Electronic Supplementary Information for

Electronic Structure, Magnetic Anisotropy and Dzyaloshinskii-Moriya Interaction in Janus $Cr_2I_3X_3$ (X = Br, Cl) Bilayers

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Table S1 Lattice constant a (Å), interlayer distance d (Å) and band gap E_g (eV) of the $AA_{up\text{-}up}$ $Cr_2I_3Br_3$ bilayer obtained from DFT, DFT-D2 and HSE06 functionals.

	DFT	DFT-D2	HSE06
a (Å)	6.79	6.69	6.69
d (Å)	4.23	3.39	3.39
E_g (eV)	0.732	0.761	1.389

A. Total DMI parameters d_{tot}

The Dzyaloshinskii-Moriya interaction (DMI) energy can be written as $\boldsymbol{E}_{DM} = \boldsymbol{d}_{12}\cdot(\boldsymbol{S}_1\times\boldsymbol{S}_2)$, where $\boldsymbol{S}_1$ and $\boldsymbol{S}_2$ represent the spins of the nearest neighbor Cr atoms.[1] The DMI vector $\boldsymbol{d}_{12}$ can be expressed as $\boldsymbol{d}_{12} = d_{12}\cdot(\boldsymbol{z}\times\boldsymbol{u}_{12})$, where $\boldsymbol{z}$ is unit vectors pointing along z and $\boldsymbol{u}_{12}$ represents unit vectors pointing from site 1 to site 2. The Cr atoms with chiral spin configuration are labeled in Fig. S1(a). Neglecting the weak interlayer contribution, the DMI energy of atom 3 can be written as

$$E_3 = \frac{1}{2}[\boldsymbol{d}_{33'}\cdot(\boldsymbol{S}_3\times\boldsymbol{S}_{3'}) + d_{32'}\cdot(\boldsymbol{S}_3\times\boldsymbol{S}_{2'}) + d_{32''}\cdot(\boldsymbol{S}_3\times\boldsymbol{S}_{2''})] + E_{other}, \quad \text{(S1)}$$

where the factor 1/2 represents the DMI energy sharing between the two sites of each bond and E_{other} includes the energy of anisotropy and symmetric exchange. Taking into account the

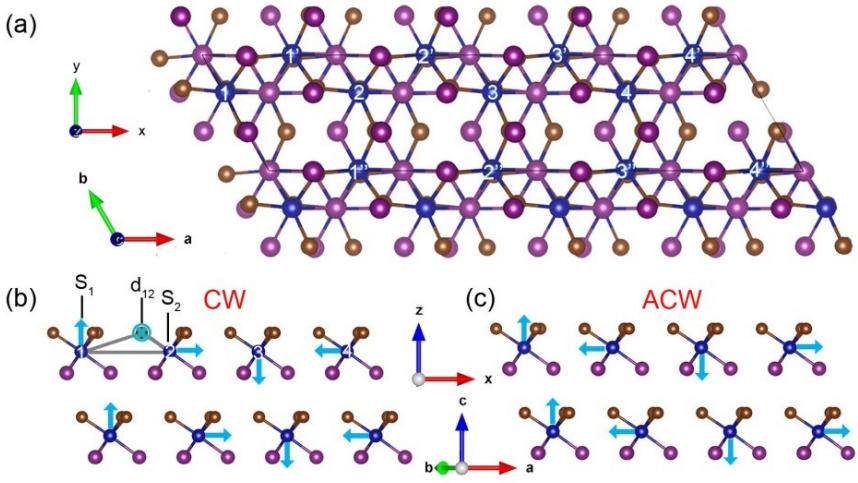

Fig. S1 (a) Top view of the $AC_{up\text{-}up}$ $Cr_2I_3Br_3$ bilayer. Side view of the $AC_{up\text{-}up}$ $Cr_2I_3Br_3$ bilayer with (b) clockwise (CW) and (b) anticlockwise (ACW) spin configurations. The schematic diagram of the DMI is shown in (b), where $\boldsymbol{S}_1$ and $\boldsymbol{S}_2$ represent the spins of the nearest neighbor Cr atoms and $\boldsymbol{d}_{12}$ is the DMI vector.

orientation of the spins, Eq. (S1) can be rewritten as

$$E_3 = \frac{\sqrt{3}}{4} d_{32'}^{y} + E_{other}, \tag{S2}$$

Hence, the energy of atom 3 for clockwise (E_{CW}) and anticlockwise (E_{ACW}) spin configurations shown in Figs. S1(b) and (c) can be expressed as

$$E_{3,\mathrm{CW}} = \frac{\sqrt{3}}{4} d_{32'}^{y} + E_{other}, \tag{S3}$$

$$E_{3,\mathrm{ACW}} = -\frac{\sqrt{3}}{4} d_{32'}^{y} + E_{other}, \tag{S4}$$

Then, the energy difference between the ACW and CW spin configurations (total DMI energy) is

$$\Delta E_{\mathrm{DMI}} = E_{\mathrm{ACW}} - E_{\mathrm{CW}} = 4\sqrt{3} d_{32'}^{-y}. \tag{S5}$$

$d_{32'}^{-y}$ represents the total DMI parameters d_{tot}, which means the strength of DMI concentrated in a atomic layer. Hence, Eq. (S5) can be rewritten as

$$d_{\mathrm{tot}} = \left(E_{\mathrm{ACW}} - E_{\mathrm{CW}}\right)/\mathrm{m}, \tag{S6}$$

where m is a constant of $4\sqrt{3}$ in the $Cr_2I_3X_3$ bilayers.

B. Method for estimating Curie temperature

Two possible magnetic configurations of the Janus $Cr_2I_3X_3$ bilayers are considered: interlayer ferromagnetic (FM) and antiferromagnetic (AFM) coupling, as shown in Fig. 1(g). Based on the 2D Heisenberg model, the interlayer Heisenberg spin Hamiltonian of $Cr_2I_3X_3$ bilayers can be expressed as

$$H_{\perp} = -J_{1\perp}\sum_{\langle ij\rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - J_{2\perp}\sum_{\langle\langle ij\rangle\rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - J_{3\perp}\sum_{\langle\langle\langle ij\rangle\rangle\rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \quad \text{(S7)}$$

where $J_{1\perp}$, $J_{2\perp}$ and $J_{3\perp}$ are the interlayer Cr nearest neighbour, next nearest neighbour and third nearest neighbour exchange interaction parameters. The $AC_{dn\text{-}up}$ model, which is the most stable among several stacking patterns of $Cr_2I_3X_3$ bilayers, is selected to calculate the Curie temperature. The $J_{1\perp}$ (in red), $J_{2\perp}$ (in black) and $J_{3\perp}$ (in green) in $AC_{dn\text{-}up}$ $Cr_2I_3X_3$ bilayer are shown in Fig. S2.

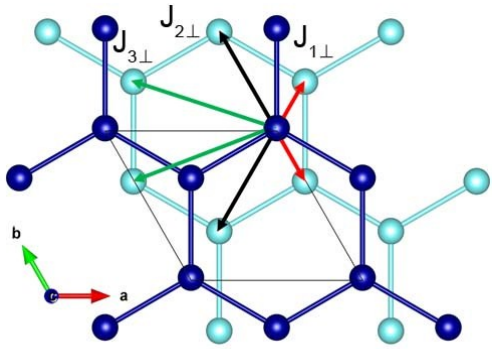

Fig. S2 The interlayer Cr nearest neighbour exchange interaction $J_{1\perp}$ (in red), next nearest neighbour exchange interaction $J_{2\perp}$ (in black) and third nearest neighbour exchange interaction $J_{3\perp}$ (in green) in the $AC_{dn\text{-}up}$ $Cr_2I_3X_3$ bilayers. The dark blue and light blue atoms represent Cr in top and bottom layers.

Using Eq. (S7), the magnetic energy of the two configurations is given by

$$E_{\mathrm{FM/AFM}} = E_0 - \left(\pm 2J_{1\perp} \pm 2J_{2\perp} \pm 2J_{3\perp}\right)\left|\boldsymbol{S}\right|^2, \tag{S8}$$

where $|\boldsymbol{S}|^2 = 3/2$. Neglecting the interlayer next nearest and third nearest neighbors exchange interaction, the exchange energy $E_{\mathrm{ex}}=E_{\mathrm{AFM}}-E_{\mathrm{FM}}$ of $\mathrm{AC_{dn\text{-}up}}$ model of $Cr_2I_3X_3$ bilayers can be expressed as

$$E_{\mathrm{ex}} = E_{\mathrm{AFM}} - E_{\mathrm{FM}} = 4J\left|\boldsymbol{S}\right|^2. \tag{S9}$$

