Supplementary Information

Two-dimensional CaFCl: Ultra-wide bandgap, strong interlayer quantum confinement, and n-type doping

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I. The band structure under uniaxial strains

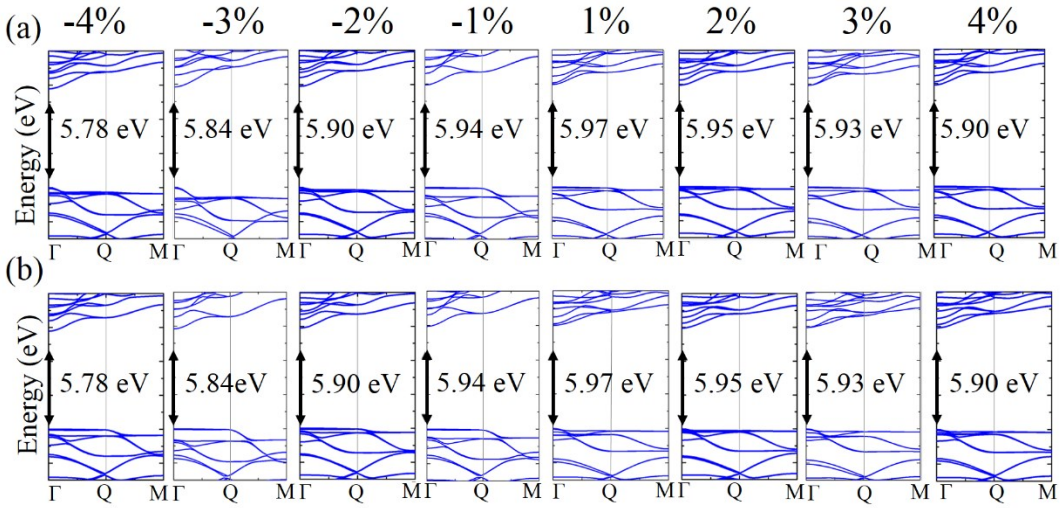

Fig. S1. The band structures of monolayer CaFCl under uniaxial strains along (a) *a* and (b) *b* directions.

II. PDOS under uniaxial strains

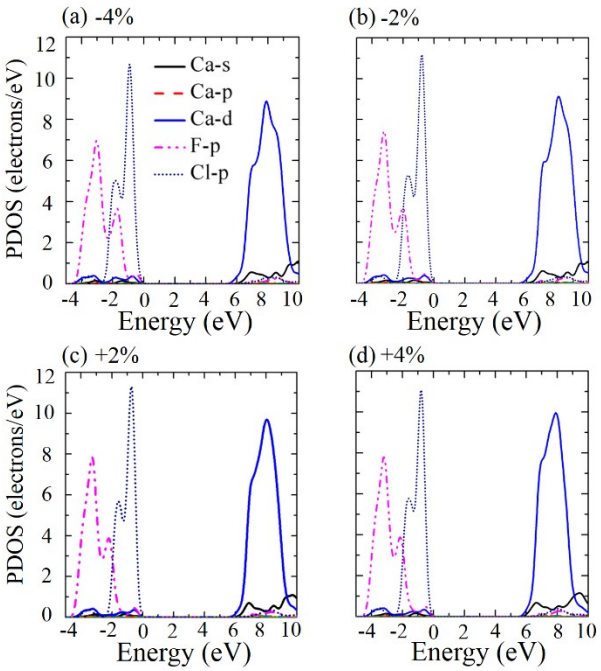

Fig. S2. The PDOS of monolayer CaFCl under -4%, -2%, 2%, and +4% uniaxial strains.

