Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Half-Metallicity in K_2CoF_4 Exfoliated Nanosheet via Defect Engineering

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The first-principles molecular dynamics (MD) simulation is performed at 300K and 400K with a canonical ensemble implemented in VASP.[1] An 4×4×1 supercell of the 2D K_2CoF_4 nanosheet containing 112 atoms is used with a Gamma-only k-point sampling. The simulation lasts for 2ps with a time step of 1.0fs.

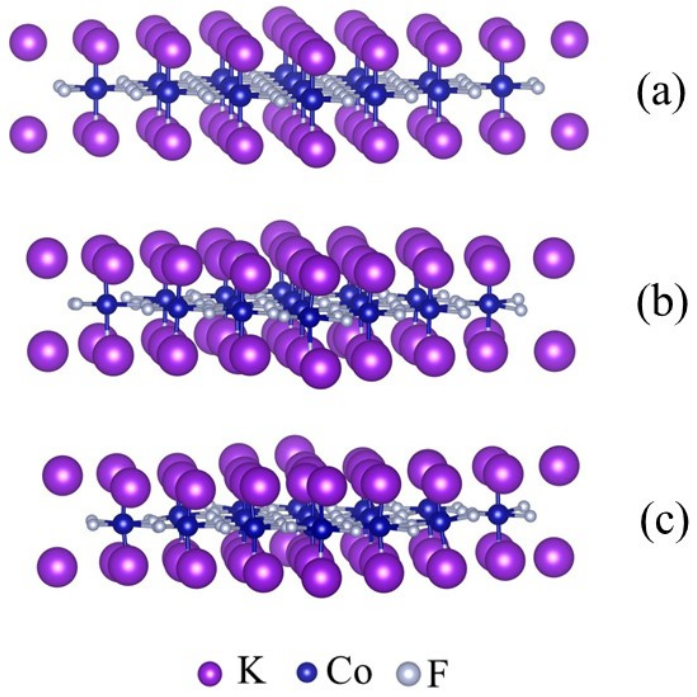

Fig.S1. (a) initial structure; (b) and (c) are the structures of K_2CoF_4 nanosheet after first-principles molecular dynamics simulation for at 300K and 400K, respectively.

We find that the intermediate states (IS1, IS2 and IS3) show a flat character in Fig.7. In order to explain these phenomena, we plot the projected density of states of F-p and Co-d orbitals and partial charge density of IS1, IS2 and IS3 in Fig.S2. As shown in Fig.S2(a, b), IS1 primarily consist of Co-dxz, and small amount of F-p states.

The corresponding partial charge density of such bands clearly shows that the electrons are localized, which lead to the flat bands.

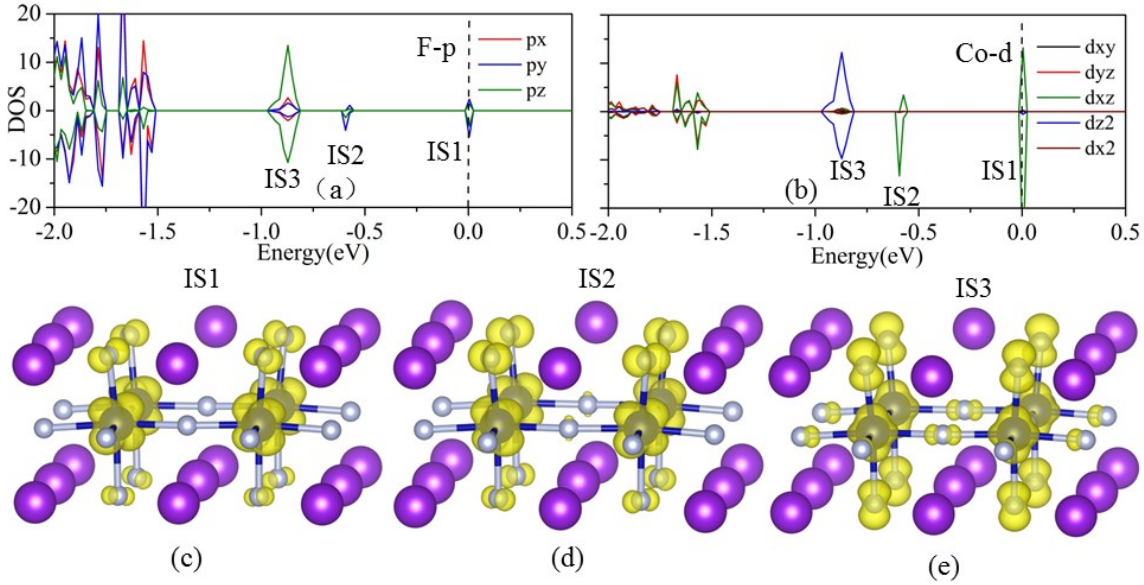

Fig.S2. (a, b) F-p and Co-d orbitals projected density of states (fermi level is presented with dashed line). (c, d and e) partial charge density of IS1, IS2 and IS3 calculated with U=4eV (isosurface value= 0.01 e/Å^3).

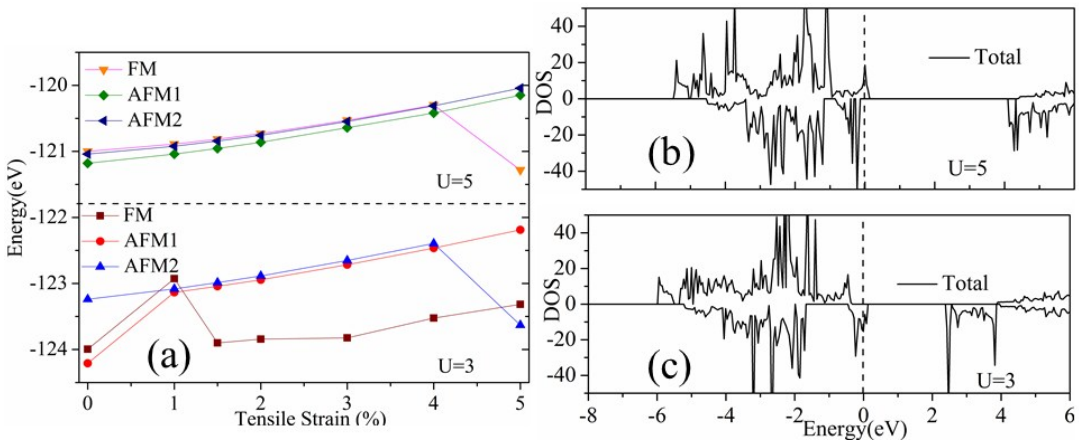

Fig.S3. Total energy for K-vacancy K_2CoF_4 nanosheet in FM, AFM1 and AFM2 states as a function of strain in (a), (b, c) show the total density of states for K-vacancy K_2CoF_4 nanosheet with U=5eV at 5% tensile strain, and U=3eV at 4% tensile strain, respectively (fermi level is presented with dashed line).

Supporting References

[1] Nosé, S., A unified formulation of the constant temperature molecular dynamics methods, J. Chem. Phys., 1984, 81, 511-519.