Electronic Supplementary Information

Temperature Dependent Magnetic Properties of Room-temperature Ferromagnetic Janus Monolayer Fe$_2$XY (X, Y = I, Br or Cl)

Chenhai Shen,[1] Xueping Li, Tianxing Wang, Guangtao Wang, and Congxin Xia*
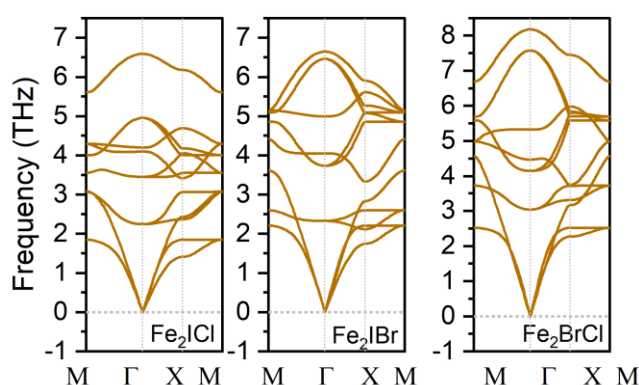
A. Phonon spectra and AIMD

Fig. S1. Phonon dispersion of Janus Fe$_2$XY (X, Y = I, Br, Cl; X ≠ Y) monolayers.
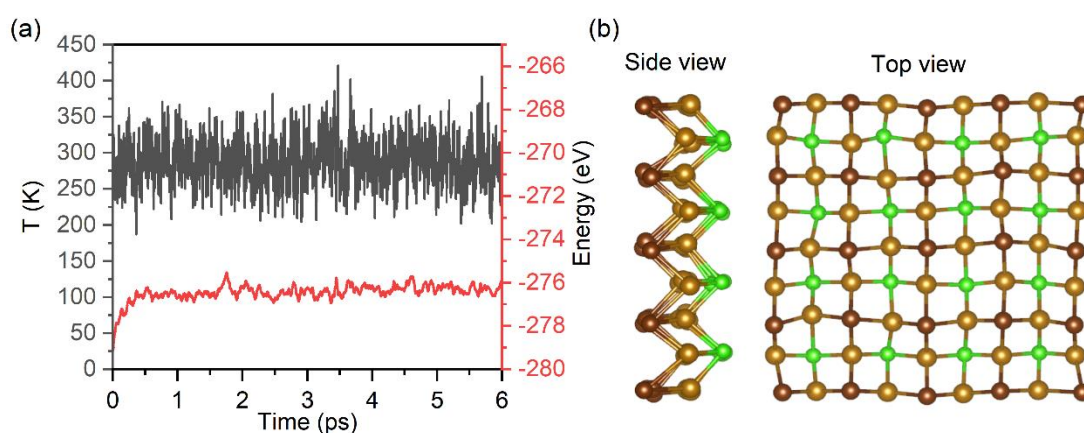

Fig. S2. The temperature and total energy fluctuations for Janus monolayer Fe$_2$BrCl with a ferromagnetic configuration at 300 K. The *ab initio* molecular dynamics simulation was conducted by the NVT ensemble with a supercell size of 4 × 4 × 1 for 6000 fs with a time step of 1 fs.

School of Physics, Henan Normal University, Xinxiang 453007, China.
E-mail: shenchenhai@htu.edu.cn, xiacongxin@htu.edu.cn