The Curie temperature T_C can be estimated based on the mean field theory[2]

$$T_C = \frac{3J}{2K_B}. \tag{S10}$$

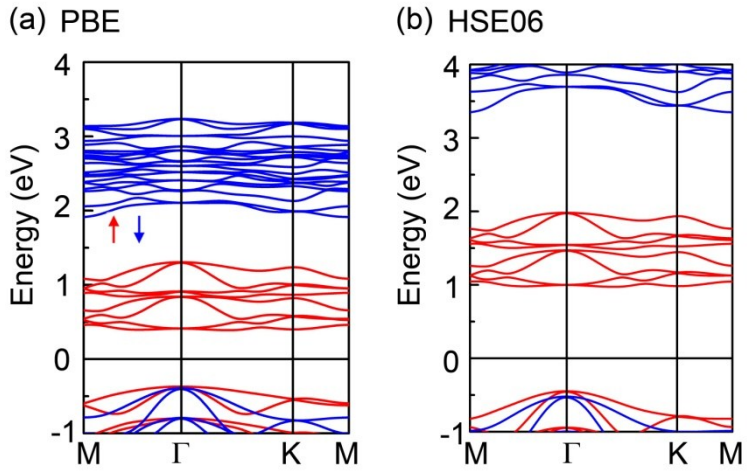

Fig. S3 Band structure of the $AA_{up\text{-}up}$ $Cr_2I_3Br_3$ bilayer calculated by (a) PBE and (b) HSE06 functionals, respectively.

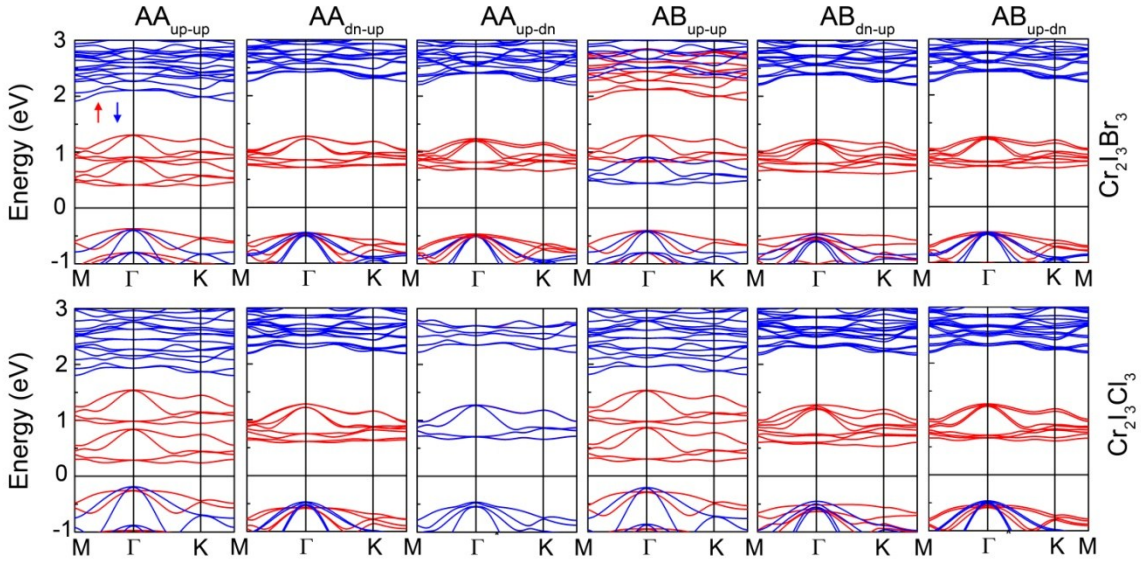

Fig. S4 Band structure of the 2D vdW Janus $Cr_2I_3Br_3$ (upper panel) and $Cr_2I_3Cl_3$ (lower panel) bilayers in $AA_{up\text{-}up}$, $AA_{dn\text{-}up}$, $AA_{up\text{-}dn}$, $AB_{up\text{-}up}$, $AB_{dn\text{-}up}$ and $AB_{up\text{-}dn}$ models. Red and blue lines represent the spin up and spin down channels, respectively.

