# Supporting Information

# Bi-stimuli assisted engineering and control of magnetic phase in monolayer CrOCl

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# **Results and Discussion**

#### S1. Structural stability of the strained system

### 1. Formation Energy

The stability of the monolayer system was studied by plotting the formation energy of the structures at different values of strains applied along different directions.



Fig S1. Formation energy of monolayer CrOCl as a function strain along various directions.

The formation energy (E<sub>form</sub>) of CrOCl is calculated using the following equation:

$$E_{form} = (aE_{Cr} + bE_{O} + cE_{Cl} - E_{CrOCl})/(a + b + c)$$

Where  $E_{Cr}$ ,  $E_0$ ,  $E_{Cl}$  are the total energies of chromium, oxygen and chlorine atoms respectively and  $E_{Crocl}$  is the total energy of a CrOCl unit cell consisting of a=b=c=2 atoms each of Cr, O and Cl.

The above equation yields an energy of 4.4 eV/atom for the unstrained CrOCl system which is quite less compared to graphene (7.85 eV/atom) and comparable to another 2D system like Fe<sub>2</sub>Si with 4.10 eV/atom (Nano letters, 17(5), 2771-2777). Similarly, Ni<sub>2</sub>X exhibited formation energies of 4.80 eV/atom and 4.42 eV/atom respectively for X=Si and Ge (Physical Chemistry Chemical Physics, 17(39), 26043-26048) and a new type of 2D transition metal borides (MnB) offers average formation energy of 6.08 eV/atom (Nanoscale Horizons, 3(3), 335-341).

From Fig. S1, it can also be observed that upon increasing the strain along various directions, the formation energy tends to remain more or less unchanged even for a high strain rate of 10%, thus displaying the structural stability of the material at these strain rates.

#### 2. Phonon band structure



Fig S2. Phonon bandstructures for strain applied along the x direction with a value of: (a) -10% (b) unstrained (c) +10%

The phonon band structure of a system is related to the stability of the system as illustrated in Fig. S2 for  $\varepsilon_x = \pm 10\%$ . For phonon dispersion calculations, the unit cell consisting of 2 atoms each of Cr, O, and Cl were considered. The kpoint mesh of 8 x 8 x 1 was taken into account, while also enabling the finite displacement method, and a phonon convergence threshold of 1 x 10<sup>-14</sup> eV was set for better accuracy. In all the three cases in Fig. S2, no occurrence of negative phonon frequencies is observed. The phonon bands start and end at the  $\Gamma$  point (zone centre). The absence of imaginary bands in all these cases indicate the stability of the structures under different strain conditions.

#### S2. Projected density of states (PDOS) and orbital bonding diagrams in xy and yz plane



Fig. S3 (a) Projected density of states (PDOS) of CrOCl in the unstrained configuration. The orbital bonding pictures in the 3 different planes: along (b) xz-plane, (c) xy-plane and (d) yz-plane.

The projected density of states (PDOS) of the system representing the contribution of various atomic orbitals involved are shown in Fig. S3(a) along with the relative orbital bonding pictures for several views in the xz, xy and yz planes depicted in Fig. S3(b, c, d).

S3. Basis Sets for total energy and electronic structure calculations



Fig S4. Total energy of CrOCl monolayer in the unstrained configuration using Gaussian and plane wave basis sets for different energy cut-off.

The above depicted plot shows the variation of total energy of CrOCl monolayer in the unstrained configuration as a function of energy cut-off used for different basis sets. The lowest total energy will provide the ground state energy of the system. From Fig. S4, it can be observed that the lowest total energy is obtained when the LCAO (linear combination of atomic orbitals / atomic cantered Gaussian basis) basis set is considered, rather than the plane wave basis. However, on increasing the cut-off energy, both methods converge and provide similar ground states. Hence both the schemes help in providing similar ground state energies at higher energy cut-off. Another point to note is that LCAO method is computationally less demanding and works very well for bulk systems compared to plane wave which requires much more processing capacity for calculations requiring high wave energy cut offs, which is another reason for specifying lower cut-offs involving plane wave calculations. This is why LCAO basis set was used for the present work.

#### S4. Effect of Hubbard U over the system

| Hubbard U <sub>eff</sub> (eV) | Energy difference ( $\Delta E = E_{FM} - E_{AFM}$ ) in meV |
|-------------------------------|--|
|                               |  |
| 5                             | 12.2   |
|                               |  |
| 6                             | 11.3   |
| 7                             | 10.7   |

Table S1. Energy difference ( $\Delta E = E_{FM} - E_{AFM}$ ) as a function of Hubbard U

In order to study the U dependence, the total energy difference (between the FM and AFM states) for unstrained CrOCl as function of different Hubbard U values was computed and the results have been depicted in Table S1. From the above table it can be observed that for a variation in U values from 5-7 eV, only a slight deviation (<1.5 meV/TM atom) in  $E_{FM} - E_{AFM}$  is observed, suggesting that the total energy difference is only slightly influenced by the aforementioned U values. Hence an effective U in the range 5 – 7 eV could be used for electronic study and total energy calculations.



Fig S5. Energy difference ( $\Delta E = E_{FM} - E_{AFM}$ ) as a function of Hubbard U

Further the energy difference as a function of U ranging from 2-7 eV was computed and plotted as shown in the Fig.S5. It can be observed that the lowest energy difference between the two magnetic configurations with the ferromagnetic ground state being the favourable one is obtained for U=7 eV.

## S5. Calculations for Electric field + Strain ( $\varepsilon_x = -4\%$ , $\varepsilon_y = 4\%$ and $\varepsilon_b = 4\%$ )

In order to observe the combined effect of external stimuli (electric field + strain) at low strain conditions additional calculations were performed considering varying electric field (5V, 10V, 15V, 20V) + strain ( $\varepsilon_x = -4\%$ ,  $\varepsilon_y = 4\%$  and  $\varepsilon_b = 4\%$ ).



Fig. S6. Critical temperature dependence on applied voltages along various crystallographic directions: (a) z, (b) x, y, z in the presence of strain.

1. As seen from Fig. S6(a), we can notice significant variations in T<sub>c</sub> for  $\varepsilon_x = -4\%$  case with a critical enhancement of ~7%, increasing the T<sub>c</sub> from 182 K to 195 K at V<sub>z</sub> = 20V. We have also noticed enhancement in T<sub>c</sub> for  $\varepsilon_y =$ 

4% where there is an elevation of critical temperature from 153 K to 156 K on varying the voltage from 0 V to 20V. Similar trend was noticed for  $\varepsilon_b$  =4%, where the T<sub>c</sub> increased from 154 K to 157 K while changing the voltage from 0 V to 20 V applied along the z direction.

2. Fig. S6(b) depicts a specific case of  $\varepsilon_b = +4\%$  with electric field being applied similar to that of  $\varepsilon_b = +8\%$  in the manuscript. Here for an applied voltage of  $V_x = V_y = V_z = 5V$ , a sudden decrease in  $T_c$  was observed and the system turns AFM above 5V. An increase in critical temperature was observed from 154 K to 203 K in the presence of  $V_z + V_y$ . However, a 30% enhancement in  $T_c$  was seen on exposing the material with  $V_z = V_x = 20V$ , where the temperature was elevated to 220 K from 154 K.