

Supporting Information

Robust Dirac spin gapless semiconductors in a two-dimensional oxalate based organic HK lattice

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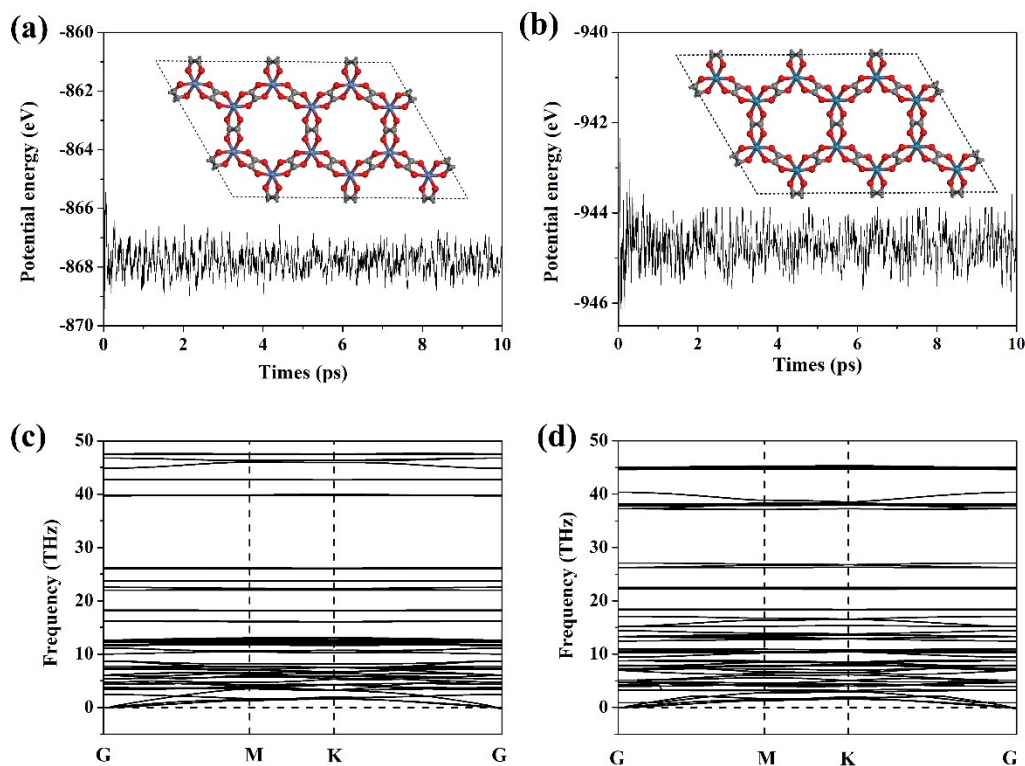


Fig. S1. (a, b) The *ab-initio* molecular dynamic (AIMD) simulation under 300 K with 10 ps timescale, and (c, d) the phonon spectrum of $\text{Ni}_2(\text{C}_2\text{O}_4)_3$ (left) and $\text{Re}_2(\text{C}_2\text{O}_4)_3$ (right), respectively.

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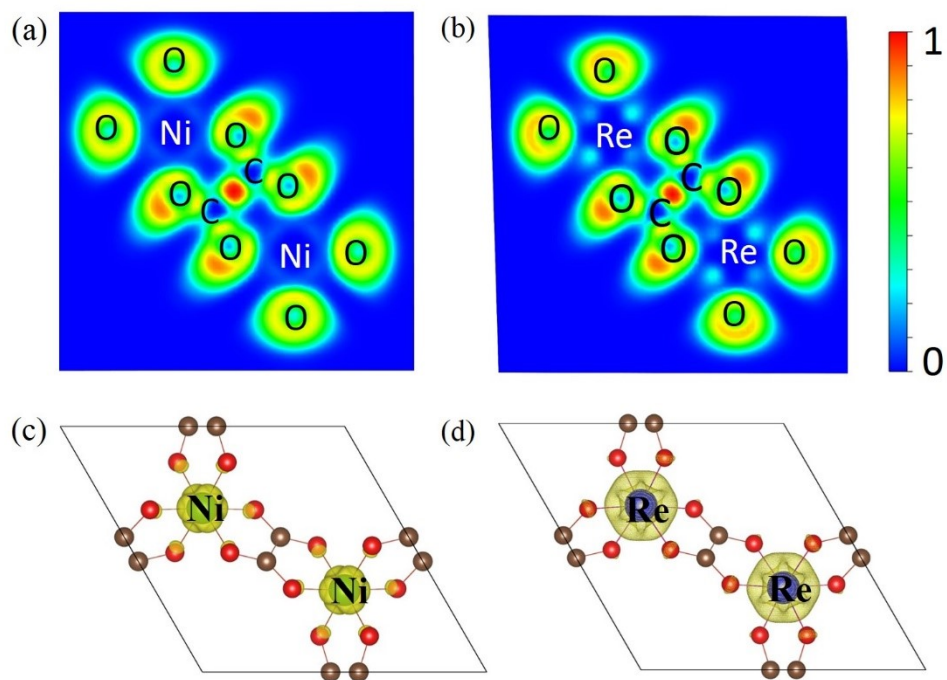


Fig S2. (a, b) The 2D plane ELF of $\text{Ni}_2(\text{C}_2\text{O}_4)_3$ and $\text{Re}_2(\text{C}_2\text{O}_4)_3$ with isosurface $0.4e/\text{Born}^3$. The red and the blue parts denote the electron accumulation and dispersion, respectively. (c, d) The spin charge density of $\text{Ni}_2(\text{C}_2\text{O}_4)_3$ (left) and $\text{Re}_2(\text{C}_2\text{O}_4)_3$ (right). Here the yellow area represents the spin charge localized region.

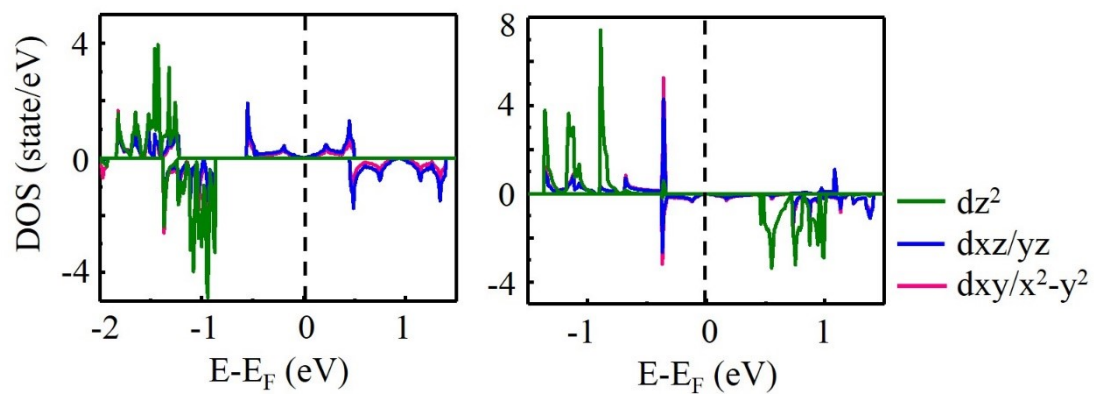


Fig S3. The PDOS of d orbitals of Ni and Re in Ni₂(C₂O₄)₃ (left) and Re₂(C₂O₄)₃ (right), respectively. The Fermi level (E_F) is set to zero.

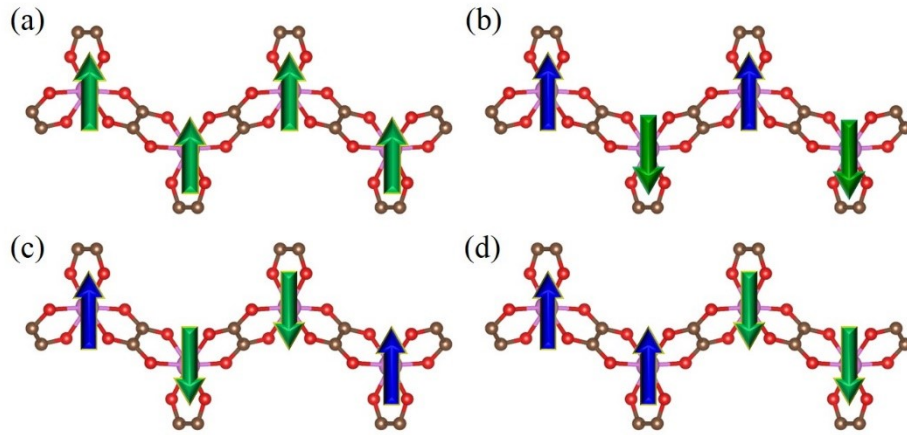


Fig. S4. Four possible magnetic configurations: (a) ferromagnet (FM), (b) Neel antiferromagnet (NAFM), (c) strip antiferromagnet (SAFM) and (d) zigzag antiferromagnet (ZAFM) for $\text{TM}_2(\text{C}_2\text{O}_4)_3$.

Scheme S1. To assess the experimental feasibility of 2D $\text{TM}_2(\text{C}_2\text{O}_4)_3$, the 3×3 supercell of Ag (111) substrate and 4×4 supercell of graphene substrate are screened to grow $\text{Ni}_2(\text{C}_2\text{O}_4)_3$ and $\text{Re}_2(\text{C}_2\text{O}_4)_3$. Their lattice mismatch is about 1.25% and 3.69%, respectively. Based on the first-principle calculations, the distance between $\text{TM}_2(\text{C}_2\text{O}_4)_3$ and substrate are about 2.5 Å and 3.3 Å for $\text{Ni}_2(\text{C}_2\text{O}_4)_3$ and $\text{Re}_2(\text{C}_2\text{O}_4)_3$. Meanwhile, the structures and magnetic properties are well retained.

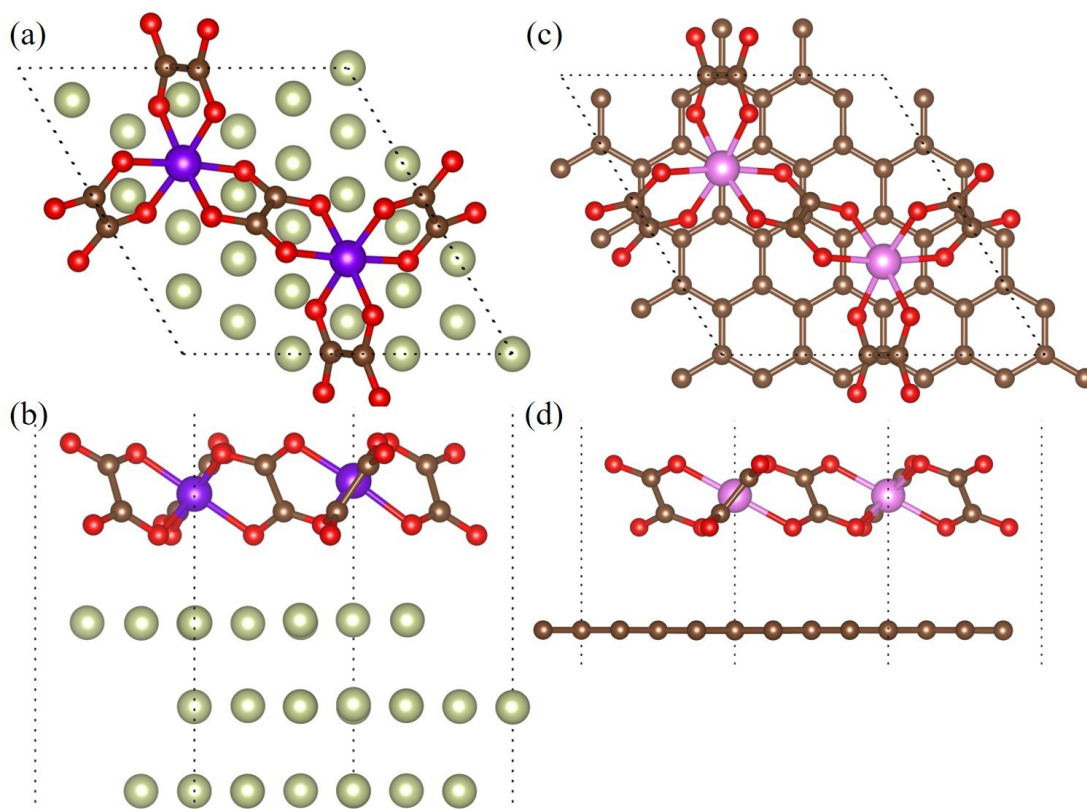


Fig. S5. (a) Top view and (b) side view of $\text{Ni}_2(\text{C}_2\text{O}_4)_3$ on Ag(111) substrate, and (c) top view and (d) side view of $\text{Re}_2(\text{C}_2\text{O}_4)_3$ on graphene monolayer.

Scheme S2. Considering the first and second nearest-neighbor magnetic exchange interactions, the Hamiltonian of a classical Heisenberg model can be written as Eq. (S1)

$$H = - \sum_{mn} J_1 M_m M_n - \sum_{kl} J_2 M_k M_l - \sum_{ij} J_3 M_i M_j \quad (\text{S1})$$

where J_1 , J_2 and J_3 are the first, second and third nearest-neighbor exchange parameters, respectively; the magnetic moments for per TM in 2D $\text{Ni}_2(\text{C}_2\text{O}_4)_3$ and $\text{Re}_2(\text{C}_2\text{O}_4)_3$ are $0.83 \mu_B$ and $1.82 \mu_B$. Based on the calculated total energies of different magnetic configurations, three different magnetic configurations are chosen.

The exchange parameters J_1 , J_2 and J_3 for $\text{Ni}_2(\text{C}_2\text{O}_4)_3$ can be derived by following equations

$$E(\text{FM}) = -(6J_1 + 12J_2 + 6J_3) M^2$$

$$E(\text{AFM}_1) = -(2J_1 - 4J_2 - 6J_3) M^2$$

$$E(\text{AFM}_2) = -(-6J_1 + 12J_2 - 6J_3) M^2$$

$$E(\text{AFM}_3) = -(-2J_1 + 12J_2 - 6J_3) M^2$$

$$J_1 = \frac{-192E_1 + 224E_2 + 192E_3}{3456M^2}$$

$$J_2 = \frac{96E_1 - 112E_2 + 120E_3}{3456M^2}$$

$$J_3 = \frac{192E_1 + 64E_2 - 192E_3}{3456M^2}$$

Here, $E_1 = E(\text{AFM}_1) - E(\text{FM})$, $E_2 = E(\text{AFM}_2) - E(\text{FM})$ and $E_3 = E(\text{AFM}_3) - E(\text{FM})$, $M = 0.83 \mu_B$.

For $\text{Re}_2(\text{C}_2\text{O}_4)_3$, it only has one kind of AFM configuration (Néel AFM as shown in Fig. S4(b)) and other two configurations are ferrimagnetic. Only considering one AFM configuration, the expression of $E(\text{FM})$ and $E(\text{AFM})$ can be written as:

$$E(\text{FM}) = -6J_1 M^2$$

$$E(\text{AFM}) = -2J_1 M^2$$

and

$$J = \frac{E}{4M^2}$$

Here, $E = E(\text{AFM}) - E(\text{FM})$, $M = 1.82 \mu_{\text{B}}$.

The calculated J_1 for $\text{Re}_2(\text{C}_2\text{O}_4)_3$ is 3.73 meV. The J_1, J_2 and J_3 for $\text{Ni}_2(\text{C}_2\text{O}_4)_3$ are 69.75 meV, -1.38 meV, and -6.28 meV, respectively, which show that the second and third neighbor coupling are very weak and can be neglected, and first neighbor magnetic exchange couplings are FM coupling.