Supporting Information for

Two-Dimensional Metal-Organic Half-Metallic Antiferromagnet: CoFePz

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Since the Fe^{2+} and Co^{2+} ions are in the similar chemical environments (binding with four N atoms), it is reasonable to assign the same U/J value to Fe^{2+} and Co^{2+} ions. Therefore, we test different U values (J = 1eV) with the same value for Fe^{2+} and Co^{2+} ions and the results are showed in Table S1 and Figure S1.It can be confirmed that the antiferromagnetic ground states robust against details of the calculations. The spin-polarized electronic band structure near the Fermi level is subject to the chosen value of U.
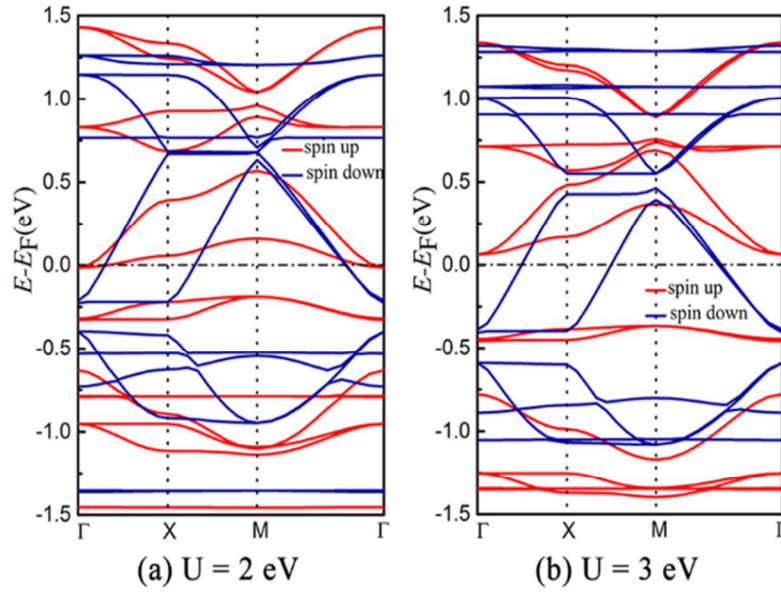

(a) U = 2 eV (b) U = 3 eV

Figure S1. Spin-polarized electronic band structures of 2D CoFePz framework in AFM state with different values of U from the DFT + U method. Γ (0, 0, 0), X (0, 1/2, 0) and M (1/2, 1/2, 0) are highly symmetric points in reciprocal space.

Table S1. Calculated lattice constants and exchange energies E_{ex} with different values of U from the DFT + U method.

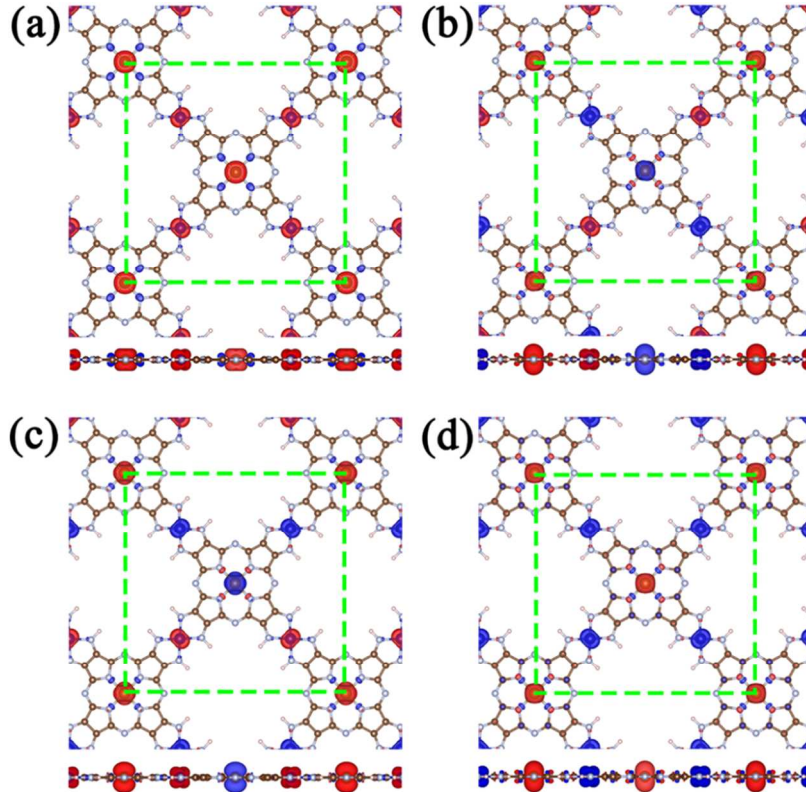| U (eV)	2	3	4
lattice constant (Å)	13.430	13.437	13.439
E_{ex} (= E_{FM} - E_{AFM}) (meV)	35	73	143

