Supporting Information for "Multiferroic Metallic Monolayer $Cu(CrSe_2)_2$"

Ke Yang,[1,2,*] Yuxuan Zhou,[2,*] Yaozhenghang Ma,[2] and Hua Wu[2,3,4,†]

[1] *College of Science, University of Shanghai for Science and Technology, Shanghai 200093, China*
[2] *Laboratory for Computational Physical Sciences (MOE), State Key Laboratory of Surface Physics, and Department of Physics, Fudan University, Shanghai 200433, China*
[3] *Shanghai Qi Zhi Institute, Shanghai 200232, China*
[4] *Shanghai Branch, Hefei National Laboratory, Shanghai 201315, China*

Table S1. The calculated magnetic anisotropy energy MAE (meV/f.u.) for $Cu(CrSe_2)_2$ using different k-meshes and cut-off (eV) energies.

k-mesh	E_{cut}	010(b)	001(c)
15×15×1	500	0	1.084
15×15×1	550	0	1.092
17×17×1	500	0	1.124

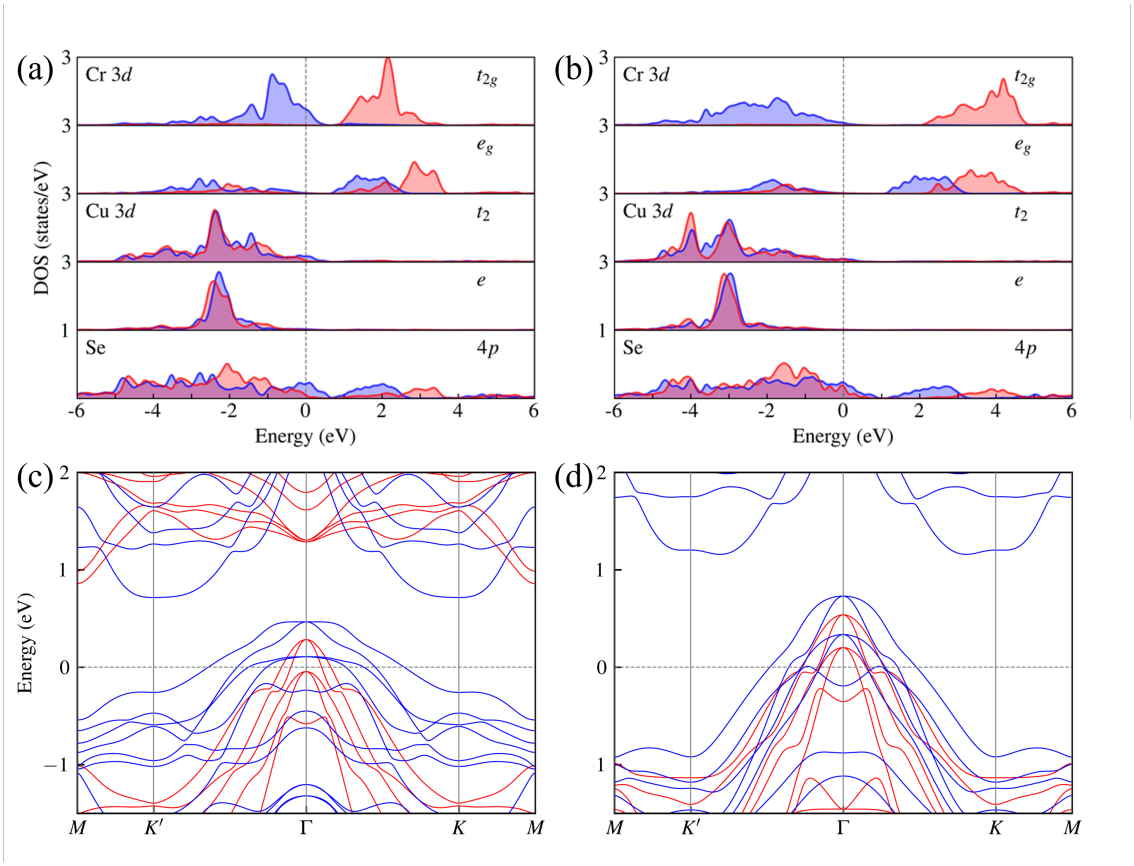

Fig. S1. The Cr 3d, Cu 3d, Se 4p, and total density of states (DOS) are shown for (a) GGA and (b) GGA+U calculations. The corresponding band structures are presented in (c) GGA and (d) GGA+U. The blue (red) curves stand for the up (down) spin channel. The Fermi level is set at zero energy.

