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Supplementary Materials

Tunable Electronic and Magnetic Properties of Monolayer and Bilayer Janus $Cr_2Cl_3I_3$: A First-Principles Study

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1. The optimizatd lattice is used fitting the total energy and the lattice parameter. The corresponding optimized lattice is 6.626 Å with fitting energy and lattice, which is agreed with the method of the global optimization.

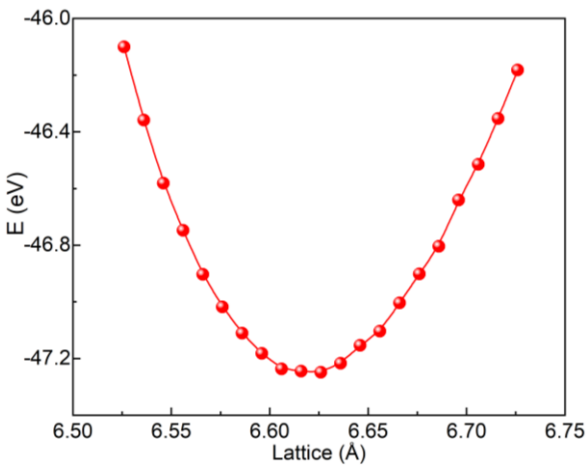

Fig. S1. The relationship between the total energy and the lattice of ML Janus $Cr_2Cl_3I_3$.

2. The band structure and density of the states of the ML CrI_3 and $CrCl_3$ with FM order are also calculated by HSE06 functional. Both CrI_3 and $CrCl_3$ show obvious spin-polarization with FM ground state. The VBM come from the contribution of Cr and Cl atoms in the ML $CrCl_3$, while VBM of CrI_3 come from the I atoms. The CBM are composed of Cl atom.

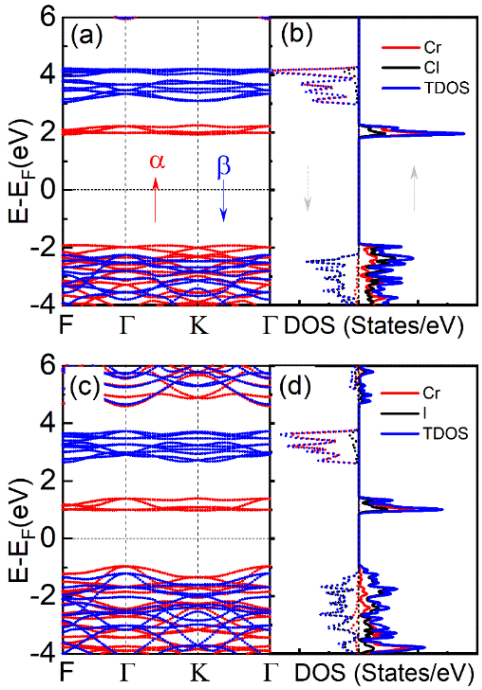

Fig. S2. The band structure (a) and density of (b) the states of ML $CrCl_3$ and (c) (d) of CrI_3, respectively. The red and blue presents spin-α and spin-β electrons, respectively. The solid and dashed lines present spin-α and spin-β electrons in the density of the states.

3. The partial band structure of the monolayer (ML) Janus CrClI with HSE06 functional is calculated. From the Fig. S1, we can find that the CBM is mainly consisted of d_{xz} and d_{yz} orbitals of Cr atoms, while the VBM is mainly consisted of Cl atoms. For the energy states in the energy range of (1.1, 1.7) and (2.7, 3.9) eV are mainly contributed by $d_{x^2\text{-}y^2}$, d_{xy}, d_{xz}, d_{yz} and d_{z^2} orbitals.

