Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Table S1. The calculated elements of 2D elastic constants matrix and the in-plane stiffness for FeI_3 monolayer.

	C_{11}	C_{22}	C_{66}	C_{12}	C_{21}	E_x	E_y
	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)
FeI_3	17.22	17.22	11.19	-5.16	-5.16	15.66	15.66

Table S2. The calculated elements of 2D elastic constants matrix and the in-plane stiffness for $FeI_{1.5}X_{1.5}$ (X=Cl, Br) monolayers.

	C_{11}	C_{22}	C_{66}	C_{12}	C_{21}	E_x	E_y
	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)	($J \cdot m^{-2}$)
$FeI_{1.5}Cl_{1.5}$	35.61	35.61	15.70	4.21	4.21	35.11	35.11
$FeI_{1.5}Br_{1.5}$	27.86	27.86	16.45	-5.04	-5.04	26.95	26.95

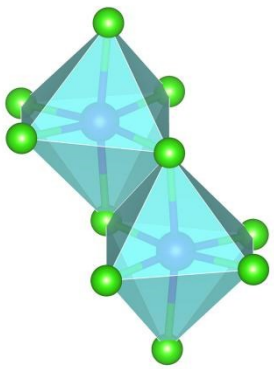

Fig. S1 Structure of the distorted FeI_6 octahedron for FeI_3 monolayer.

