Supporting Information

Cr_2TiC_2-based double MXenes: Novel 2D bipolar antiferromagnetic semiconductor with gate-controllable spin orientation towards antiferromagnetic spintronics

Junjie He*[,1,2], Guangqian Ding[2], Chengyong Zhong[1,2], Shuo Li[3], Dengfeng Li[2], Gang Zhang*[,4]

[1] *Institute for Advanced Study, Chengdu University, Chengdu 610100, P. R. China*

[2]*School of Science, Chongqing University of Posts and Telecommunications, Chongqing 400065, P. R. China.*

[3]*Faculty of Science, Charles University in Prague, Hlavova 8, 128 43 Prague 2, Czech Republic.*

[4]*Institute of High Performance Computing, A*STAR, Singapore 138632.*

1. The band structure of symmetrical functionalization of double MXenes $Cr_2TiC_2F_2$

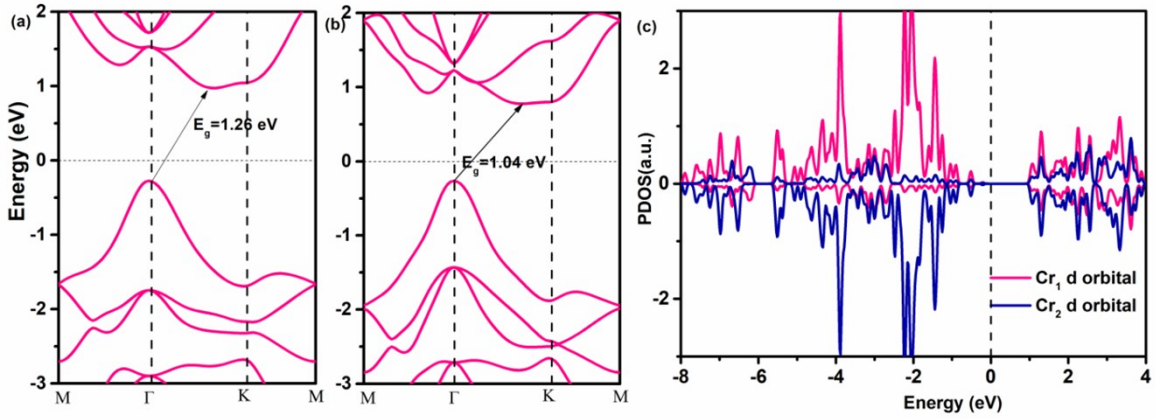

Figure S1: (a) $Cr_2TiC_2F_2$ and (b) $Cr_2TiC_2Cl_2$. (c) The PDOS of *d* states are shown for the Cl-bonded (Cr_1) and F-bonded (Cr_2) Cr atoms in $Cr_2TiC_2F_2$, respectively. The Fermi level is set to zero.

2. Non-collinear magnetic structure.

The triangular spin lattice typically results in a frustration, which have been found in has been found in $CrSe_2$.[1] Therefore, we consider a non-collinear AFM (NCAFM) configuration with the spin vectors of the nearest-neighboring Cr atoms at 120° to each other as one of possibility of the spin arrangement in Cr_2TiC_2FCl MXenes system in Figure S2. Luckily, by considering the spin-orbital coupling, our calculation indicate the AFM-c state as more stable, which gains $E_{NCAFM/AFM-c}=E_{NCAFM}-E_{AFM-c}$=339 meV over the described NCAFM phase. Furthermore, we calculate the magnetic anisotropy energy E_{MAE} between magnetic moment in plane and out of plane for the Cr_2TiC_2FCl MXenes, which defined by $E_{MAE}=E_{in-plane}-E_{out-of-plane}$, the E_{MAE} turns out to be positive and equal to 1.221 meV, indicating an out-of-plane orientation of magnetism moment. Thus, the Cr_2TiC_2FCl system shows the AFM-c ground states with out-of-plane magnetic anisotropy.

