

## Supplementary Material

# Electrical control of topological spin textures in two-dimensional multiferroics

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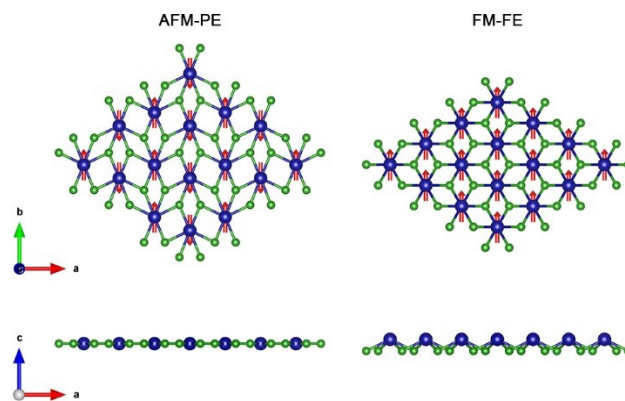
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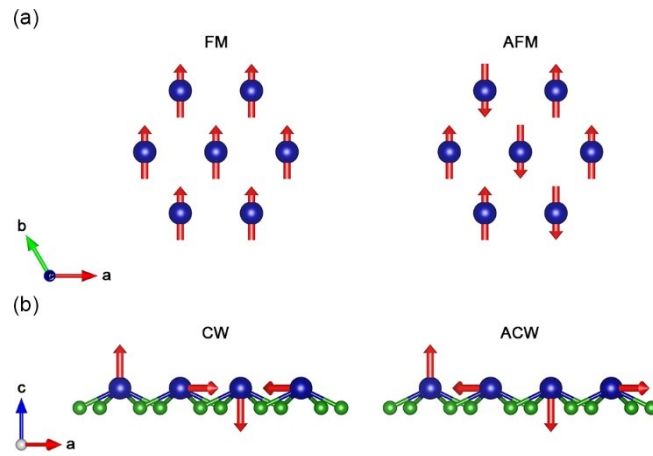
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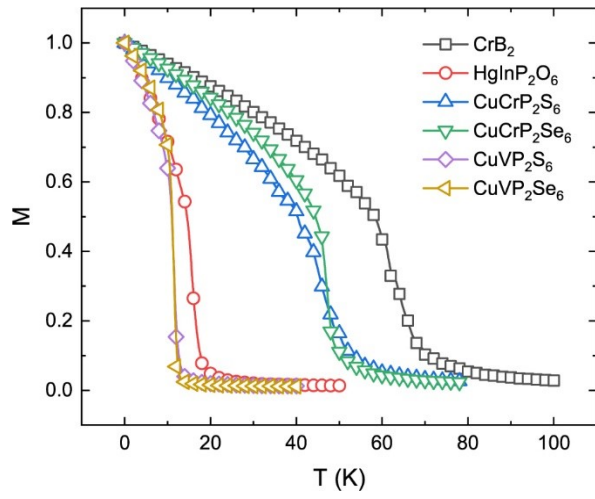
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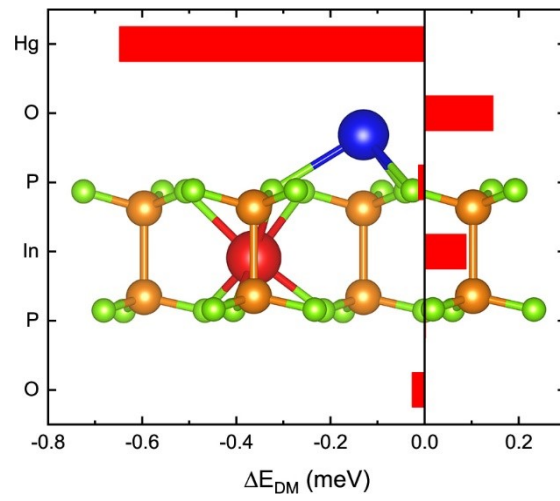
**Fig. S1.** The schematic structures of paraelectric (PE) phase CrB<sub>2</sub> (left panel) and ferroelectric (FE) phase CrB<sub>2</sub> (right panel). For PE CrB<sub>2</sub>, it adopts the antiferromagnetic (AFM) ground state. And for FE CrB<sub>2</sub>, it adopts the ferromagnetic (FM) ground state.



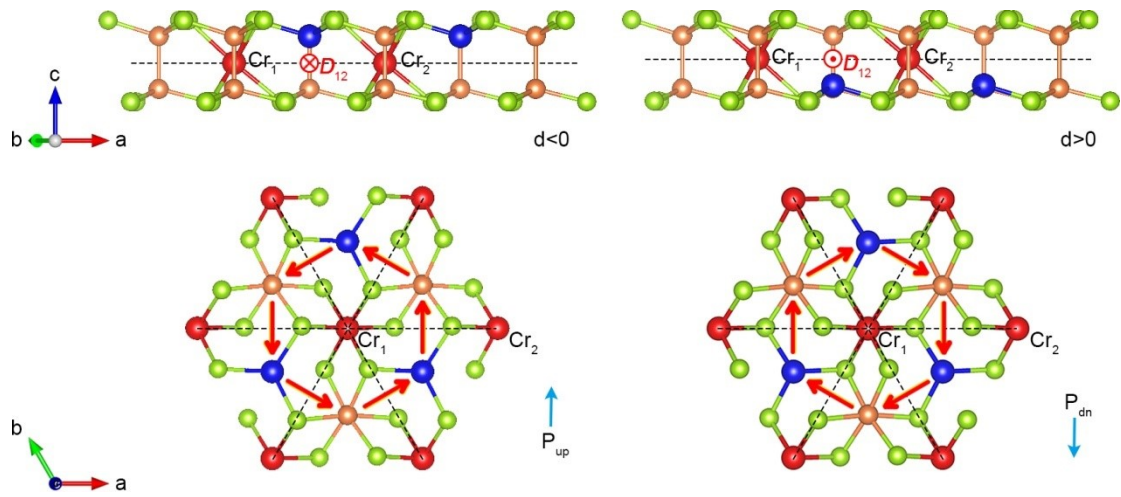
**Fig. S2.** Spin configurations for the calculations of (a) Heisenberg exchange coupling parameter  $J$  and (b) Dzyaloshinskii–Moriya interaction parameter  $d$ , showing the ferromagnetic (FM)/antiferromagnetic (AFM) collinear configurations and clockwise (CW)/anticlockwise (ACW) chiral configurations, respectively.



**Fig. S3.** Calculated normalized magnetization versus temperature for CrB<sub>2</sub>, HgInP<sub>2</sub>O<sub>6</sub>, CuCrP<sub>2</sub>S<sub>6</sub>, CuCrP<sub>2</sub>Se<sub>6</sub>, CuVP<sub>2</sub>S<sub>6</sub>, and CuVP<sub>2</sub>Se<sub>6</sub> monolayers.



**Fig. S4.** Atomic-layer-resolved localization of DMI energies  $\Delta E_{DM}$  for HgInP<sub>2</sub>O<sub>6</sub> monolayer.



**Fig. S5.** The DMI vectors  $\mathbf{D}_{12}$  (red vectors) between the nearest neighbors of Cr atoms (red balls) in  $\text{CuCrP}_2\text{Se}_6$  with opposite polarizations, where the displacement of Cu atoms (blue balls) along the plane normal causes out-of-plane polarization.