Supporting Information

Robust Spin Manipulation in 2D Organometallic Kagome Lattice: A First-principles Study

Peng Wang,[a] Xue Jiang,[b,*] Jun Hu,[c] Biao Wang,[a] Tingwei Zhou,[a] Hongkuan Yuan,[a] Jijun Zhao[b]

[a]*School of Physical Science and Technology, Southwest University, Chongqing 400715, China*

[b]*Key Laboratory of Materials Modification by Laser, Ion and Electron Beams (Dalian University of Technology), Ministry of Education, Dalian 116024, China*

[c]*College of Physics, Optoelectronics and Energy, Soochow University, Suzhou, Jiangsu 215006, China*

[*] E-mail: jiangx@dlut.edu.cn

Supplementary Note

In the work of Lado *et al.* (Ref. 16), they have included the anisotropy exchange term (λ) in the Hamiltonian of the Heisenberg model. For CrI_3 monolayer, this term takes a significant effect on the calculated Curie temperature (T_C). Because we cannot get the correct value of λ under our DFT framework. To clarify the underlying effect of the anisotropic exchange term, we construct the same Hamiltonian as in Ref. 16 and the anisotropy exchange term is included:

$$H = -\sum_{i,j} J_C \vec{S}_i \bullet \vec{S}_j - \sum_i D(S_i^z)^2 - \sum_{i,j} \lambda S_i^z S_j^z$$

We assume λ equals to 0, 0.1D, 0.3D, 0.5D and 0.7D for the $Re_3C_{12}N_{12}H_{12}$ system. Then, Monte Carlo simulations are performed to calculate T_C. The variations of the heat capacity (C_V) with respect to the temperature are shown in **Fig. S3**. It can be seen that the introduction of anisotropy exchange term will enhance the value of T_C, from approximately 330 K (λ=0) to 440 K (λ=0.7D). Hence, we believe the Curie temperature obtained by solving the Heisenberg model in the main text is a lower limit value and the current results are reliable.

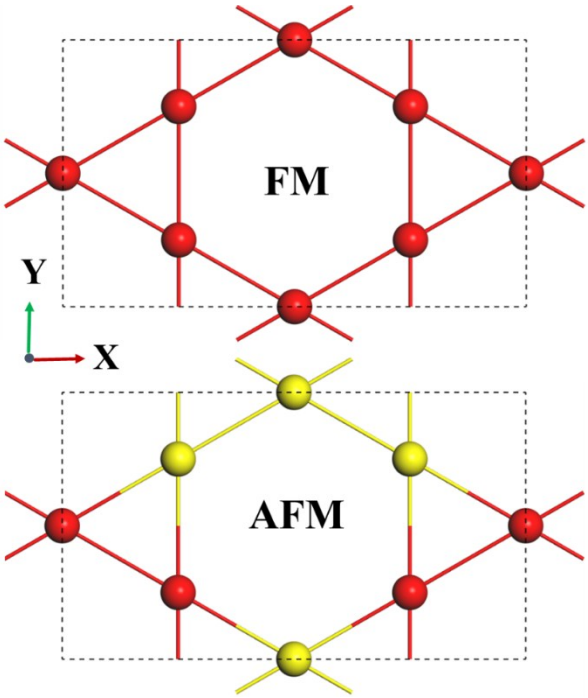

Fig. S1 FM and AFM structures in a $\sqrt{3}\times1$ supercell of the $M_3C_{12}X_{12}$ monolayer. For clarity, only metal atoms are shown and the red and yellow ball refers atoms with magnetic moment along two opposite directions, i.e., spin up and spin down.

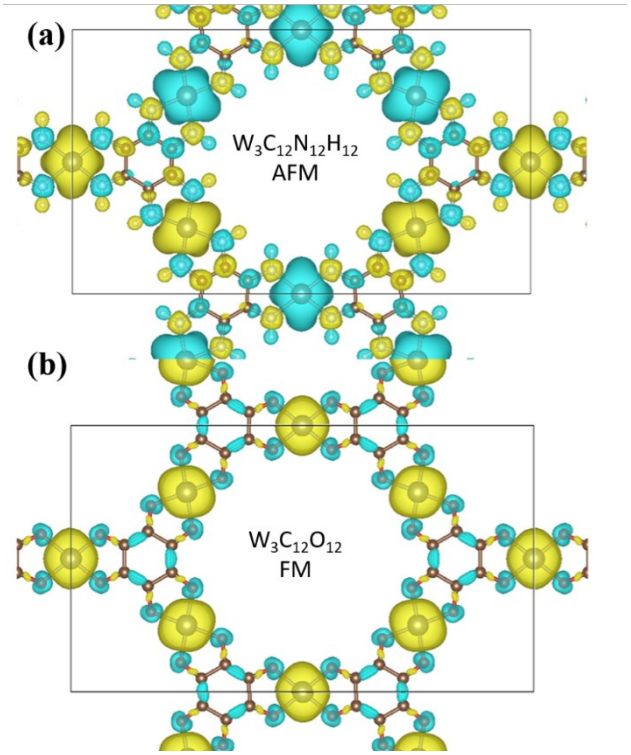

Fig. S2 The spin charge density of $W_3C_{12}N_{12}H_{12}$ and $W_3C_{12}O_{12}$ in a $\sqrt{3} \times 1$ supercell. Here, the spin charge density is defined as $\rho = \rho_{up} - \rho_{down}$. The value of the isosurfaces is 0.0015 e/Å^{-3}. Yellow and cyan surfaces correspond to the isosurfaces of net spin up and spin down density, respectively.