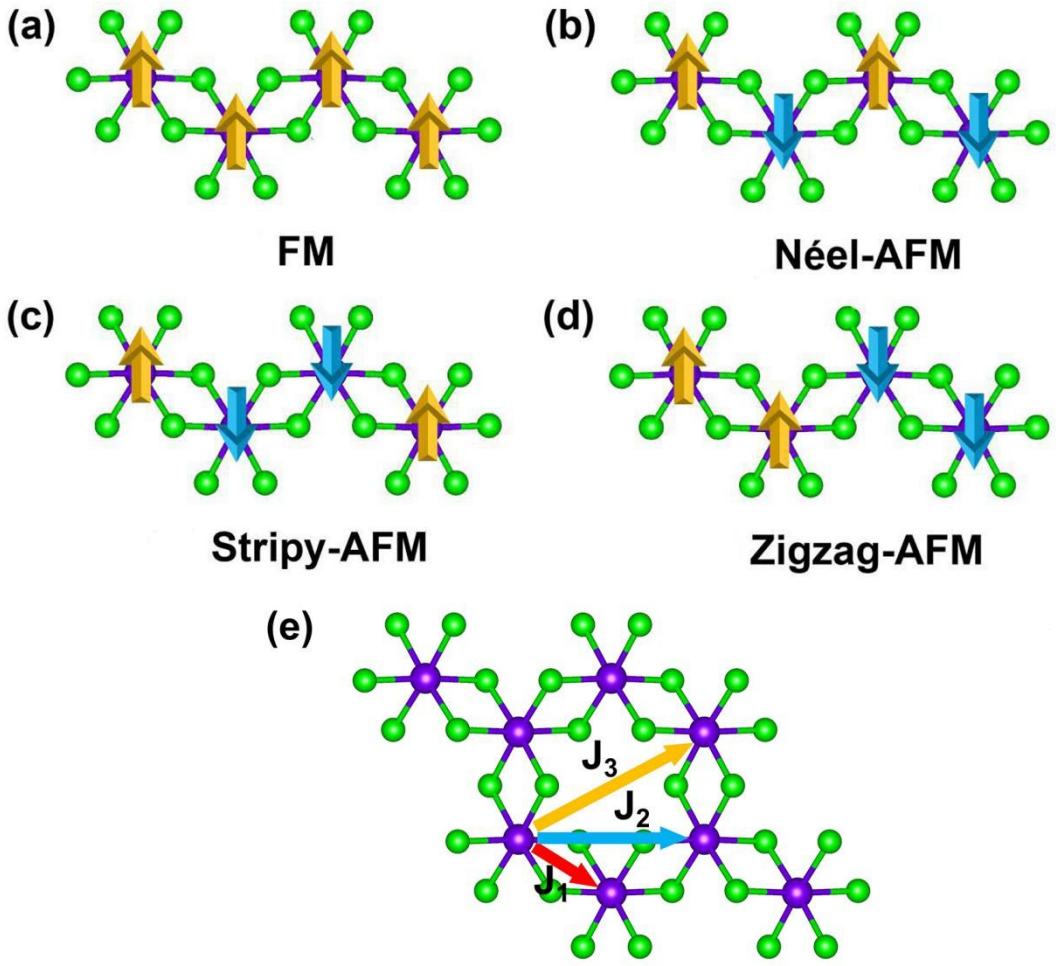

Fig. S2 Four possible magnetic configurations of (a) FM, (b) Néel-AFM, (c) Stripy-AFM, and (d) Zigzag AFM of FeI_3 monolayer, and (e) the nearest (J_1), next-nearest (J_2), next next-nearest (J_3) magnetic coupling parameters, the yellow and blue arrows represent spin-up and spin-down states of the Fe atoms, respectively.

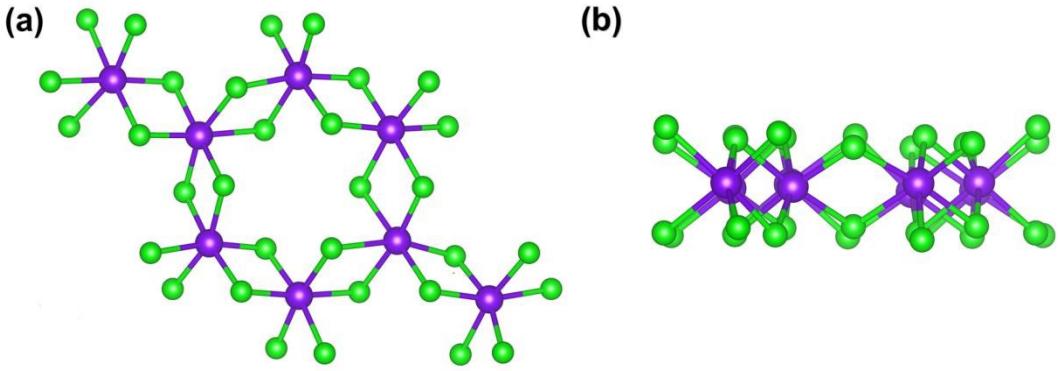

Fig. S3 (a) Top and (b) side views for the snapshots of atomic configurations for FeI_3 monolayer at the end of 5ps AIMD simulation at 300K.

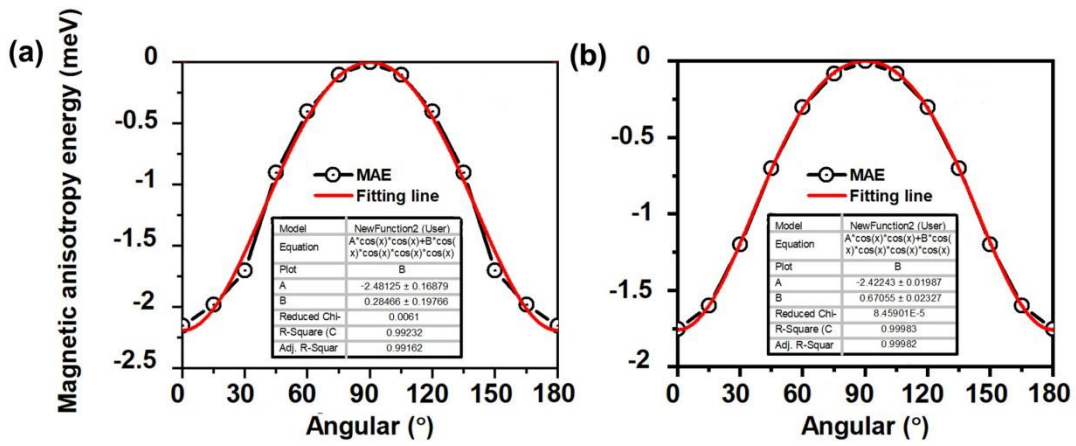

Fig. S4 Magnetic anisotropy energy (MAE) as a function of azimuthal angle for (a) FeI_3 monolayer, and (b) FeI_3 monolayer with 0.1e per unit cell of electron doping. Red solid lines are fitting lines; the inset table lists the standard errors for the fitting slope.

