Supporting Information for Effect of metal-ligand interactions on magnetic characteristics of two-dimensional Kagome structured perthiolated coronene (PTC) metal-organic frameworks (MOFs)

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In this study, we investigated the bonding characteristics of the system by using Bader charge transfer analysis. Subsequently, after examining the energy band projection diagrams of PTC-Mn, we extended our analysis to calculate the energy band structures of PTC-Fe and PTC-Co, which are quite different from that of PTC-Mn. Notably, the energy band structures of PTC-Fe and PTC-Co exhibit a distinct flat-band structure with Van Hove singularities, indicative of the Kagome lattice. This observation confirms the potential simplification of these systems as Kagome structures with only connecting point metal atoms for investigating their magnetic properties. This also implies possible topological electro-magnetic properties of these two systems. It is rather unfortunate that the

PTC-Mn system, which has the highest magnetic transition temperature, does not exhibit such electronic structure.

When considering non-collinear magnetic states, we find that none of the three configurations shown in Fig. S4(a-c) represents the ground state of the system. However, for PTC-Mn, the energy difference between the two configurations, AFM-S1 and AFM-S3, while greater than that of the conventional FM configuration, remains extremely small. Similarly, in the PTC-Fe system, the energy difference between AFM-S2 and the ground state FM is also modest. These observations suggest that both systems may exhibit more complex spin textures under certain conditions.

Table S1. Relative energies of PTC-TM (Mn, Fe, Co) in different non-collinear magnetic states with respect to FM states, ($E-E_{FM}$)

System	FM (meV)	AFM-S1 (meV)	AFM-S2 (meV)	AFM-S3 (meV)
PTC-Mn	0	0.2	952.1	0.2
PTC-Fe	0	51.6	0.1	51.6
PTC-Co	0	384.8	384.8	227.8

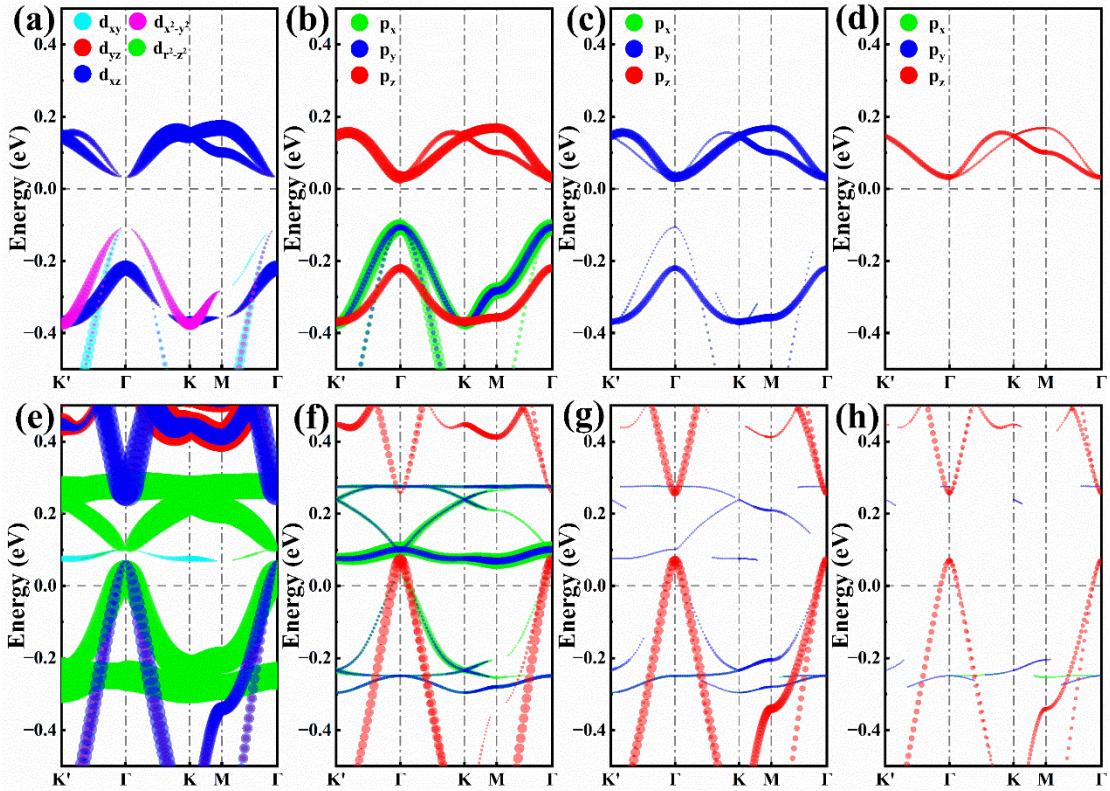

Fig. S1. Orbital-resolved band structure of PTC-Fe monolayer. The *d*-orbital projections of the Mn atom with minority and majority electrons are shown in (a) and (e), respectively. The *p*-orbital projections of the S atom in minority and majority channels are illustrated in (b) and (f). Projections of the *p*-orbitals of the C-18 atom in minority and majority channels are displayed in (c) and (g), while (d) and (h) depict the *p*-orbital projections of the C-6 atom in minority and majority channels.

