Intrinsic magnetism and biaxial strain tuning in two-dimensional metal halide $V_3X_8$ ($X = F, Cl, Br, I$) from first principles and Monte Carlo simulation

Haibo Xiao$^{a,*}$, Xiaonan Wang $^a$, Ruilong Wang $^a$, Lingfang Xu $^a$, Shiheng Liang$^a$, Changping Yang $^{a,*}$

$^a$Hubei Key Laboratory of Ferro & Piezoelectric Materials and Devices, Faculty of Physics & Electronic Science, Hubei University, Wuhan, 430062, People's Republic of China

Corresponding authors' information:

Haibo Xiao (Hubei University, Wuhan, China), E-mail: xhb7142@hubu.edu.cn

Changping Yang (Hubei University, Wuhan, China), E-mail: cpyang@hubu.edu.cn

Tel: +86-27-88665447

Fig S1. Orbital-resolved electronic band structures of $V_3Br_8$ with different spin component ($\uparrow$) and ($\downarrow$) spins. The Fermi level is set as zero. The V-d and Br-p orbitals are mapped with different colors and sizes.
Fig. S2. Electronic band structures of (a, b) \( V_3F_8 \), (c) \( V_3Cl_8 \) and (d, e) \( V_3I_8 \) calculated with GGA+\( U_{\text{eff}} \). The Fermi level is set as zero. The V-d and X (X = F, Cl, Br, I)-p orbitals are mapped with different colors and sizes.

Fig. S3. The first \( (J_1) \) and second nearest-neighboring \( (J_2) \) exchange interaction in \( V_3F_8 \) (\( V_3I_8 \)) structures.
According to the FM and AFM configurations shown in Fig. 2, the energy function are expressed as follows:

\[ J_1 \text{ and } J_2 \text{ are calculated with the following equations:} \]

\[ E_{FM} = E_0 - 24J_1S^2 - 24J_2S^2 \]
\[ E_{AFM_1} = E_0 - 8J_1S^2 + 8J_2S^2 \]
\[ E_{AFM_2} = E_0 + 8J_1S^2 - 8J_2S^2 \]

Where \( S \) represents the local magnetic moment of V atom in 2D monolayer \( V_3I_8 \). Then the exchange parameters can be obtained as: \( J_1 = 10.79 \text{ meV}(7.46 \text{ meV}) \), and \( J_2 = -1.97 \text{ meV}(1.64 \text{ meV}) \).

Fig. S4 (a) The \( E_{AFM} - E_{FM} \) calculated with GGA+U\(_{\text{eff}}\) as a function of biaxial strain for 2D \( V_3I_8 \). (b-g) Spin-resolved electronic band structures using GGA+U\(_{\text{eff}}\) with respect to biaxial strains from -10% to 8% for \( V_3I_8 \), respectively.

Fig. S5 (a) Strain-dependent the \( E_{AFM} - E_{FM} \) with GGA+U for 2D \( V_3F_8 \). (b-g) The evolution of band structures using GGA+U\(_{\text{eff}}\) under biaxial strains from -8% to 8% for \( V_3F_8 \).
Fig. S6 (a) The energies as a function of biaxial strain for two-dimensional metal halide V$_3$Cl$_8$ with different magnetic configurations. (b-f) Calculated spin-resolved electronic band structures with respect to biaxial strains from compressed stage of -10% to tensile strain of 5% of AFM metal halide V$_3$Cl$_8$.

Fig. S7 The energy variation of $E_{FM}$ - $E_{AFM}$ and calculated magnetic moment per unit cell versus carrier doping. The positive and negative value indicate electron and hole doping in the neutral system, respectively.
Fig. S8 The evolution of band structure under the variation of carrier concentration for V3I8.