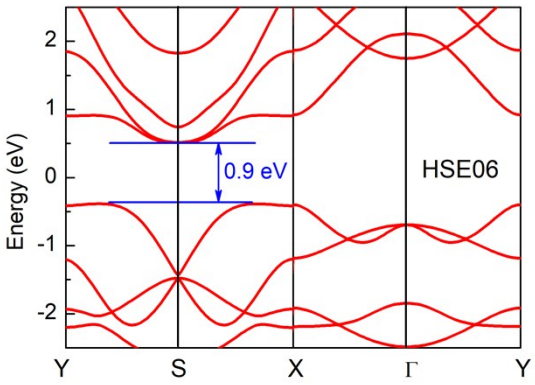

Figure S8 Band structure of FeAs-III on the HSE06 level.

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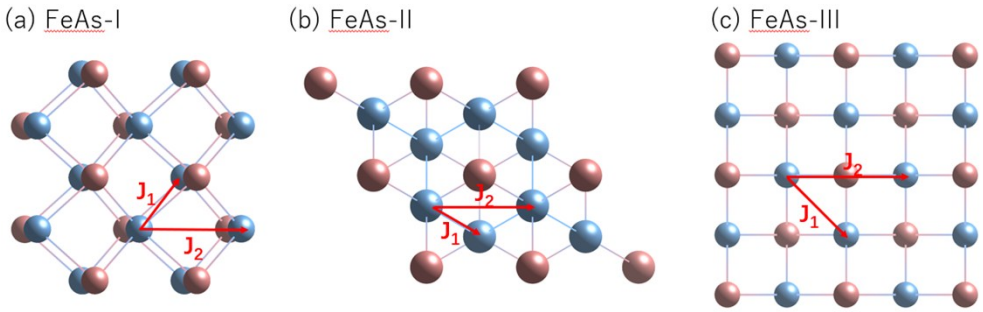

Figure S9 This picture indicates the exchange coupling J_1 and J_2 for the three structures.

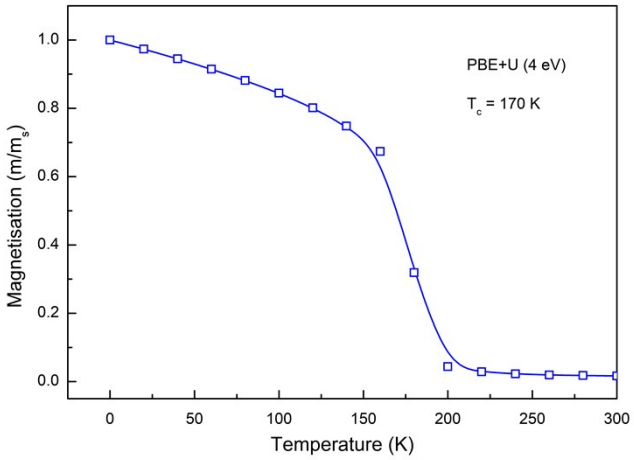

Figure S10 MC result for FeAs-II, showing a critical temperature of about 170 K.

Table S1. Energy difference (per atom, in eV) $\Delta E = E_{FM} - E_{AFM}$, magnetic ground state, space group and formation energy E_f (in eV) for virious FeAs and Fe_2As monolayers.

FeAs	ΔE **(PBE+U)**	**Ground state**	**Space group**	$\mathbf{E_f}$
IV	-0.201	FM	P2/m	-0.162
V	-0.1155	FM	C2/m	-0.155
VI	0.258	AFM	Pmm2	0.160
VII	0.122	AFM	Pmm2	0.191
VIII	0.002	AFM	Pmmm	0.223
IX	0.129	AFM	Pmma	0.240
X	0.0035	AFM	Pma2	0.356
Fe_2As				
I	-0.007	FM	Cmmm	0.036
II	-0.171	FM	P-6m2	0.061
III	-0.166	FM	Pmmm	0.107

Table S2 Calculated elastic constants (N/m) for FeAs-I, -II and -III.

FeAs	$\mathbf{C_{11}}$	$\mathbf{C_{12}}$	$\mathbf{C_{44}}$
I	46.8	43.5	23.8
II	75.9	51.6	0.8
III	39.2	-36.9	17.3

Table S3. Exchange constants (in meV) and critical temperature (T_c/T_N) for FeAs-I and III monolayers, calculated by the PBE+U method with different U values.

U (eV)	**I-J_1**	**I-J_2**	**I-T_c (K)**	**III-J_1**	**III-J_2**	**III-T_N (K)**
3.0	47.78	7.56	605	-38.5	3.9	335
4.0	50.0	8.15	645	-39.9	4.38	350
6.0	57.4	8.19	710	-42.4	4.45	360