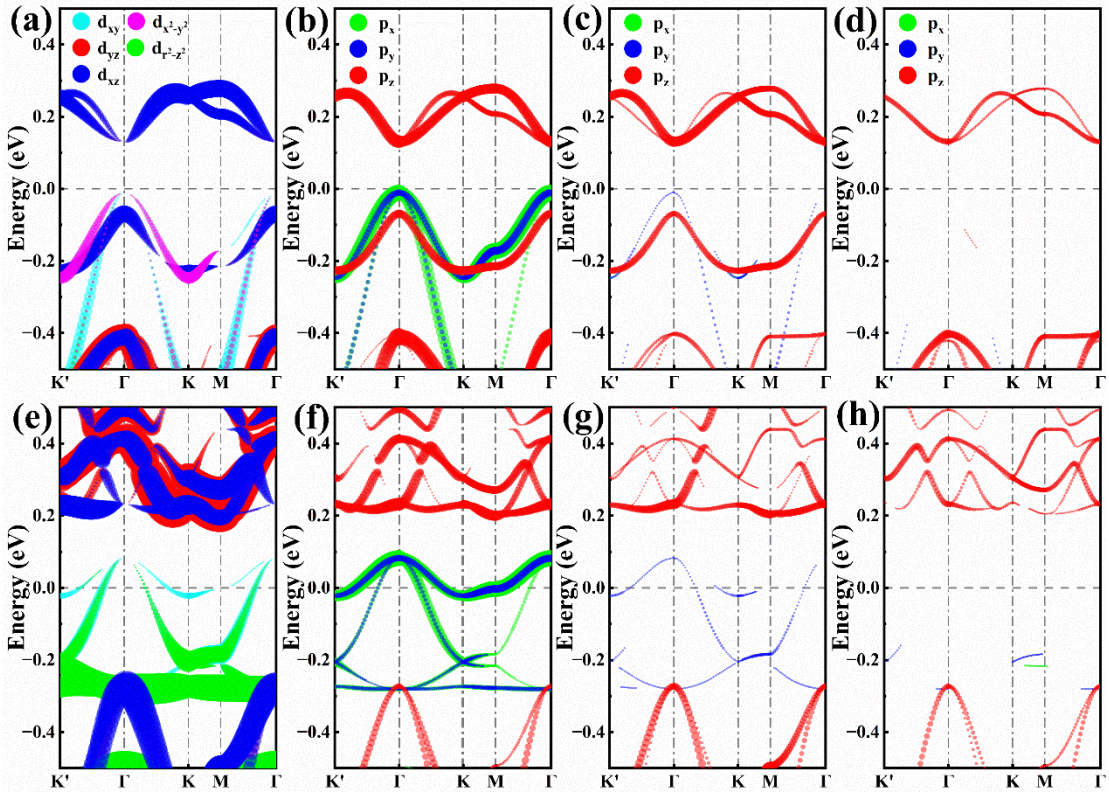

Fig. S2. Orbital-resolved band structure of PTC-Co monolayer. The *d*-orbital projections of the Mn atom with minority and majority electrons are shown in (a) and (e), respectively. The *p*-orbital projections of the S atom in minority and majority channels are illustrated in (b) and (f). Projections of the *p*-orbitals of the C-18 atom in minority and majority channels are displayed in (c) and (g), while (d) and (h) depict the *p*-orbital projections of the C-6 atom in minority and majority channels.