Figure S2. Top and side views of the spin density isosurfaces in a $\sqrt{2} \times \sqrt{2} \times 1$ supercell at absolute spin-density value $|\rho_\uparrow - \rho_\downarrow| = 0.04$ electrons per $Å^3$ for four magnetic coupling configurations. (a) illustrates ferromagnetic (FM) configuration; (b), (c), (d) are three different antiferromagnetic (AFM) configurations and are labeled as AFM_{rod}, AFM_{zigzag}, AFM respectively. Red and blue isosurfaces correspond to positive and negative spin density, respectively. The $\sqrt{2} \times \sqrt{2} \times 1$ supercell is denoted by the dashed rectangle.

Table S2. Calculated total energies E_{tot} relative to the energy of AFM configuration of four magnetic coupling configurations (FM, AFM_{rod}, AFM_{zigzag} and AFM) in a $\sqrt{2} \times \sqrt{2} \times 1$ supercell.

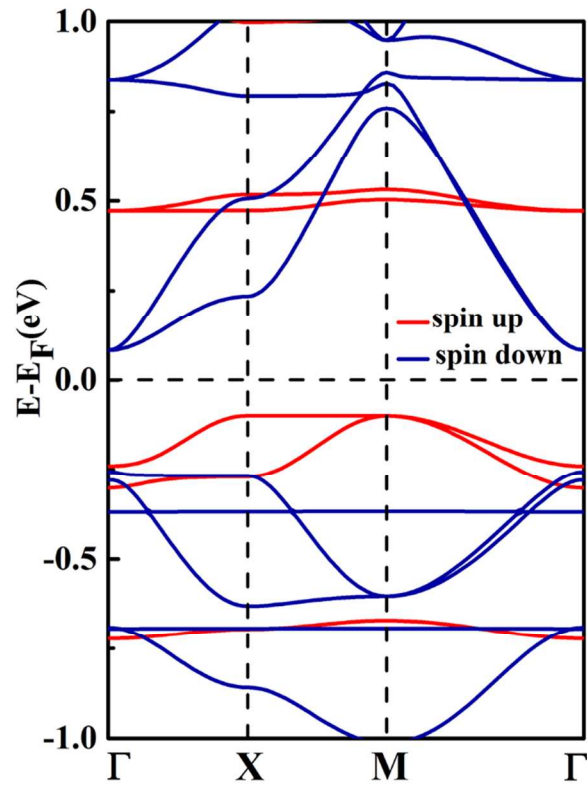|	FM	AFM_{rod}	AFM_{zigzag}	AFM
E_{tot}(meV)	293.1	181.7	168.4	0

