Supporting Information

Two-dimensional Multiferroic FeCl with Room Temperature Ferromagnetism and Tunable Magnetic Anisotropy via Ferroelectricity

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Table S1 The optimized lattice constants of 2D FeCl crystal within PBE and HSE06 functionals, respectively.

	PBE(Å)	**HSE(Å)**
a	3.22	3.30
b	3.61	3.57
Fe-Fe	2.54	2.58
Fe-Cl	2.37/2.40	2.39/2.42
d	3.71	3.71

Supporting Figures

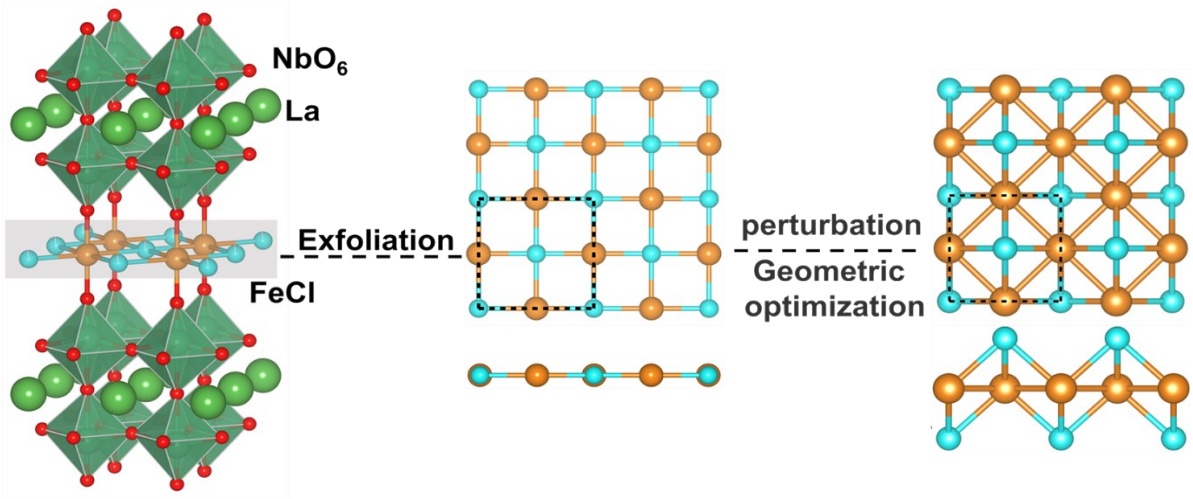

Fig. S1 The possible exfoliation process of paraelectric FeCl monolayer from layered oxyhalide (FeCl)LaNb2O7.

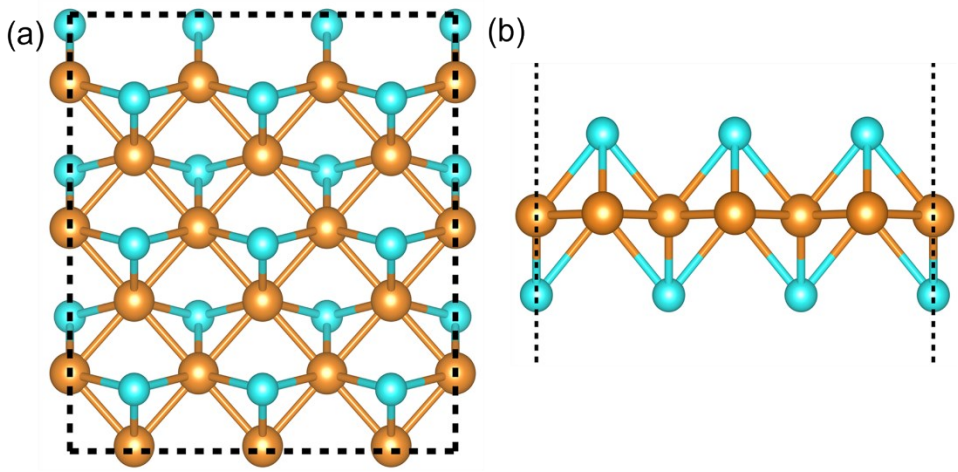

Fig. S2 The structural snapshots of 2D FeCl crystal at time of 5 ps during BOMD simulation under the temperatures of 300K.