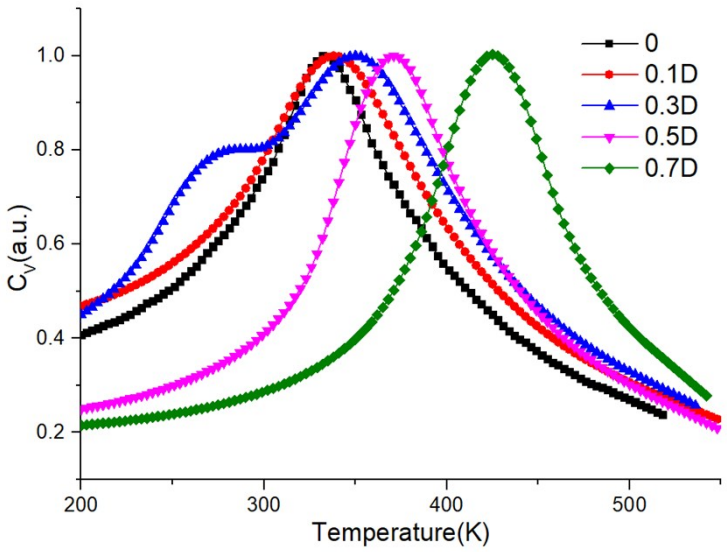

Fig. S3 Variation of the heat capacity (C_V, in arbitrary unit) with respect to the temperature for $Re_3C_{12}N_{12}H_{12}$ with the anisotropic exchange parameter λ equals to 0, 0.1D, 0.3D, 0.5D and 0.7D, respectively.

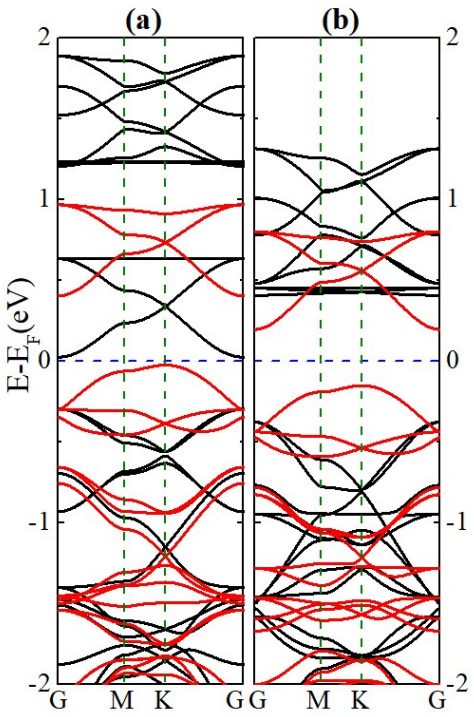

Fig. S4 Spin-polarized band structure of $Re_3C_{12}S_{12}$ (a) and $Os_3C_{12}S_{12}$ (b) monolayers. Red and black lines correspond to spin up and spin down bands, respectively.

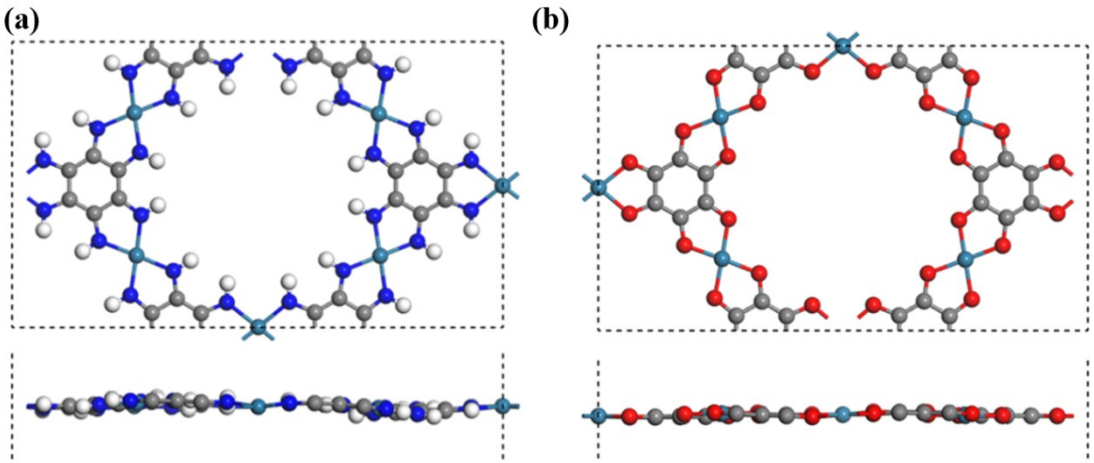

Fig. S5 Top and side views of the snapshot of $Re_3C_{12}N_{12}H_{12}$ and $Re_3C_{12}O_{12}$ in the $\sqrt{3} \times 1$ supercell after 10 ps of MD simulation at 400 K.

Table S1 Reaction heat (ΔH) of $M_3C_{12}X_{12}$ monolayers per formula unit. The unit is in eV.

	Ta	W	Re	Os
$M_3C_{12}S_{12}$	–8.67	–11.17	–11.43	–15.99
$M_3C_{12}N_{12}H_{12}$	–5.75	–7.04	–5.61	–10.59
$M_3C_{12}O_{12}$	–10.69	–8.2	–5.81	–7.71