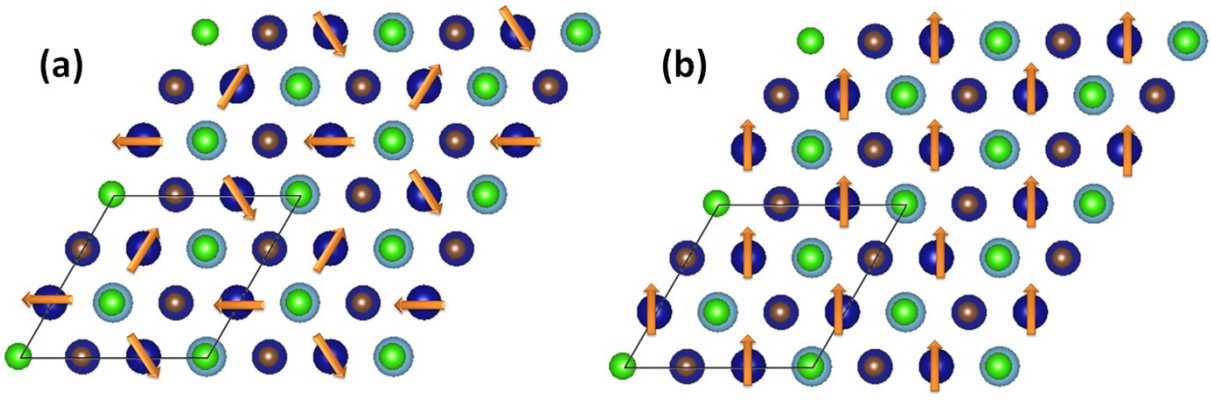

Figure S2: A 2D $(\sqrt{3}\times\sqrt{3})R30°$ cell is considered.(a) non-collinear AFM (NCAFM) configuration. (b) AFM-c configuration with SOC. The yellow arrows denote the different magnetic configuration. The arrow of magnetic moment in (b) are shown in *xy* plane for visualization purpose. (in fact, the arrow is always in out of plane under consideration)

3. The Atomic Model for Mixed functionalization Cr_2TiC_2 double MXenes

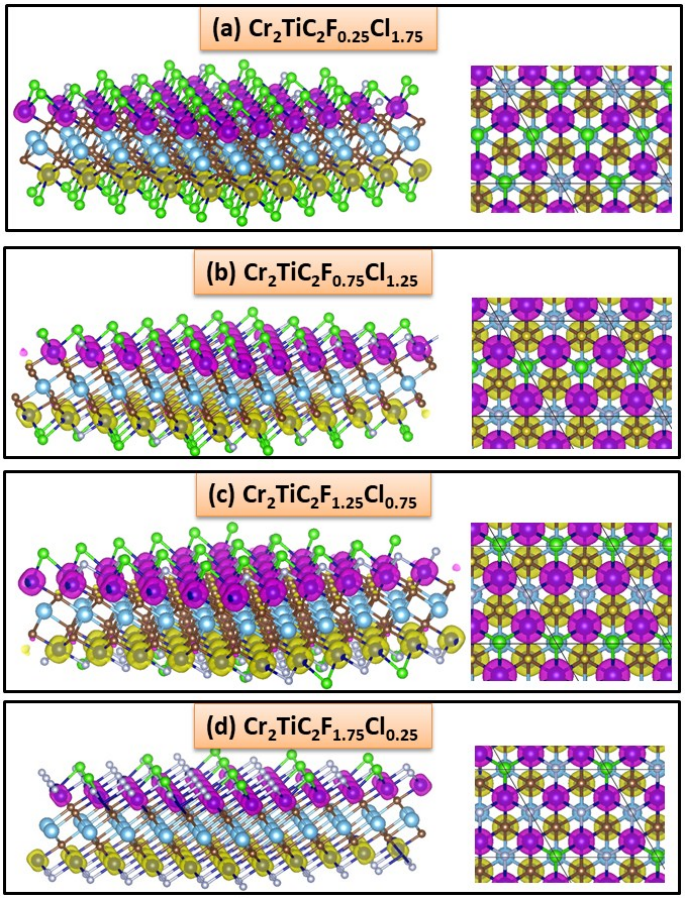

Figure S3: The various atomic model of $Cr_2TiC_2F_xCl_{2-x}$ systems, x=0.25 (a), 0.75 (b), 1.25 (c),1.75 (d) with spin-polarized charge density are considered. The blue and yellow colors represent the spin up and spin down spin-polarized charge densities, respectively. The iso-surface is set to 0.03 e/Å^3.

Reference:

[1] H. Y. Lv *et al. Phys. Rev. B*, 2015, **92**(21), p.214419.