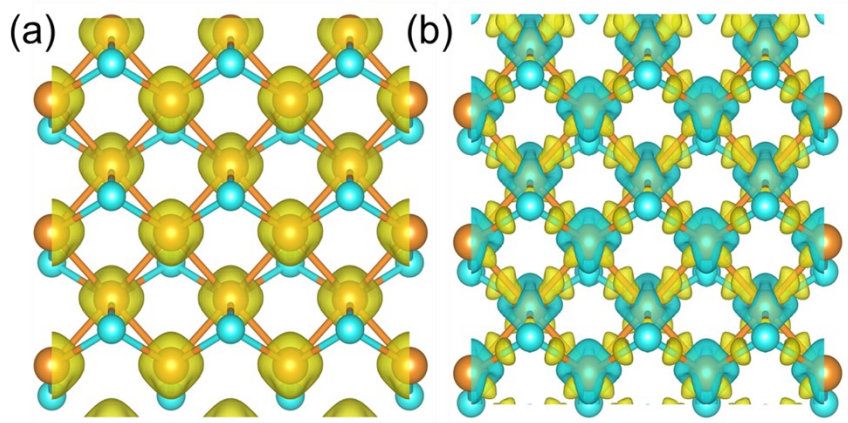

Fig. S3 The spin charge density distribution with an isosurface value of 0.02 e/Bohr^3, (b) The deformation charge density distribution with an isosurface value of 0.01e/Bohr^3, Blue and yellow denote the charge density depletion and accumulation, respectively.

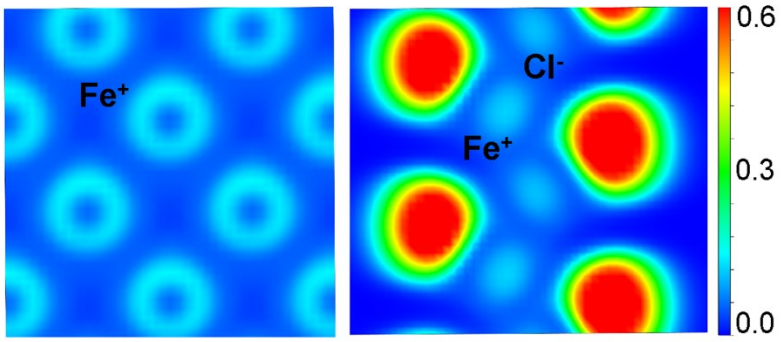

Fig. S4 The Fig.s left and right represent top view of the ELF pattern containing the Fe-Fe bonds and cross-sectional ELF pattern containing Fe-Cl bonds, respectively.