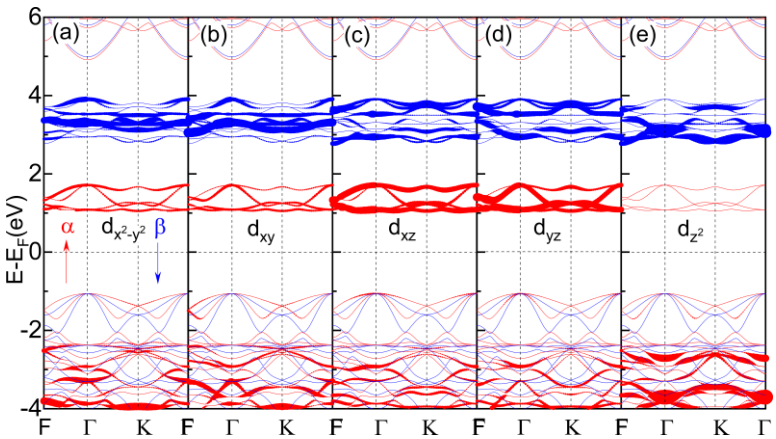

Fig. S3. The band structure of the ML Janus $Cr_2Cl_3I_3$ with HSE06 functional. The states near the fermi-level are mainly contributed by the Mo atoms. The band structure of monolayer of $Cr_2Cl_3I_3$ with the contribution of (a) $d_{x^2\text{-}y^2}$, (b) d_{xy}, (c) d_{xz}, (d) d_{yz}, and (e) d_{z^2} orbitals of Cr atoms.

4. The PDOS of the ML Janus $Cr_2Cl_3I_3$ are calculated by HSE06 functional. The VBM are mainly contributed by the p_y and p_x orbitals of I atoms, the CBM mainly comes from the contribution of the d_{xz}, d_{yz}, d_{xy}, $d_{x^2\text{-}y^2}$ of Cr atoms. And this is consistent with the analysis of the band structure.

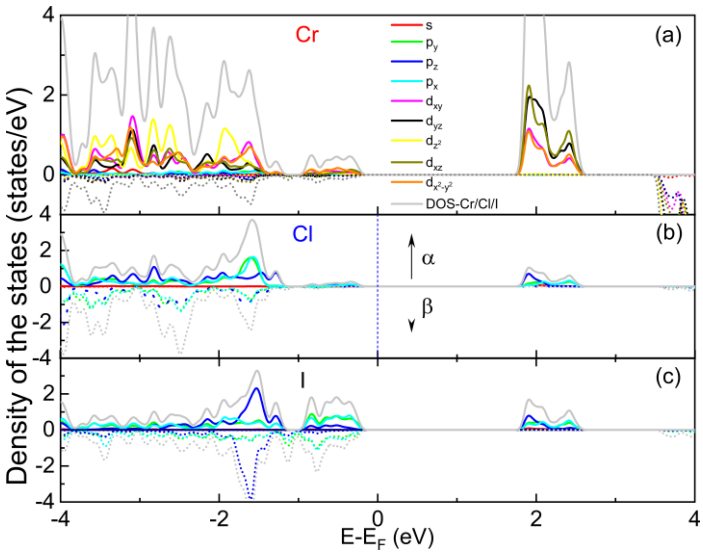

Fig. S4. The partial density of the state of the ML $Cr_2Cl_3I_3$ is calculated by HSE06 functional. The (a) Cr, (b) Cl and (c) I atoms projected density of the state of the ML $Cr_2Cl_3I_3$, respectively.

5. For the ML Janus $Cr_2Cl_3I_3$, the spin-orbital coupling (SOC) effect the electronic properties. Therefore, the band structure with projections with SOC is also calculated with HSE06. There is obvious spin splitting at points of high symmetry, especial at Γ and K point. There is spin splitting energy of 119 meV at K point.

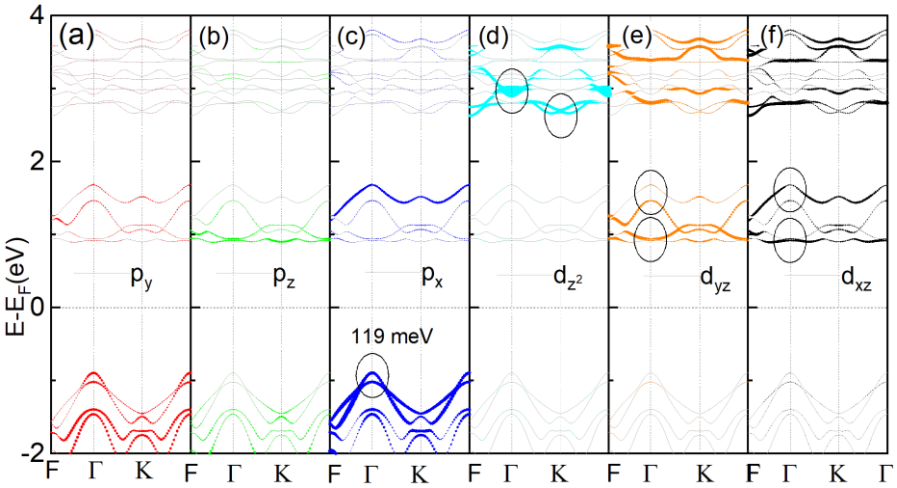

Fig. S5. The band structure of ML $Cr_2Cl_3I_3$ calculated by HSE06 including spin-orbital coupling, and the weights of (a) p_y, (b) p_z, (c) p_x, (d) d_{z^2}, (e) d_{yz}, and (f) d_{xz} orbitals of the Cr atoms, respectively.

