Supporting Information

FeSi$_2$: a two-dimensional ferromagnet containing planar hexacoordinate Fe atoms

Ying Zhao $^1$, Qinxi Liu $^1$, Jianpei Xing $^1$, Xue Jiang $^1$*, Jijun Zhao $^{1,2}$

$^1$Key Laboratory of Materials Modification by Laser, Ion and Electron Beams (Ministry of Education), Dalian University of Technology, Dalian 116024, China

$^2$Key Laboratory for Intelligent Nano Materials and Devices of the Ministry of Education, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China

Fig. S1 Evolution of total energy for FeSi$_2$ monolayer at (a) 300 K, (b) 600 K, and (c) 900 K based on AIMD simulations. The insets show snapshots of the FeSi$_2$ monolayer equilibrium structure at different temperatures at the end of the 10-ps AIMD simulation.

* Corresponding Authors. Email: jiangx@dlut.edu.cn (Xue Jiang)
Fig. S2 (a) ELF maps corresponding to slices of FeSi$_2$ monolayer perpendicular to the (001) direction with an isosurface of 0.84 au. (b) Differential charge density slices of FeSi$_2$ monolayer perpendicular to the (001) direction with an isosurface of 0.008 e·Å$^{-3}$. Blue and red colors represent charge accumulation and depletion, respectively.
Fig. S3 Polar diagrams of the (a) Young’s modulus and (b) Poisson’s ratio of FeSi$_2$ monolayer.
Fig. S4 Spin-polarized band structures of FeSi$_2$ monolayer calculated using the HSE06 functional.
Fig. S5 Ferromagnetic (FM) state and seven different antiferromagnetic (AFM) states. Red and green indicate spin up and spin down, respectively. The exchange parameters of $J_i$ ($i = 1–7$) are marked in the FM state.
**Fig. S6** Simulated constant-current STM images of FeSi$_2$ monolayer at a bias voltage of $-1.32$ eV at 2 Å above the Si (110) surface.
Fig. S7 DOS diagram of the freestanding FeSi$_2$ monolayer (a) with and (b) without the Si (110) substrate.
**Table S1.** The energy differences of different AFM states of FeSi$_2$ monolayer relative to the FM state in a 6×4×1 supercell, with unit is eV.

| System | FM | AFM1 | AFM2 | AFM3 | AFM4 | AFM5 | AFM6 | AFM7 |
|--------|----|------|------|------|------|------|------|------|
| FeSi$_2$ | 0  | 2.09 | 1.72 | 0.86 | 1.43 | 1.70 | 1.75 | 2.10 |
Table S2. The calculated exchange parameters $J_i$ ($i = 1–7$) of FeSi$_2$ monolayer at different distances $d_i$.

| $i$ | $d_i$ (Å) | $J_i$ (meV) |
|-----|-----------|-------------|
| 1   | 2.62      | 7.5         |
| 2   | 5.24      | −4          |
| 3   | 5.96      | 2.3         |
| 4   | 6.51      | 3           |
| 5   | 7.85      | 0.85        |
| 6   | 7.93      | −1.8        |
| 7   | 9.86      | 1.13        |