III. The band structure under biaxial strains

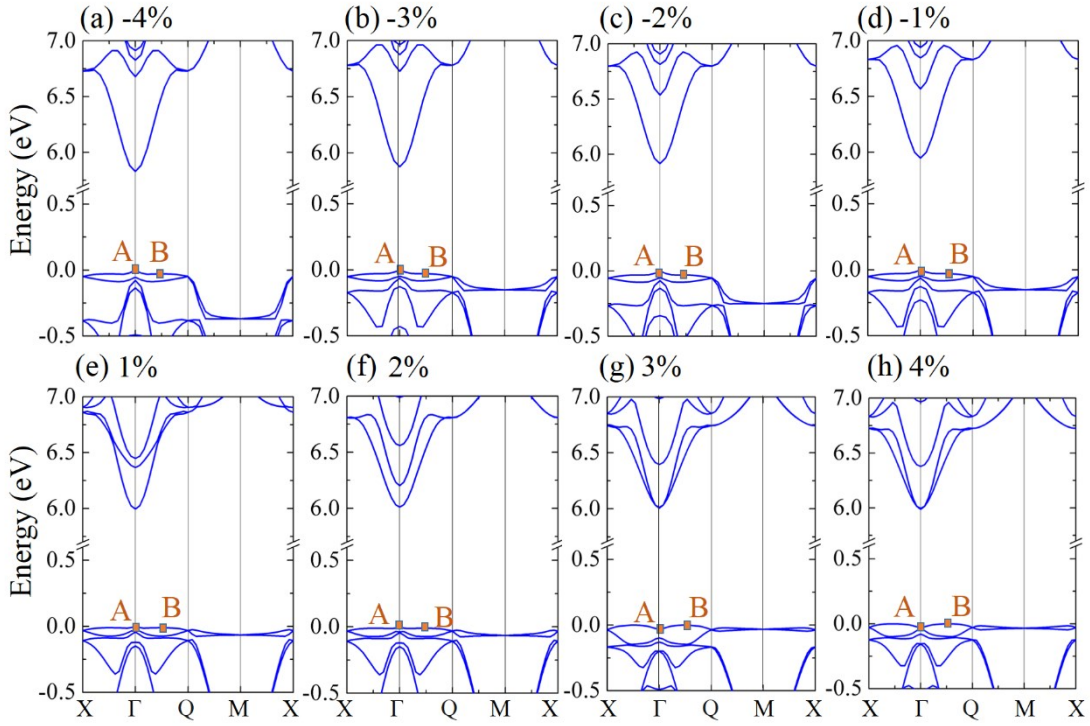

Fig. S3. The band structures of monolayer CaFCl under different biaxial strains.