Table S2. The magnetic anisotropy energy (MAE, meV/f.u.), exchange interaction parameter (J, meV), and Curie temperature (T_C, K) by GGA + SOC and GGA + SOC + U for different U (eV) values.

		MAE	J_{top}	J_{bottom}	J_{inter}	T_C
GGA		−2.54	−9.46	−10.36	−1.58	360
U_{Cu}	U_{Cr}	MAE	J_{top}	J_{bottom}	J_{inter}	T_C
6	4	−1.08	−5.70	−6.55	−0.18	190
6	5	−1.40	−5.89	−7.09	−0.03	178
7	4	−0.95	−5.73	−6.53	−0.19	190
7	5	−1.30	−5.91	−7.04	−0.04	178

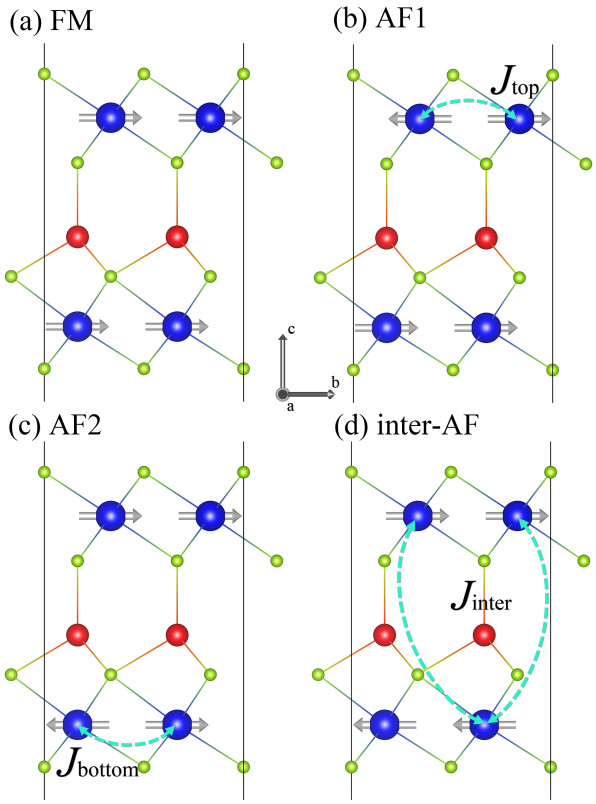

Fig. S2. The four magnetic structures of $Cu(CrSe_2)_2$ marked with three exchange parameters.

Table S3. Relative total energies ΔE (meV/f.u.) and local spin moments (μ_B) for the $Cu(CrSe_2)_2$ monolayer. The derived three exchange parameters are J_{top} =–5.70 meV and J_{bottom} =–6.55 meV, and the interlayer J_{inter} =–0.18 meV. The T_C of 190 K is estimated by MC simulation.

state	Energy	$M_{Cr_{top}}$	$M_{Cr_{bottom}}$	M_{Cu}
FM	0	3.04	3.12	−0.05
AF1	60.17	±2.93	3.12	−0.03
AF2	52.47	3.05	±2.98	−0.01
inter-AF	2.38	3.05	−3.12	0.01

Table S4. The relative total energy (meV/f.u.) for the different states is calculated using the four-state method, employing a 3×4 supercell to calculate the Dzyaloshinskii-Moriya (DM) parameters (meV) of the top layer $CrSe_2$. This involves a pair of nearest Cr ions along the b-axis in the top layer.

τ_1	τ_2	$(0,\tau_1 S,0);\ (0,0,\tau_2 S)$	$(0,0,\tau_1 S);\ (\tau_2 S,0,0)$	$(\tau_1 S,0,0);\ (0,\tau_2 S,0)$
+	+	0	0	0
−	+	0.247	0.457	124.801
+	−	124.045	−0.169	0.061
−	−	125.836	−0.714	124.938
		$D_x = 0.09$	$D_y = -0.08$	$D_z = 0.09$

Table S5. The relative total energy (meV/f.u.) for the different states is calculated using the four-state method, employing a 3×4 supercell to calculate the DM parameters (meV) of the bottom layer $CrSe_2$. This involves a pair of nearest Cr ions along the b-axis in the bottom layer.