B. Effects of U_{eff} on the lattice constant and magnetic ground state

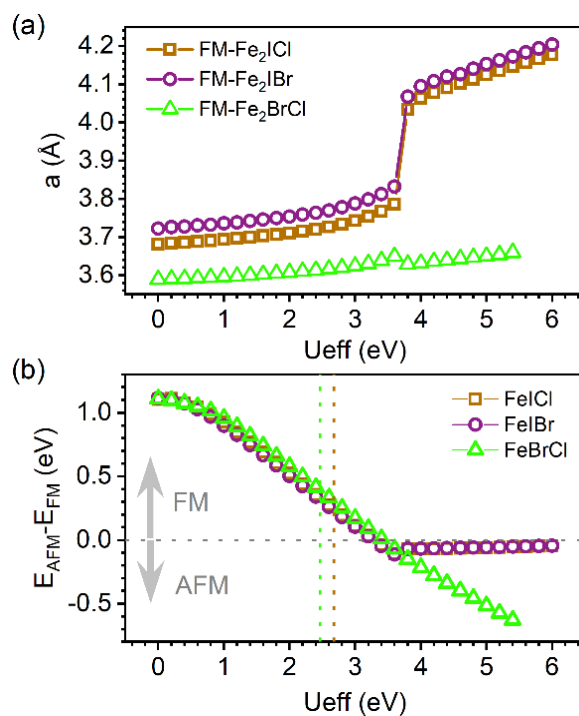Fig. S3 Effects of electronic correlation on lattice constants and magnetic ground states. (a)the variation of lattice constant with the effective on-site Coulomb interaction U_{eff}, and (b)the variation of the energy difference between antiferromagnetic and ferromagnetic states as a function of U_{eff}.
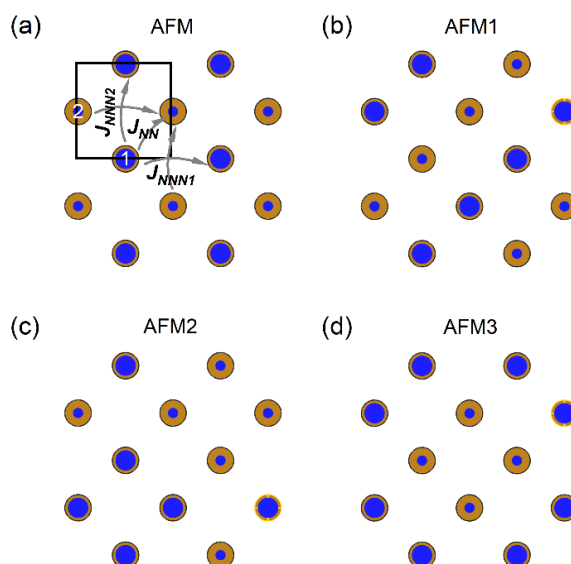
C. Magnetic exchange constants

Fig. S4. Four configurations of antiferromagnetic ordering, where the ligands are omitted and only the magnetic Fe atoms are retained, and the large (small) blue circles labeled in Fe atoms represent the spins perpendicular to the paper pointing outward (inside).

When calculating the magnetic exchange parameters, we consider the interaction of one Fe atom with its nearest neighbors (NN) and with its next nearest neighbors (NNN). For Fe$_2XY$, each Fe atom is surrounded by four NN Fe atoms and four NNN Fe atoms. However, we shall note that the interactions of one Fe atom with its NNN atoms are classified into two types: One is the interaction between one Fe and its NNN atoms connected via X atoms, another is between one Fe and its NNN connected via Y atoms. As shown in Fig. S4(a), the former is defined as J_{NNN1} and the latter as J_{NNN2}. For the FM configuration of $4 \times 4 \times 1$ supercell, we can get:

$$4E_{FM} = 8 \times \left[4 \times \frac{1}{2} \times (-J_{NN}) + 2 \times \frac{1}{2} \times (-J_{NNN1}) + 2 \times \frac{1}{2} \times (-J_{NNN2}) \right] + E_{other}$$
$$= -16J_{NN} - 8J_{NNN1} - 8J_{NNN2} + E_{other} \tag{1}$$

where E_{FM} is the total energy of the unit cell of Fe$_2XY$, J_{NN} is the magnetic exchange parameter between one Fe with its NN Fe atoms, and E_{other} is other energy of $4 \times 4 \times 1$ supercell.

Similarly, for a particular Fe atom in the AFM configuration shown in Fig. S4(a), it has four NN atoms with opposite spin and four NNN atoms with same spin. Hence, we can get:

$$E_{AFM} = 8 \times \left[4 \times \frac{1}{2} \times J_{NN} + 2 \times \frac{1}{2} \times (-J_{NNN1}) + 2 \times \frac{1}{2} \times (-J_{NNN2}) \right] + E_{other}$$
$$= 16J_{NN} - 8J_{NNN1} - 8J_{NNN2} + E_{other}. \tag{2}$$

For the AFM1 configuration shown by Fig. S4(b), we can get:

$$E_{AFM1} = 8 \times \left[2 \times \frac{1}{2} \times (-J_{NN}) + 2 \times \frac{1}{2} \times J_{NN} + 2 \times \frac{1}{2} \times J_{NNN1} + 2 \times \frac{1}{2} \times J_{NNN2} \right] + E_{other}$$
$$= 8J_{NNN1} + 8J_{NNN2} + E_{other}. \tag{3}$$