IV. The relaxed structures of TTF/CaFCl

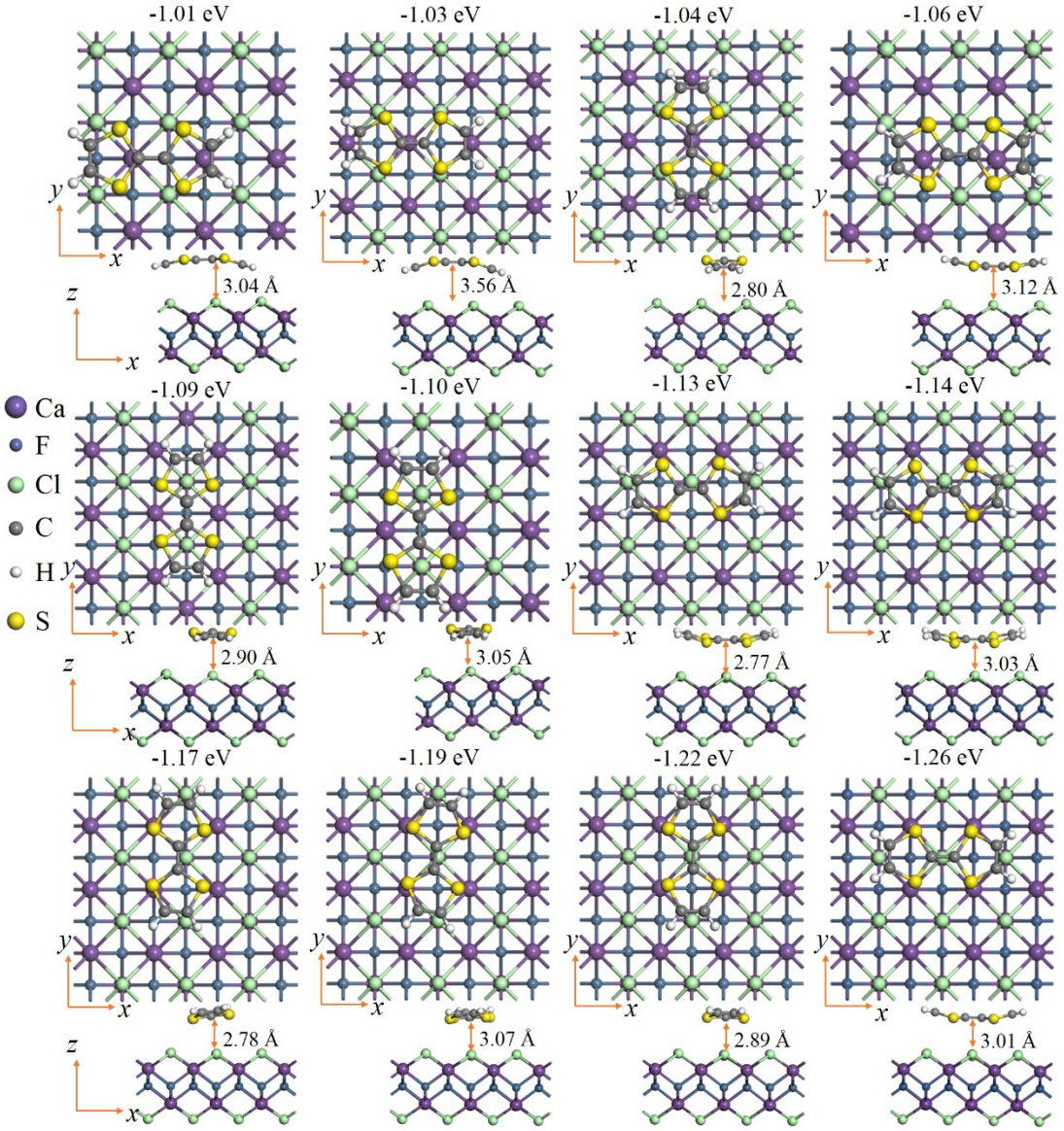

Fig. S4. Top and side views of optimized structures for TTF interacting with CaFCl monolayer. The adsorption energies and heights between the molecule and the CaFCl monolayer are also illustrated.

V. Computation detail of the minimum vertical distance

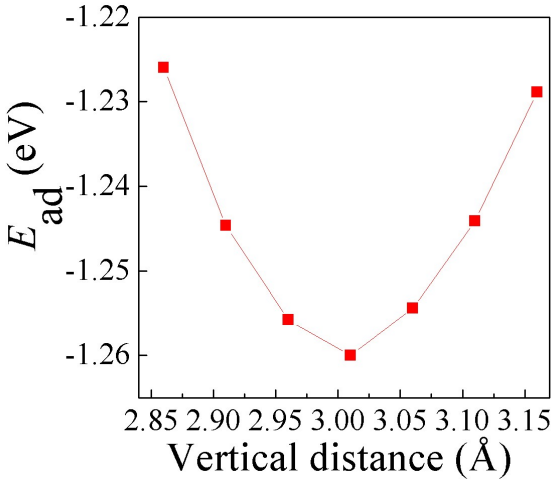

Fig. S5. Relative adsorption energies of the most stable configuration for TTF/CaFCl as a function of the vertical distance.

VI. The highest occupied molecular orbital of the TTF/CaFCl system

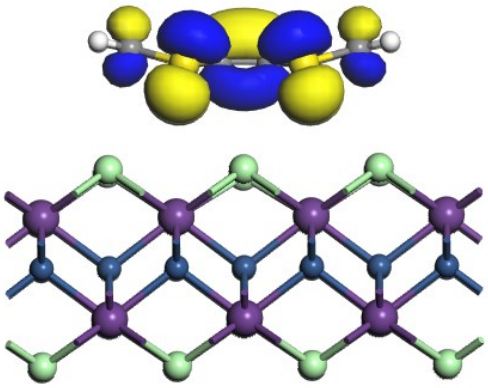

Fig. S6. The highest occupied molecular orbital of TTF/CaFCl at Γ point. The isovalue is set to 0.03 Å^{-3}.

VII. Computation detail of in-plane Young's modulus (Y_{2D})

The in-plane Young's modulus along *a* or *b* can be derived from $\frac{\Delta E}{S_0} = Y_{\mathrm{D}} \frac{(\Delta l / l_0)^2}{2}$, where ΔE is the total energy difference under each strain, and S_0 is the lattice area of monolayer CaFCl. We use $\Delta l/l_0$ ranging from -1.5% to 1.5% to fit the value of Y_{2D} (Fig.

S7).

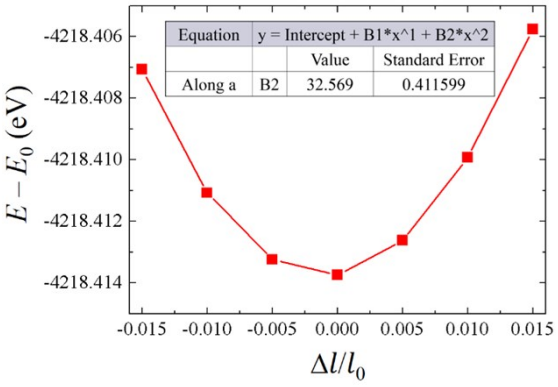

Fig. S7. In-plane Young's modulus of monolayer CaFCl along *a* direction. The fitted curve along *b* direction is consistent with that along *a* direction.