τ_1	τ_2	$(0,\tau_1 S,0);\ (0,0,\tau_2 S)$	$(0,0,\tau_1 S);\ (\tau_2 S,0,0)$	$(\tau_1 S,0,0);\ (0,\tau_2 S,0)$
+	+	0	0	0
−	+	0.487	−1.012	121.444
+	−	121.441	0.563	0.319
−	−	122.763	−1.167	122.580
		$D_x = 0.17$	$D_y = -0.11$	$D_z = 0$

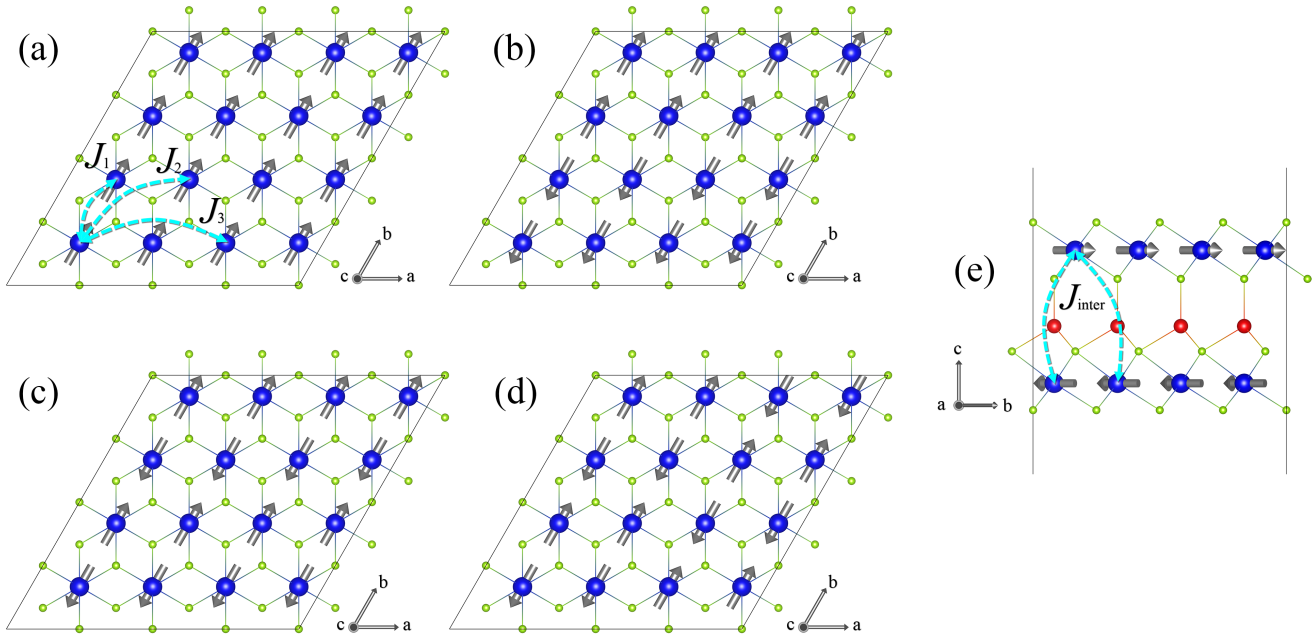

Fig. S3. The five magnetic configurations of $Cu(CrSe_2)_2$ are utilized to estimate the second-nearest neighbor J_2, and the third-nearest neighbor J_3 within the $CrSe_2$ layer.

$$
\begin{aligned}
E_{\mathrm{FM,FM}} &= [3(J_{\mathrm{top1}} + J_{\mathrm{top2}} + J_{\mathrm{top3}}) + 3(J_{\mathrm{bottom1}} + J_{\mathrm{bottom2}} + J_{\mathrm{bottom3}}) + 3J_{\mathrm{inter}}]\, S^2 \\
E_{\mathrm{AF1,FM}} &= [(J_{\mathrm{top1}} - J_{\mathrm{top2}} - J_{\mathrm{top3}}) + 3(J_{\mathrm{bottom1}} + J_{\mathrm{bottom2}} + J_{\mathrm{bottom3}})]\, S^2 \\
E_{\mathrm{AF2,FM}} &= [(-J_{\mathrm{top1}} - J_{\mathrm{top2}} + J_{\mathrm{top3}}) + 3(J_{\mathrm{bottom1}} + J_{\mathrm{bottom2}} + J_{\mathrm{bottom3}})]\, S^2 \\
E_{\mathrm{AF3,FM}} &= [(-J_{\mathrm{top1}} + J_{\mathrm{top2}} - J_{\mathrm{top3}}) + 3(J_{\mathrm{bottom1}} + J_{\mathrm{bottom2}} + J_{\mathrm{bottom3}})]\, S^2 \\
E_{\mathrm{FM,FM'}} &= [3(J_{\mathrm{top1}} + J_{\mathrm{top2}} + J_{\mathrm{top3}}) + 3(J_{\mathrm{bottom1}} + J_{\mathrm{bottom2}} + J_{\mathrm{bottom3}}) - 3J_{\mathrm{inter}}]\, S^2 \\
E_{\mathrm{FM,AF1}} &= [3(J_{\mathrm{top1}} + J_{\mathrm{top2}} + J_{\mathrm{top3}}) + (J_{\mathrm{bottom1}} - J_{\mathrm{bottom2}} - J_{\mathrm{bottom3}})]\, S^2 \\
E_{\mathrm{FM,AF2}} &= [3(J_{\mathrm{top1}} + J_{\mathrm{top2}} + J_{\mathrm{top3}}) + (-J_{\mathrm{bottom1}} - J_{\mathrm{bottom2}} + J_{\mathrm{bottom3}})]\, S^2 \\
E_{\mathrm{FM,AF3}} &= [3(J_{\mathrm{top1}} + J_{\mathrm{top2}} + J_{\mathrm{top3}}) + (-J_{\mathrm{bottom1}} + J_{\mathrm{bottom2}} - J_{\mathrm{bottom3}})]\, S^2
\end{aligned}
\tag{S1}
$$

Table S6. The derived exchange parameters (meV) up to the third-nearest neighbors within the $CrSe_2$ layer. The T_C of 185 K is estimated by MC simulation.

type	J_1	J_2	J_3
top layer	−4.59	−0.73	0.15
bottom layer	−6.23	−0.50	0.56
inter-layer	−0.26	/	/

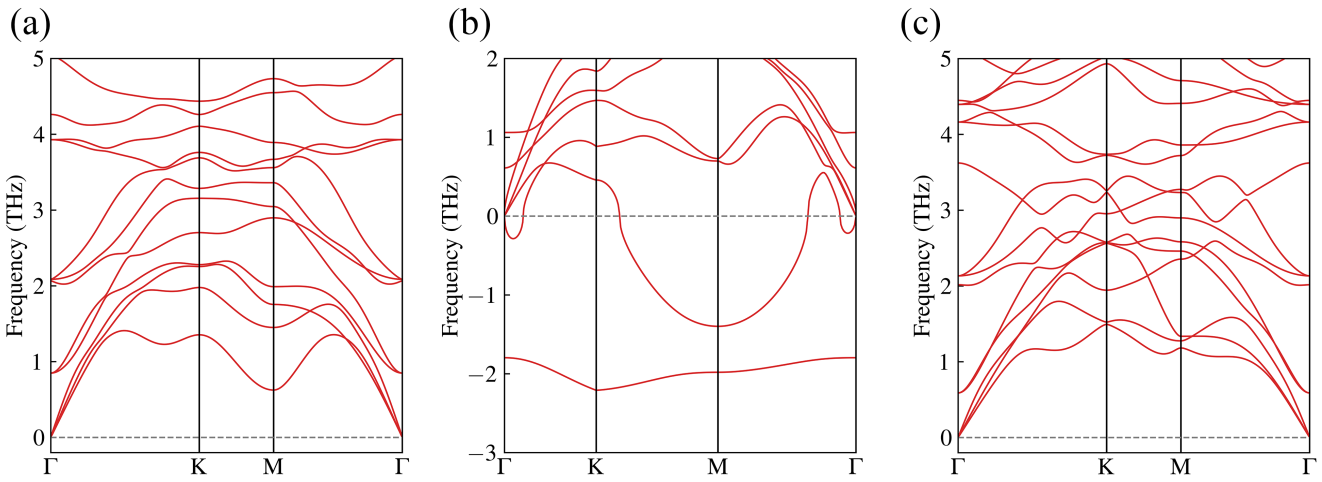

Fig. S4. The phonon spectrum of (a) FE (±P) (b) PE', and (c) PE states, see also Fig. 3 in the main text.