For the AFM2 configuration shown by Fig. S4(c), we can get:

$$E_{AFM2} = 8 \times \left[2 \times \frac{1}{2} \times (-J_{NN}) + 2 \times \frac{1}{2} \times J_{NN} + 2 \times \frac{1}{2} \times J_{NNN1} + 2 \times \frac{1}{2} \times (-J_{NNN2}) \right] + E_{other}$$
$$= 8J_{NNN1} - 8J_{NNN2} + E_{other}. \tag{4}$$

For the AFM3 configuration shown by Fig. S4(d), we can get:

$$E_{AFM3} = 8 \times \left[2 \times \frac{1}{2} \times (-J_{NN}) + 2 \times \frac{1}{2} \times J_{NN} + 2 \times \frac{1}{2} \times (-J_{NNN1}) + 2 \times \frac{1}{2} \times J_{NNN2} \right] + E_{other} \quad (5)$$
$$= -8J_{NNN1} + 8J_{NNN2} + E_{other}.$$

Next, by subtracting Eq. 1 from Eq. 2, we get:

$$E_{AFM} - 4E_{FM} = 32J_{NN}. \quad (6)$$

Subtracting Eq. 4 or 5 from Eq. 3 gets

$$E_{AFM1} - E_{AFM2} = 16J_{NNN2} \quad (7)$$

or

$$E_{AFM1} - E_{AFM3} = 16J_{NNN1} \quad (8)$$
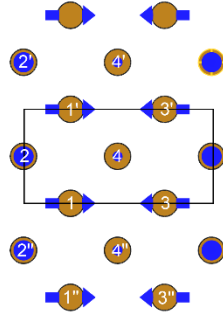
D. DMI parameters

Fig. S5. Configuration of clockwise (CW) spin rotation. The rectangle of black lines is a unit cell of the CW configuration, and the order of observing spin rotation is from 2 to 1 to 4 and then to 3.

Figure S5 shows the configuration of clockwise spin rotation.

$$E_{cw} = -2 \times [\frac{1}{2}(-\frac{\sqrt{2}}{2}D_{14} - \frac{\sqrt{2}}{2}D_{12} - \frac{\sqrt{2}}{2}D_{12''} - \frac{\sqrt{2}}{2}D_{14''}) - \frac{1}{2}[2 \times J_{NNN2} - 2 \times J_{NNN1})]$$
$$-2 \times [\frac{1}{2}(-\frac{\sqrt{2}}{2}D_{43'} - \frac{\sqrt{2}}{2}D_{41'} - \frac{\sqrt{2}}{2}D_{41} - \frac{\sqrt{2}}{2}D_{43}) - \frac{1}{2}(2 \times J_{NNN1} - 2 \times J_{NNN2})] \quad (9)$$
$$+E_{other}$$

where the multiplicator 2 in front of the square brackets means that the energy of atoms 1 (4) and the energy of atom 3 (2) are equal. Because the magnitude of the DMI constants between one Fe and its four NN atoms is equal, Equation 9 can simplify as

$$E_{CW} = 4\sqrt{2}D_{NN} + E_{other}. \quad (10)$$

Similarly, for the ACW configuration, we can get

$$E_{ACW} = -4\sqrt{2}D_{NN} + E_{other}. \quad (11)$$

Subtracting Eq. 11 from Eq. 10 leads to

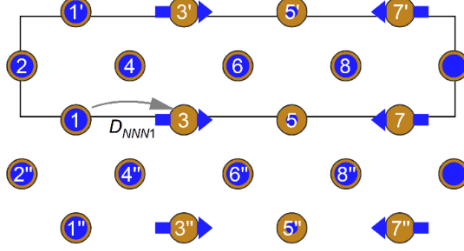$$E_{CW} - E_{ACW} = 8\sqrt{2}D_{NN} \tag{12}$$

Fig. S6. Configuration of clockwise (CW) spin rotation only in the sub lattice 1. The rectangle of black lines is a unit cell of the CW configuration, and the order of observing spin rotation is from 1 to 3 to 5 and then to 7.

Figure S6 shows another CW spin configuration, which aims to calculation the DMI parameter (D_{NNN1}) between a particular Fe atom and its NNN atoms bridged to it via the X atom. In this configuration, the magnetic sublattice 2 remains FM ordering while the magnetic sublattice 1 possesses a CW configuration. Thus, the magnetic exchange coupling of a magnetic atom with its NN atoms cancels mutually in the $4 \times 4 \times 1$ supercell. For the exchange coupling of a magnetic atom with its NNN atoms, its size in the CW configuration is equal to that in the ACW configuration. Meanwhile, the DMI interaction between a magnetic atom and its NN atoms will cancels mutually in the supercell. For simplicity, these interactions will be omitted in the following expression.

$$E_{CW} = 4 \times [-\frac{1}{2} \times (-D_{35}) - \frac{1}{2} \times (-D_{31})] + E_{other} = 4D_{NNN1} + E_{other} \tag{13}$$

For the ACW configuration, we can get

$$E_{ACW} = 4 \times (-\frac{1}{2} \times D_{35} - \frac{1}{2} \times D_{31}) + E_{other} = -4D_{NNN1} + E_{other} \tag{14}$$

Hence,

$$E_{CW} - E_{ACW} = 8D_{NNN1} \tag{15}$$

Similarly, for the CW configuration only in sublattice 2, we can get

$$E_{CW} - E_{ACW} = 8D_{NNN2} \tag{16}$$

E. Curie temperature
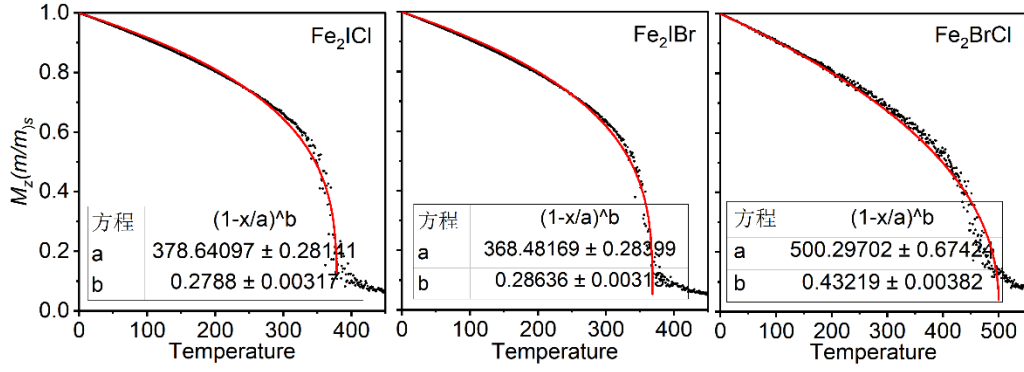

Fig.S7. Curie temperature of Janus monolayers Fe$_2$ICl, Fe$_2$IBr and Fe$_2$BrCl.

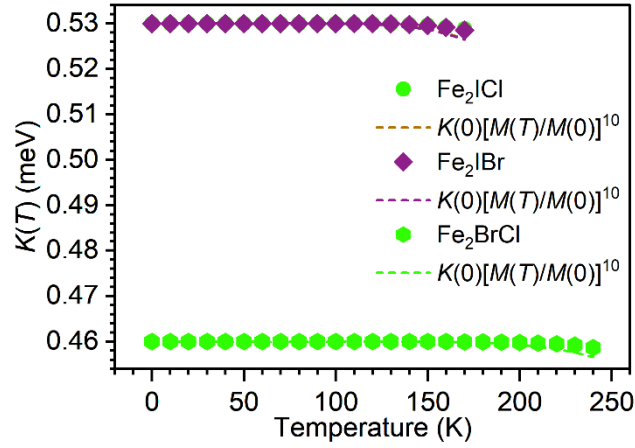# F. Temperature-dependent *K* based on the tenth-power law

Fig.S8. Temperature-dependent uniaxial anisotropy constant *K* of Janus monolayer Fe$_2XY$ (*X* = I and Br, *Y* = Br and Cl, *X* ≠ *Y*), which is obtained by the rescaling parameter based on the tenth-power law. The rescaling parameters is 8.0, 8.2 and 8.6 for Fe$_2$ICl, Fe$_2$IBr and Fe$_2$BrCl monolayers. In this case, we can observe a subtle decrease in *K*(*T*) as temperature increases within $T \lesssim 0.5T_C$. This observation indicates that the coercive field will slightly decreases with the increasing temperature in this temperature region.