Supporting Information

Cr$_2$TiC$_2$-based double MXenes: Novel 2D bipolar antiferromagnetic semiconductor with gate-controllable spin orientation towards antiferromagnetic spintronics

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1. The band structure of symmetrical functionalization of double MXenes

**Cr₂TiC₂F₂**

![Energy bands and PDOS](image)

**Figure S1:** (a) Cr₂TiC₂F₂ and (b) Cr₂TiC₂Cl₂. (c) The PDOS of d states are shown for the Cl-bonded (Cr₁) and F-bonded (Cr₂) Cr atoms in Cr₂TiC₂F₂, 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 CrSe₂.¹ 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 Cr₂TiC₂FCl MXenes system in Figure S2. Luckily, by considering the spin-orbital coupling, our calculation indicate the AFM-c state as more stable, which gains $E_{NCAFM/AFM-c} - E_{AFM-c} = 339$ 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 Cr₂TiC₂FCl 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 Cr₂TiC₂FCl 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)](image)
3. The Atomic Model for Mixed functionalization Cr$_2$TiC$_2$ double MXenes

Figure S3: The various atomic model of Cr$_2$TiC$_2$F$_x$Cl$_{2-x}$ 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 isosurface is set to 0.03 e/A$^3$.

Reference: