Electronic Supporting Information

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

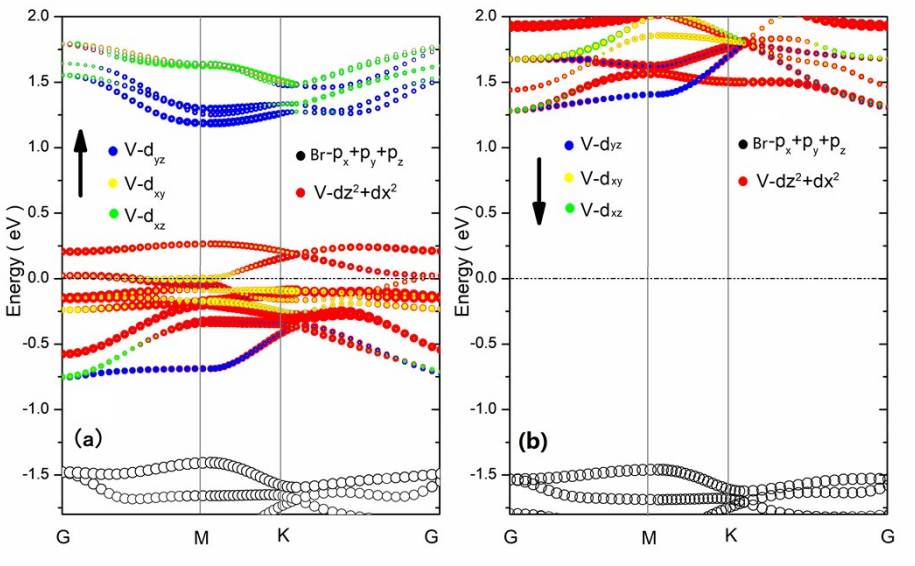

Fig S1. Orbital-resolved electronic band structures of V_3Br_8 with different spin component (↑) and (↓) spins. The Fermi level is set as zero. The V-d and Br-p orbitals are mapped with different colors and sizes.

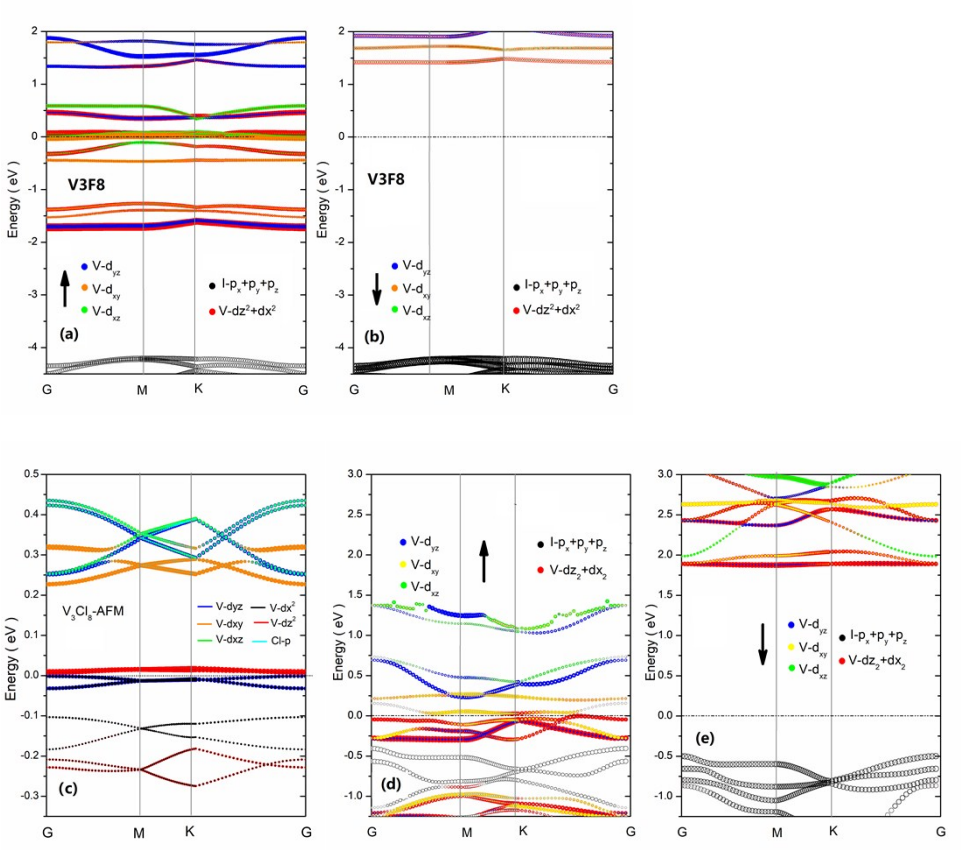

Fig. S2. Electronic band structures of (a, b) V_3F_8, (c) V_3Cl_8 and (d, e) V_3I_8 calculated with GGA+U_{eff}. The Fermi level is set as zero. The V-d and X (X = F, Cl, Br, I)-p orbitals are mapped with different colors and sizes.

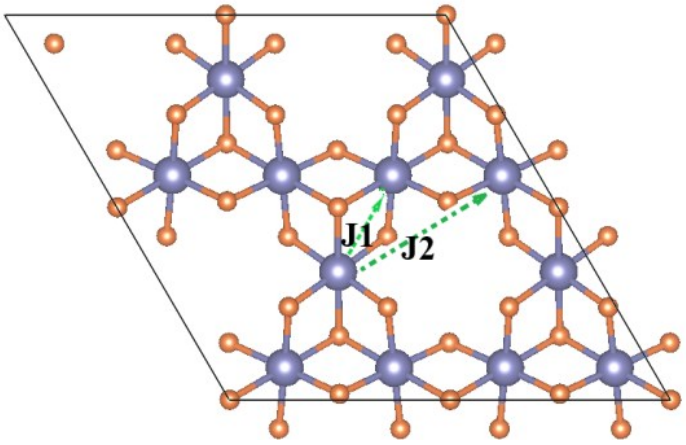

Fig .S3 The first (J_1) and second nearest-neighboring (J_2) exchange interaction in V_3F_8 (V_3I_8) structures.

According to the FM and AFM configurations shown in Fig. 2, the energy function are expressed as follows:

J_1 and J_2 are calculated with the following equations:

$$E_{FM} = E_0 - 24J_1S^2 - 24J_2S^2$$
$$E_{AFM_1} = E_0 - 8J_1S^2 + 8J_2S^2$$
$$E_{AFM_3} = E_0 + 8J_1S^2 - 8J_2S^2$$

Where S represents the local magnetic moment of V atom in 2D monolayer V_3F_8 (V_3I_8). Then the exchange parameters can be obtained as: J_1 = 10.79 meV(7.46 meV), and J_2 = -1.97 meV(1.64 meV).

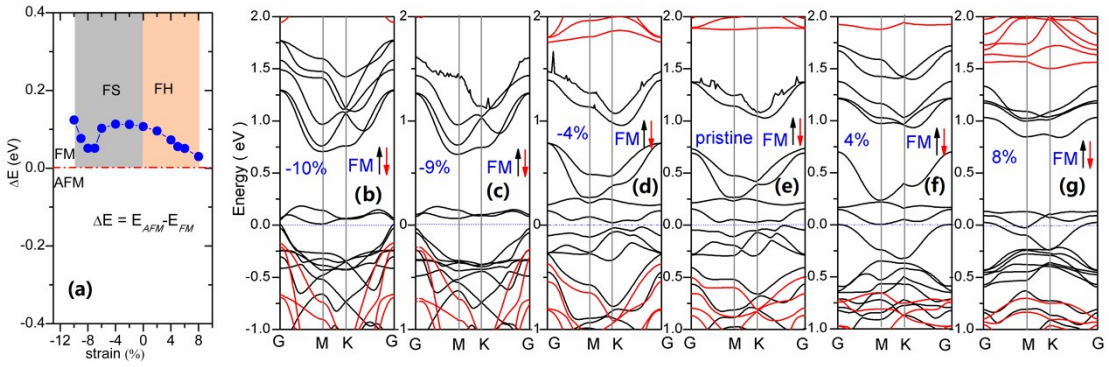

Fig. S4 (a) The E_{AFM}– E_{FM} calculated with GGA+U_{eff} as a function of biaxial strain for 2D V_3I_8. (b-g) Spin-resolved electronic band structures using GGA+U_{eff} with respect to biaxial strains from -10% to 8% for V_3I_8, respectively.

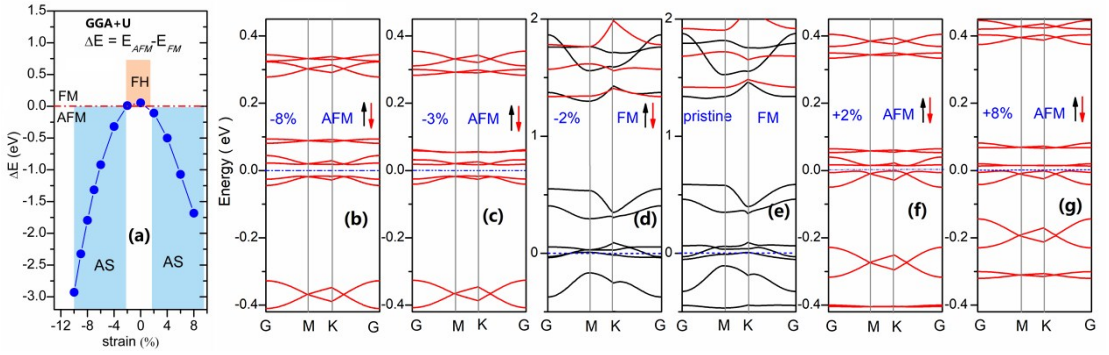

Fig. S5 (a) Strain-dependent the E_{AFM}– E_{FM} with GGA+U for 2D V_3F_8. (b-g) The evolution of band structures using GGA+U_{eff} under biaxial strains from -8% to 8% for V_3F_8.

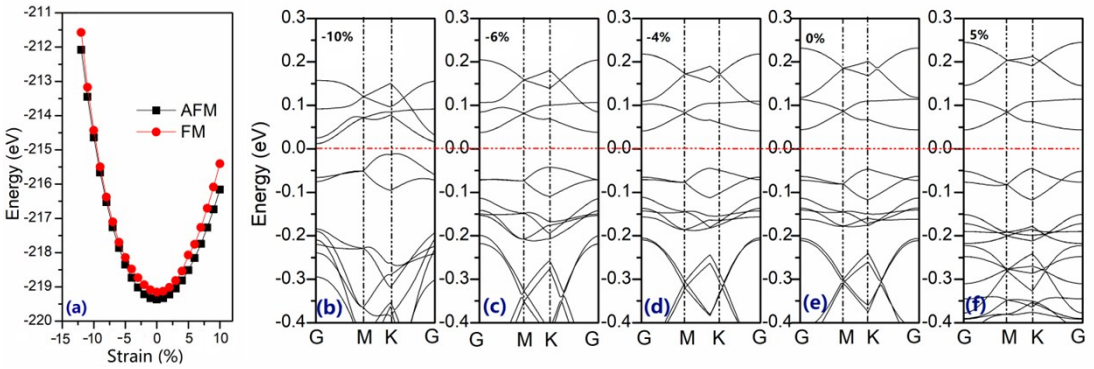

Fig.S6 (a) The energies as a function of biaxial strain for two-dimensional metal halide V_3Cl_8 with different magnetic configurations. (b-f) Calculated spin-resolved electronic band structures with respect to biaxial strains from compressed stage of -10% to tensile strain of 5% of AFM metal halide V_3Cl_8.