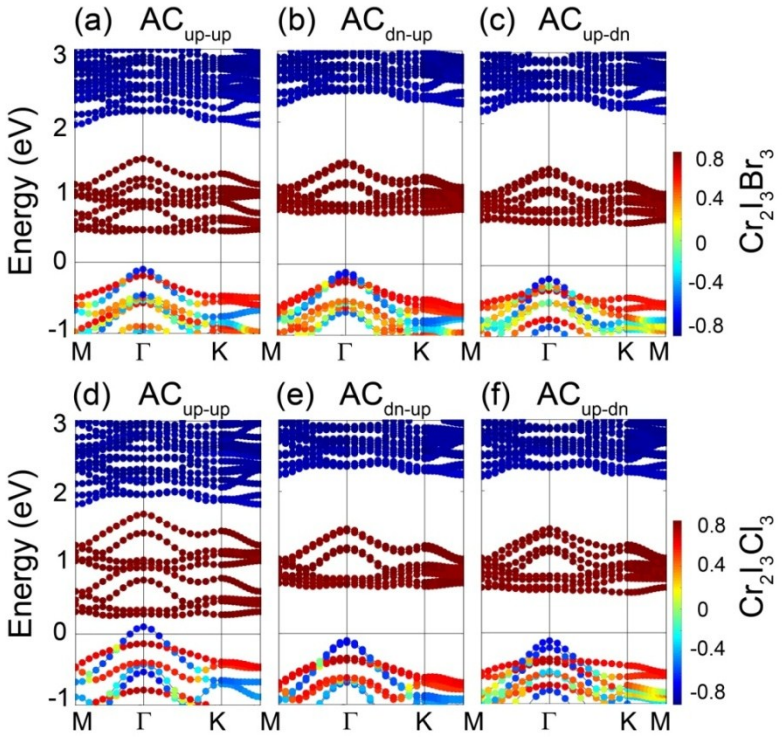

Fig. S5 Band structure of the 2D vdW Janus $Cr_2I_3X_3$ (X = Br, Cl) bilayers in (a)(d) $AC_{up\text{-}up}$, (b)(e) $AC_{dn\text{-}up}$ and (c)(f) $AC_{up\text{-}dn}$ models with SOC. The Fermi energy is set to zero. The spin projection along the z direction is depicted as the color scale.

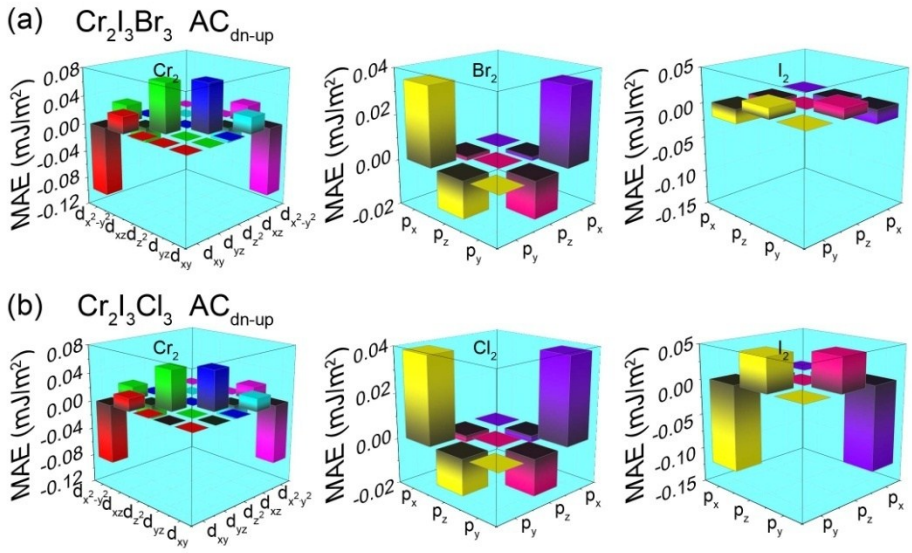

Fig. S6 The orbital resolved MAE of (a) Cr_2, Br_2 and I_2 atoms in the $AC_{dn\text{-}up}$ $Cr_2I_3Br_3$ bilayer and (b) Cr_2, Cl_2 and I_2 atoms in the $AC_{dn\text{-}up}$ $Cr_2I_3Cl_3$ bilayer.

References

1 A. Fert, N. Reyren and V. Cros, *Nat Rev Mater*, 2017, **2**, 17031.

2 L. Webster and J. A. Yan, *Phys. Rev. B*, 2018, **98**, 144411.