Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

Intrinsic ferromagnetism semiconductivity realized in a new MoS_2 monolayer

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Table S1. The calculated lattice constants *a* (*b*), relative energy E_r, atomic density and band gap E_G.

	H	T	D_2
a, b (Å)	3.188, 3.188	3.206, 3.206	4.790, 3.574
E_r (eV)	0	0.79	1.26
n (MoS_2/ Å^2)	0.98	0.97	0.29
E_G (eV)	1.90	Metallic	0.21

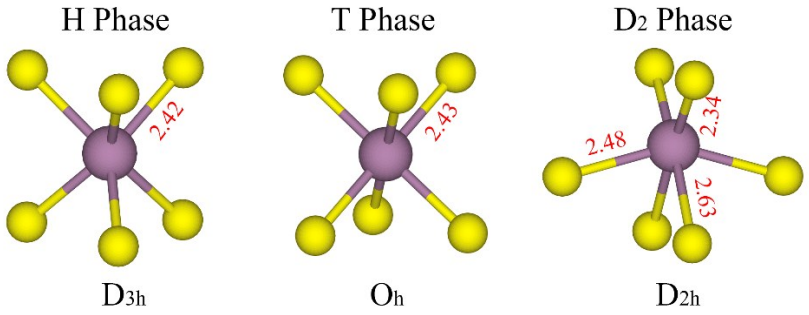

Fig. S1 Different MoS_6 symmetries and the calculated Mo-S bond lengths for H, T and D_2 phases.

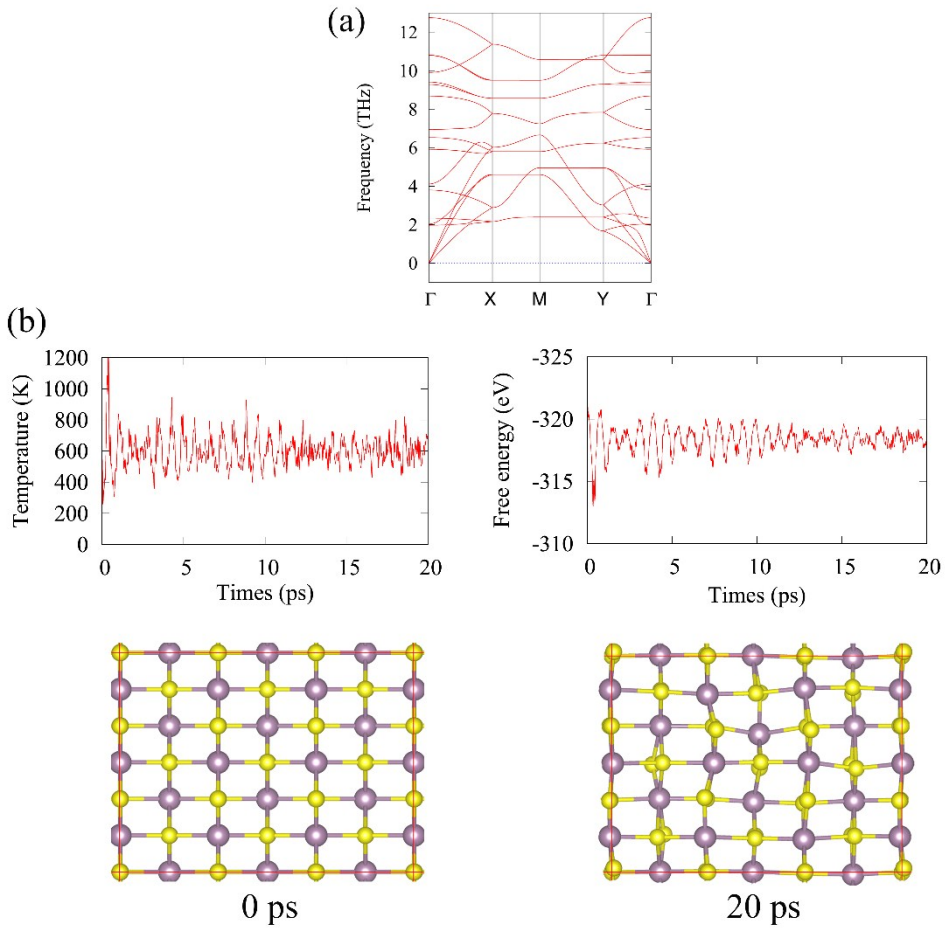

Fig. S2 (a) Phonon spectrum and (b) MD simulation at 600K.

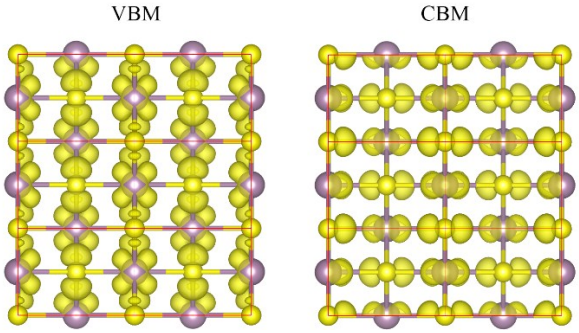

Fig. S3 Real space partial charge density for VBM and CBM.

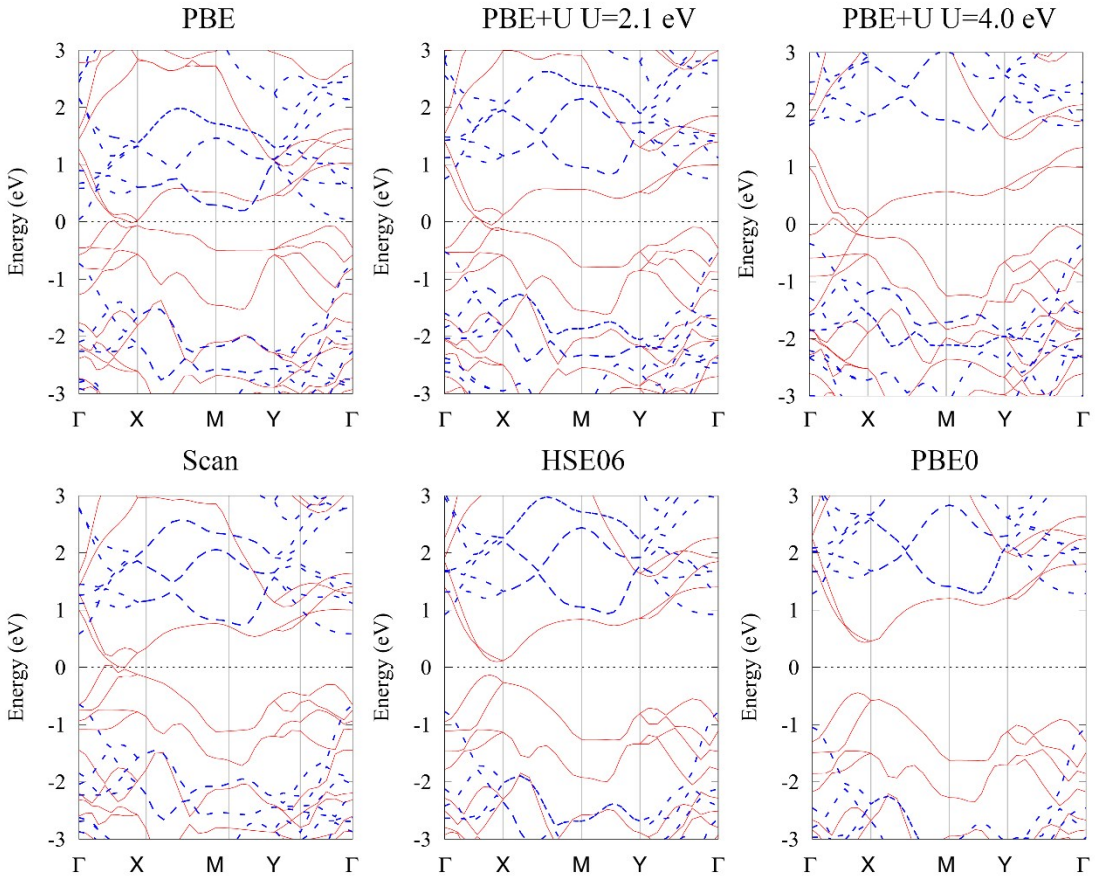

Fig. S4 Electronic band structure by using different functional.

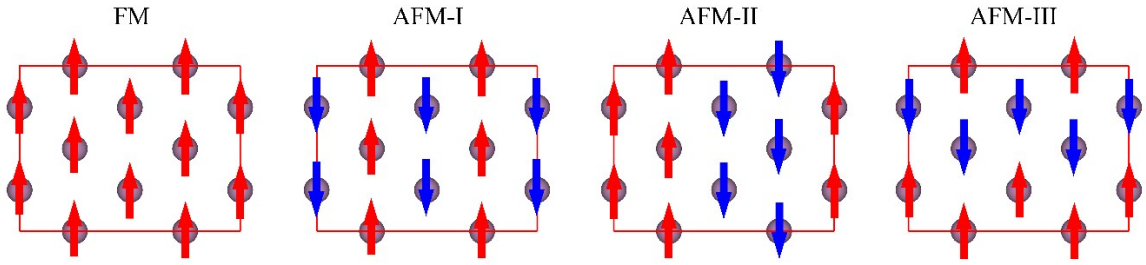

Fig. S5 The 2×2 Mo supercells for ferromagnetic ground state and three antiferromagnetic states to calculate the exchange coupling strength. The relative energy for these magnetic states are 0, 0.207, 0.339 and 1.601 meV, respectively.