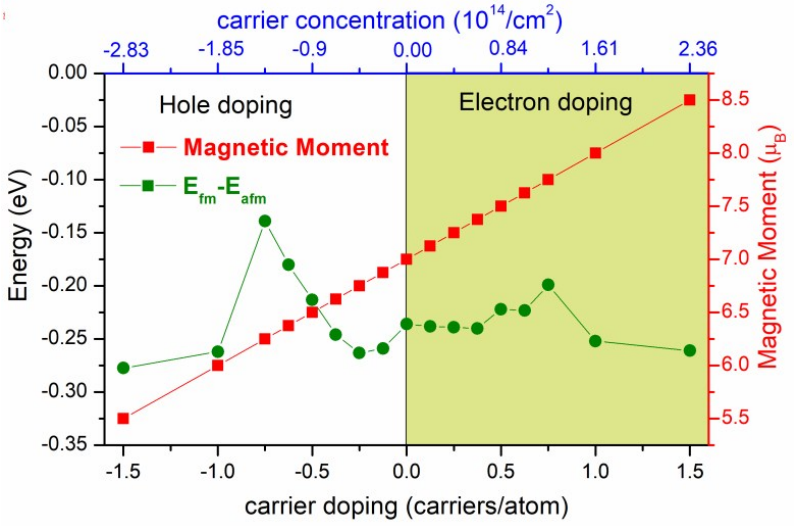

Fig. S7 The energy variation of $\mathbf{E_{FM}}$– $\mathbf{E_{AFM}}$ and calculated magnetic moment per unit cell versus carrier doping. The positive and negative value indicate electron and hole doping in the neutral system, respectively.

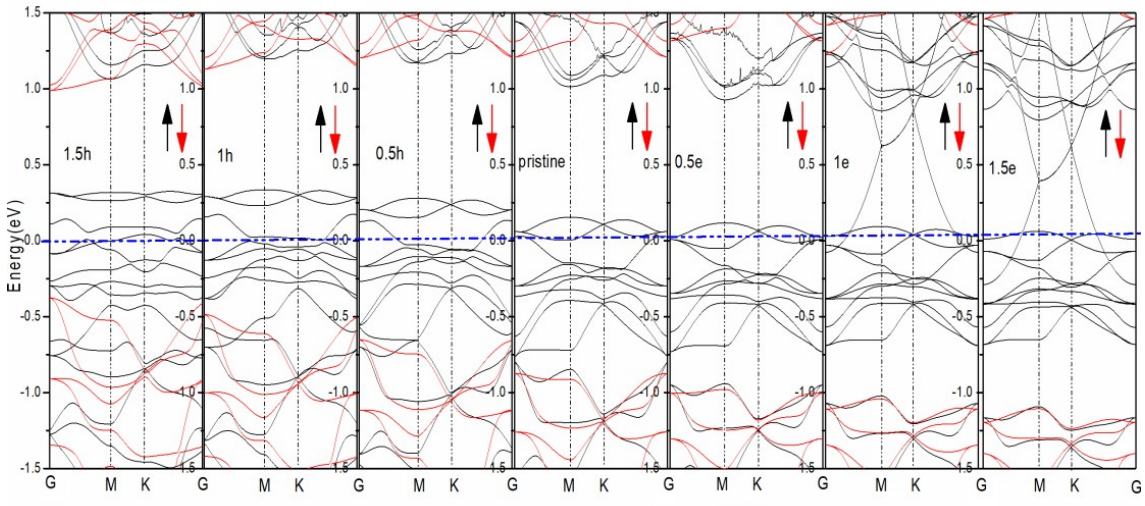

Fig. S8 The evolution of band structure under the variation of carrier concentration for V3I8.