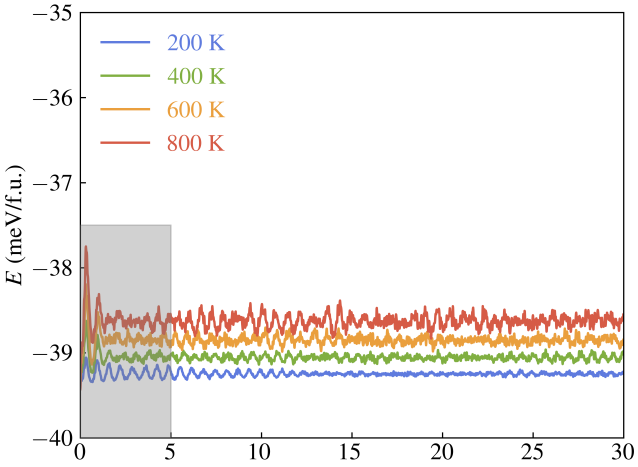

Fig. S5. The total-energy evolution in the ab initio molecular dynamics simulation.

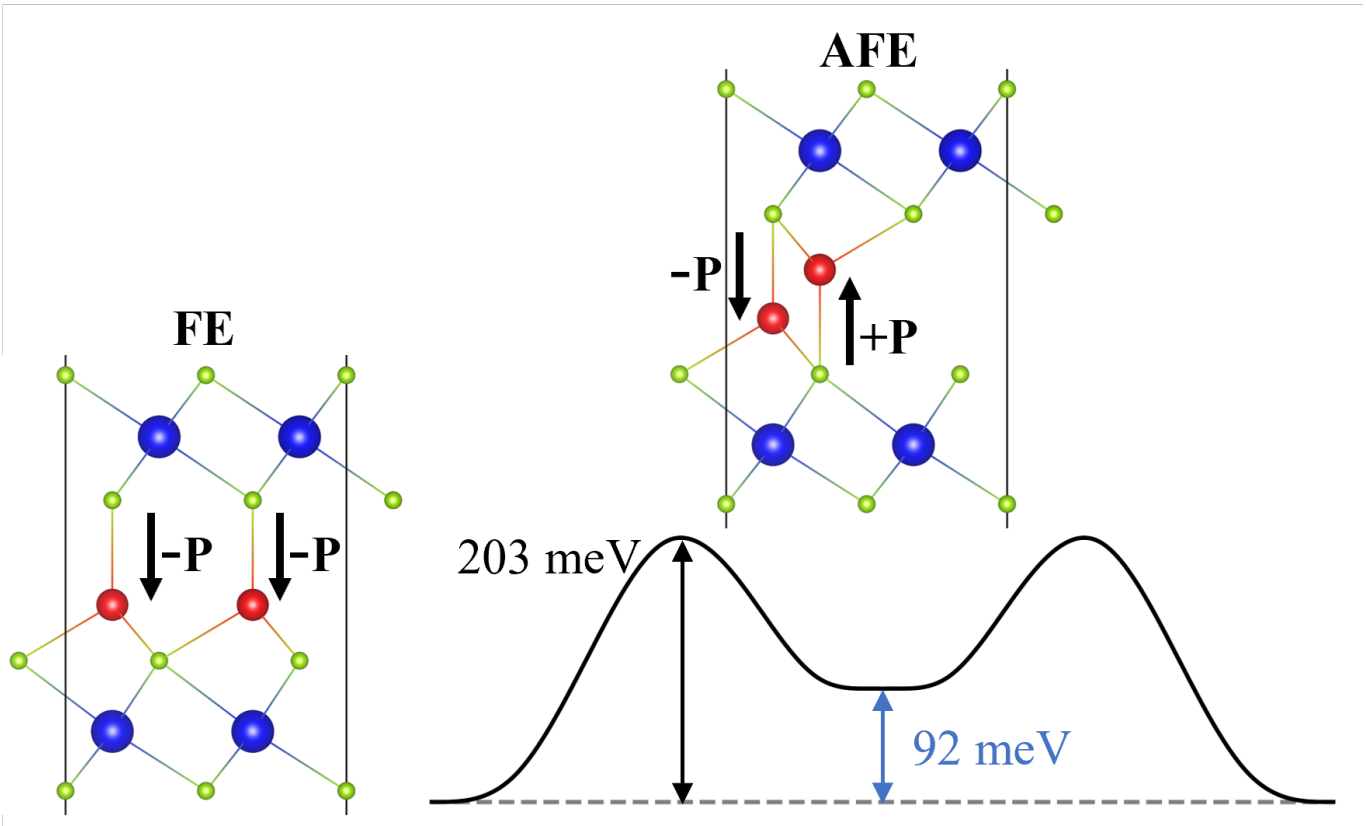

Fig. S6. The FE and anti-FE structure. The anti-FE one is less stable by 92 meV/f.u.