Note 1: Computational detail for exchange coupling strength

According to equation 1, the energies arising from spin exchange for FM, AFM-I, AFM-II and AFM-III can be written as,

$$E_{FM} = -8J_1M^2 - 16J_2M^2 - 8J_3M^2, \quad \text{(S1)}$$

$$E_{AFM-I} = -8J_1M^2 + 16J_2M^2 - 8J_3M^2, \quad \text{(S2)}$$

$$E_{AFM-II} = -8J_1M^2 + 8J_3M^2, \quad \text{(S3)}$$

$$E_{AFM-III} = +8J_1M^2 - 8J_3M^2. \quad \text{(S4)}$$

Therefore, J_1, J_2 and J_3 can be obtained,

$$J_2 = \frac{E_{AFM-I} - E_{FM}}{32M^2}, \quad \text{(S5)}$$

$$J_3 = \frac{(E_{AFM-II} - E_{AFM-I}) + 16J_2M^2}{16M^2}, \quad \text{(S6)}$$

$$J_1 = \frac{(E_{AFM-III} - E_{AFM-I}) + 16J_2M^2}{16M^2}. \quad \text{(S7)}$$

Note 2: **Monte Carlo (MC) simulation**

A 24×24 spin grid is chosen for MC simulation. Spin Hamiltonian induced by magnetic coupling takes equation 1. In each MC step, the spin can flip randomly in all directions. If its energy change $\Delta E \leq 0$, we accept this flip. If $\Delta E > 0$, we accept this flip with a probability of $e^{-\Delta E/(k_B T)}$. To obtain the transition temperature, we perform MC simmulations at 0~700 K with a step of 10 K. At each temperature, we first run 10^8 MC steps to achive the equilibrium and following 10^8 MC steps are used for sampling. The Curie temperature is taken as the critical point of the specific heat, defined as $C(T) = (\langle E^2 \rangle - \langle E \rangle^2)/T^2$.

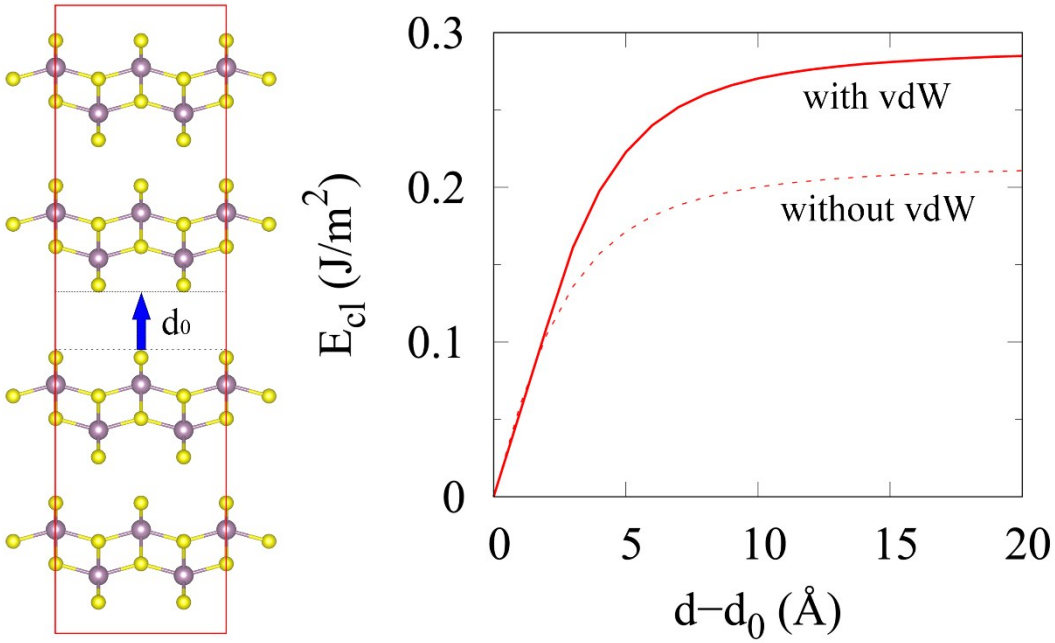

Fig. S6 Cleave scheme and energy for D_2 MoS_2 phase.