Figure S3. Band structure of the 2D CoFePz at the FM state near the Fermi level. Γ (0,0,0), X $(0, 1/2, 0)$ and M $(1/2, 1/2, 0)$ represent the highly symmetric points in reciprocal space.
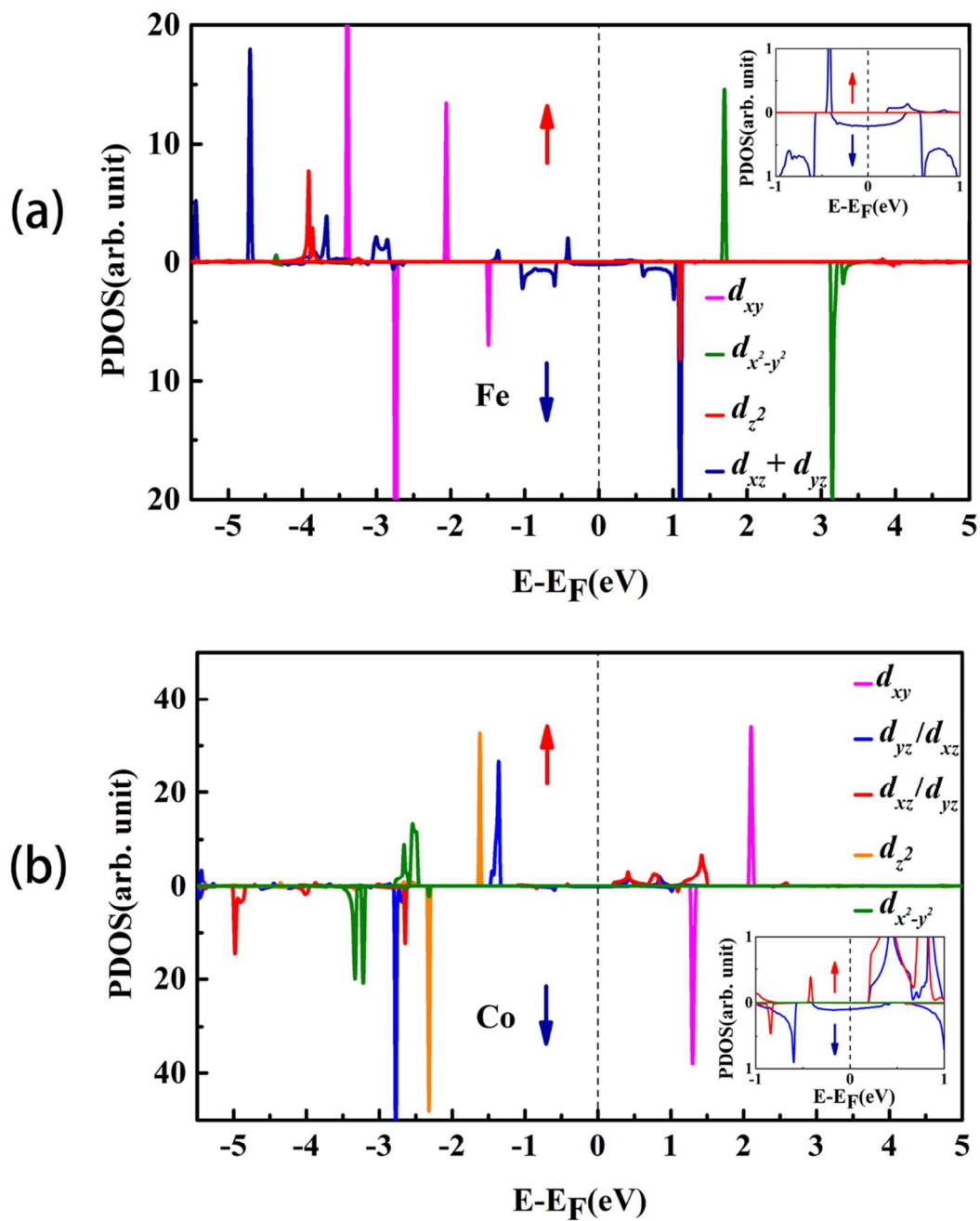
Figure S4. The partial electron density of states (PDOS) projected onto the 3d orbitals of Fe, Co atoms. The energy at the Fermi level was set to zero. "Up" and "down" arrows denote the spin-up and down polarization, respectively. The enlarged views of the PDOS near the Fermi level (from -1eV to 1eV) were plotted as the insets of this figure.