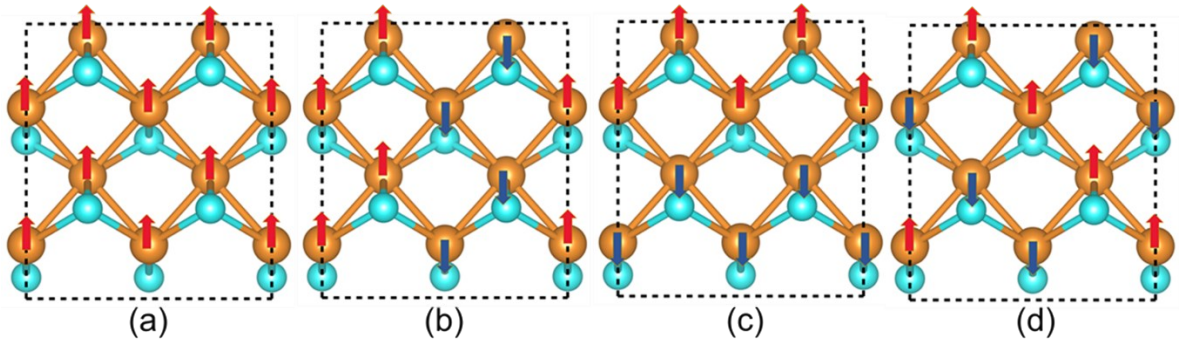

Fig. S5 (a), (b), (c) and (d) are the ferromagnetic (FM), antiferromagnetic AF1, AF2 and AF3 states, respectively. The black and blue arrows show the spin up and down orientations, respectively. The brown and green spheres represent the Fe and Cl atoms, respectively.

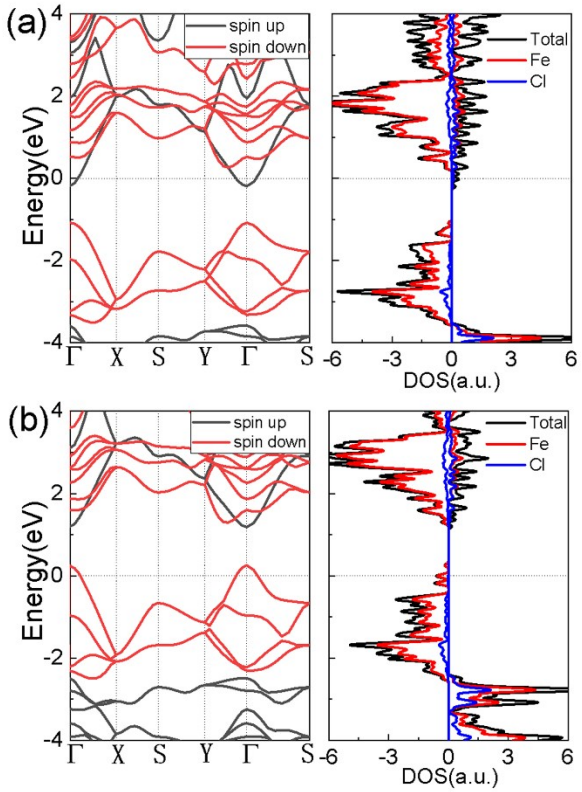

Fig. S6 Band structures and atom projected density of states for (a) electron-doped and (b) hole-doped FeCl monolayers under electron and hole doping, respectively. The Fermi levels are all set to zero.

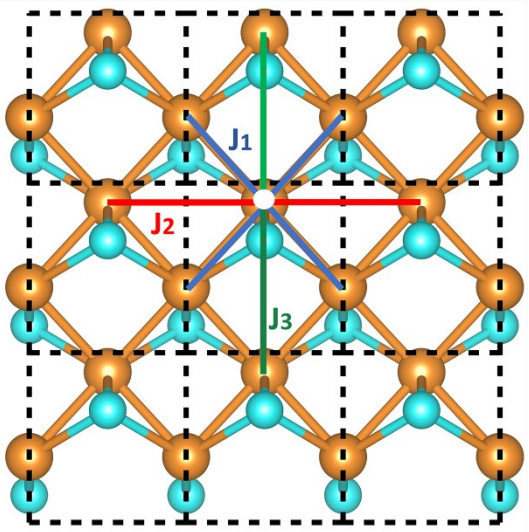

Fig. S7 The spin exchange parameters J_1, J_2, and J_3， representing the first-nearest, second-nearest and third-nearest neighboring exchange parameters, respectively.

Calculation of Young's modulus

The Young's modulus of 2D FeCl is calculated to estimate the in-plane stiffness. The calculated in-plane Young's modulus is about 43 N m^{-1}, which is comparable to germanene (42 N $m^{-1)}$, implying that 2D FeCl is mechanically stable.[1]

1. L.-M. Yang, I. A. Popov, T. Frauenheim, A. I. Boldyrev, T. Heine, V. Bačić and E. Ganz, *Phys. Chem. Chem. Phys.*, 2015, **17**, 26043-26048.