Ferroelectrically tunable magnetic skyrmions in two-dimensional multiferroics

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Supplementary Note 1: Computational details of first-principles calculations and MC simulations

Our first-principles calculations are performed on the basis of the density-functional theory (DFT), as implemented in the Vienna Ab initio Simulation Package (VASP) [1-3]. Considering the exchange-correlation interaction, the generalized gradient approximation (GGA) in the form of Perdew-Burke-Ernzerhof functional is utilized [4]. The cutoff energy is set to 500 eV, and the convergence criterion of total energy is set to $10^{-5}$ eV. Structures are fully relaxed until the force on each atom is less than $10^{-2}$ eV/Å, and a $15 \times 15 \times 1$ $k$-point mesh is used to sample the Brillouin zone. For magnetic parameters calculations, a $2\times1$ supercell with a $k$-point mesh of $16 \times 32 \times 1$ is used, while in DMI calculations, a $4\times1$ supercell with a $k$-point mesh of $8 \times 32 \times 1$ is used. To avoid interaction between adjacent layers, a vacuum space of 30 Å is adopted. To describe the strong correlated correction of Co-3d electrons, the GGA+U method is employed [5], and the U value is chosen as 2 eV.

Using the magnetic parameters obtained from first-principles calculations, parallel tempering Monte-Carlo (MC) simulations with the Metropolis algorithm are carried out to get the energy minimum spin textures [9]. The spin textures are obtained based on a $210 \times 210 \times 1$ supercell with $1 \times 10^5$ MC steps performed for each temperature (from 660 K gradually cooling down to 0 K).

![Table](https://example.com/tables.png)

**Table S1.** Magnetic parameters of Co$_2$NF$_2$ and Co$_2$NF$_2$/MoSe$_2$.
Supplementary Note 2: The formulas of {1\(e_x\), 1\(e_y\)} and {2\(e_x\), 2\(e_y\)}.

\[1\(e_x\) = \sqrt{2/3}d_{xy} - \sqrt{1/3}d_{xz},\]
\[1\(e_y\) = \sqrt{2/3}d_{x^2 - y^2} - \sqrt{1/3}d_{yz},\]
\[2\(e_x\) = \sqrt{1/3}d_{xy} + \sqrt{2/3}d_{xz},\]
\[2\(e_y\) = \sqrt{1/3}d_{x^2 - y^2} + \sqrt{2/3}d_{yz}.\] (S1)

**Fig. S1.** (a) Phonon spectra and (b) AIMD simulation results of Co\(_2\)NF\(_2\). Insert in (b) is snapshot of the structure for Co\(_2\)NF\(_2\) taken from the end of AIMD simulations (500 K and 5 ps).

**Fig. S2.** Four different magnetic configurations used to obtain the magnetic parameters of \(J\), \(\lambda\) and \(K_C\).
Fig. S3. PDOS for $d$ orbitals of (a) Co1 atom and (b) Co2 atom in Co$_2$NF$_2$.

Fig. S4. (a) Two spin-spiral configurations of CW and ACW employed to obtain $^{d \parallel}$. (b) Zero-field magnetic skyrmions of Co$_2$NF$_2$. The color map specifies the OP spin component and arrows indicate the IP component. (c) Topological charge Q in Co$_2$NF$_2$ as a function of magnetic field.
Fig. S5. Crystal structure of Co$_2$NF$_2$/MoSe$_2$ from top view, with dashed diamond indicating the unit cell.

**Supplementary Note 3: The underlying physical mechanism for the variation of $K_C$ between Co$_2$NF$_2$ and Co$_2$NF$_2$/MoSe$_2$.**

$K_C$ is substantially weakened for FE phases of Co$_2$NF$_2$/MoSe$_2$. To reveal the source of this character, we investigate the underlying physics for the single ion anisotropy $K_C$. It's well known that the single ion anisotropy originates from the joint effect of crystal field splitting and SOC. Through SOC effect, the isotropy spin angular momentum is coupled with the anisotropy orbital angular momentum induced by crystal field splitting, leading to $K_C$. According to the perturbation theory of SOC effect, $K_C$ can be estimated as follows [10]:

$$K_C = \xi^2 \sum_{M,\sigma,u,o,\sigma'} |\langle u,\sigma | L^C_x | o,\sigma' \rangle|^2 - |\langle u,\sigma | L^C_z | o,\sigma' \rangle|^2 E_{u,o,\sigma} - E_{o,o',\sigma'},$$

where $u$ and $o$ correspond to the unoccupied and occupied states of magnetic Co atoms near the Fermi level, respectively. $E_{u/o,\sigma}$ is the band energy of the state, and the spin indices $\sigma/\sigma' = \pm 1$, referring to the two orthogonal spin states. The negative (positive) value of the $K_C$ indicates OP (IP) single ion anisotropy.

In free-standing Co$_2$NF$_2$, as shown in **Fig. S3(a)**, $1a\downarrow$ is dominated in unoccupied states of Co1 atom, while the occupied states of Co1 atom is composed of $1e_{x'y}\downarrow$, $2e_{x'y}\uparrow$ and $1e_{x'z}\uparrow$. According to **Eq. S1** and **S2**, $1a\downarrow$, $1e_{x'y}\downarrow$ is negative, while $1a\downarrow$, $2e_{x'y}\uparrow$ and $1a\downarrow$, $1e_{x'z}\uparrow$ are positive. Here, $(u, o)$ represents the contribution from the SOC effect of the unoccupied state $u$ and occupied state $o$ to $K_C$. Owing to the small energy splitting between $1e_{x'y}\downarrow$ and $2e_{x'y}\uparrow$, $1a\downarrow$, $1e_{x'y}\downarrow$ and $1a\downarrow$, $2e_{x'y}\uparrow$ cancel each other, leading to a weak combined effect on $K_C$. Therefore, $K_C$ is mainly contributed by $1a\downarrow$, $1e_{x'y}\uparrow$, which is positive and consists with the OP magnetic anisotropy in Co$_2$NF$_2$. In Co$_2$NF$_2$/MoSe$_2$, as shown in **Fig. S6**, the energy splitting between $1e_{x'y}\downarrow$ and $2e_{x'y}\uparrow$ becomes larger as compared with that in Co$_2$NF$_2$. Thus, the negative $1a\downarrow$, $1e_{x'y}\downarrow$ makes a greater contribution, indicating the substantially weakened $K_C$ in Co$_2$NF$_2$/MoSe$_2$. 

**Fig. S6.** PDOS for $d$ orbitals of magnetic Co atom in (a) $+P\uparrow$ state and (b) $-P\downarrow$ state of Co$_2$NF$_2$/MoSe$_2$.