6. The band structure of the ML Janus $Cr_2Cl_3I_3$ under the biaxial strains, with ε = -7.7%, -7.8%, -7.9%, -8.0% using HSE06 functional. For ε = -7.7% ,the ML CrClI is at FM ground state. The ML CrClI is BMS with direct band gap of 1.514 eV, and both VBM and CBM are K and Q point (middle point between Γ and K point), respectively. As the ε increases to the -8.0%, the ML CrClI is transferred from the FM to the AFM ground state. The corresponding ML CrClI has spin-unpolarized gap of 1.474 eV.

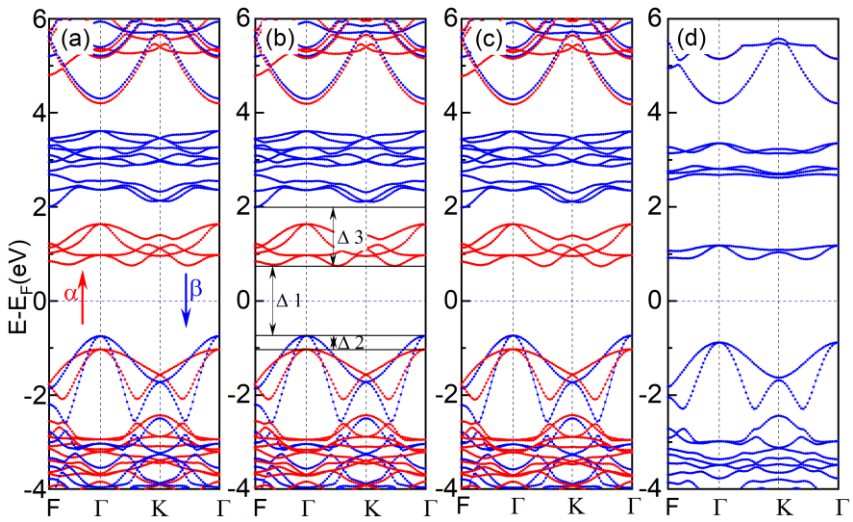

Fig. S6. The band structure of ML Janus $Cr_2Cl_3I_3$ under the biaxial strains with (a) ε = -7.7% (b) ε = -7.8% (c) ε = -7.9%, and (d) ε = -8.0%, respectively. The lines with red, blue and green dots represent the weights of the Mo, S and Se atoms, respectively.

7. The phonon spectrum and phonon density of the monolayer CrI_3 and $CrCl_3$. The phonon spectra, phonon density of states are calculated using finite displacement method, in which we use a supercell with dimension of 4×4×1 for CrI_3 and $CrCl_3$. In the calculation of phonon spectra, and the geometries are fully relaxed until energy and force is converged to 10^{-8} eV and 10^{-3} eV/Å, respectively.

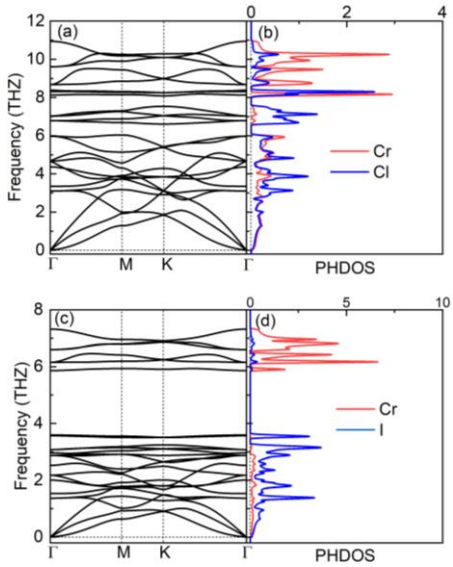

Fig. S7. Phonon spectra, phonon density of states (PHDOS) of ML (a, b) of $CrCl_3$, and (c, d) of CrI_3, respectively.