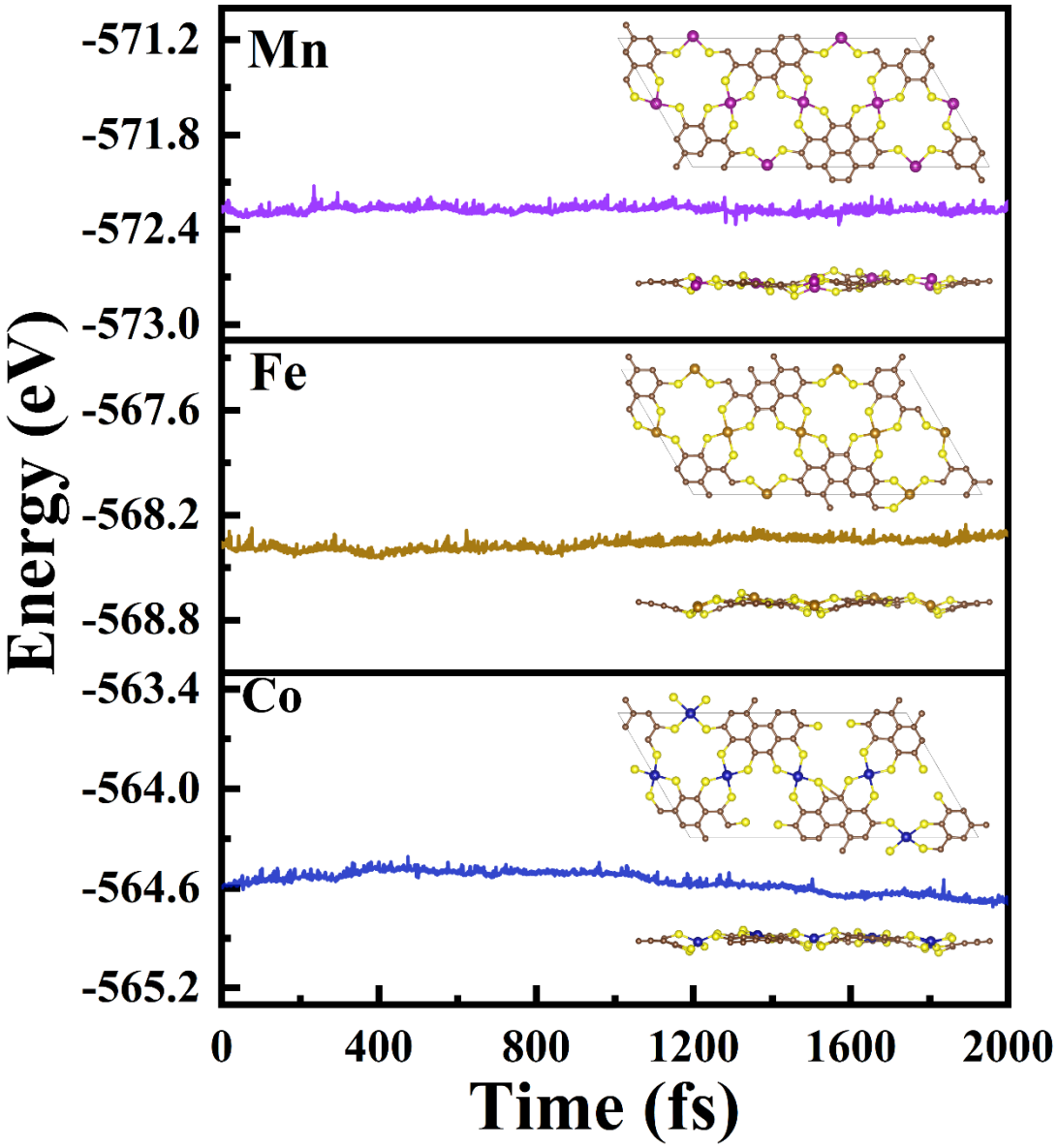

Fig. S3. Total energy changes of PTC-Mn, Fe, Co at 300 K within 2 ps according to AIMD simulation. The insets are the top and side views of the corresponding structure after molecular dynamics simulation.

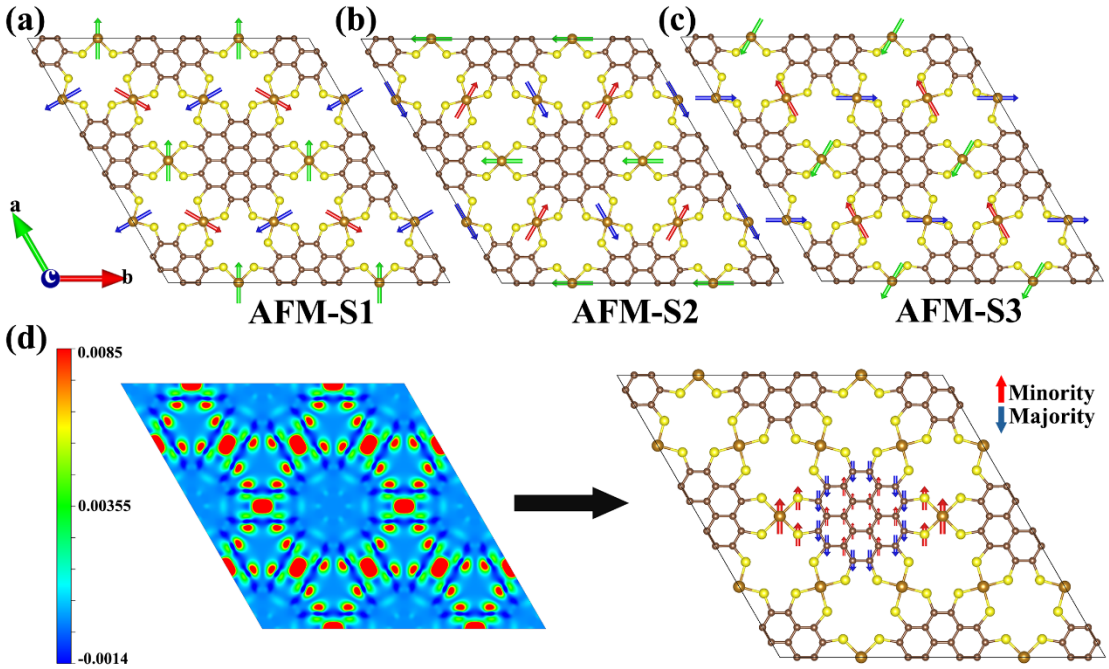

Fig. S4. The specific arrangements of magnetic moments for the three non-collinear magnetic configurations are illustrated in panels (a) to (c). Panel (d) depicts the spin density of PTC-Fe along with a schematic representation of the oscillating spin evolution between a pair of metal atoms.