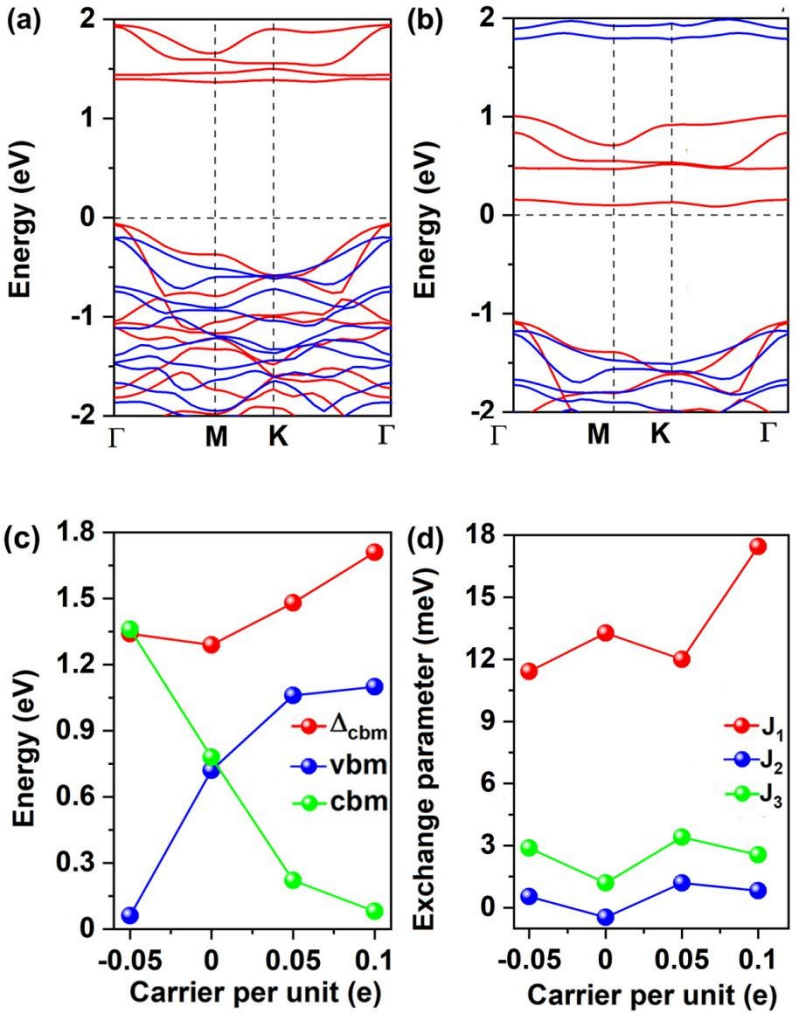

Fig. S5 Spin-resolved electronic band structure of FeI_3 monolayer with (a) -0.05e and (b) 0.1e per unit cell of carrier doping, the Fermi level is set as 0 eV. (c) valence band maximum (VBM), conduction band minimum (CBM), Δ_{cbm} and (d) nearest (J_1), next-nearest (J_2), next next-nearest exchange parameter (J_3) of FeI_3 monolayer with carrier doping ranging from -0.05e to 0.1e per unit cell. Positive and negative values represent the electron and hole doping, respectively.

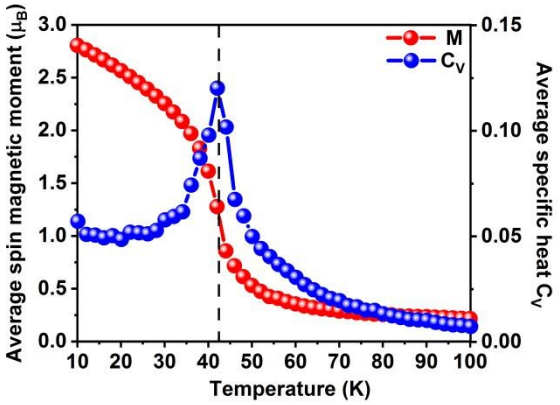

Fig. S6 On-site magnetic moments of Cr atoms and the specific heat C_v as function of temperature based on Heisenberg model for CrI_3 monolayer, the value of J and A are obtained from Ref 21.

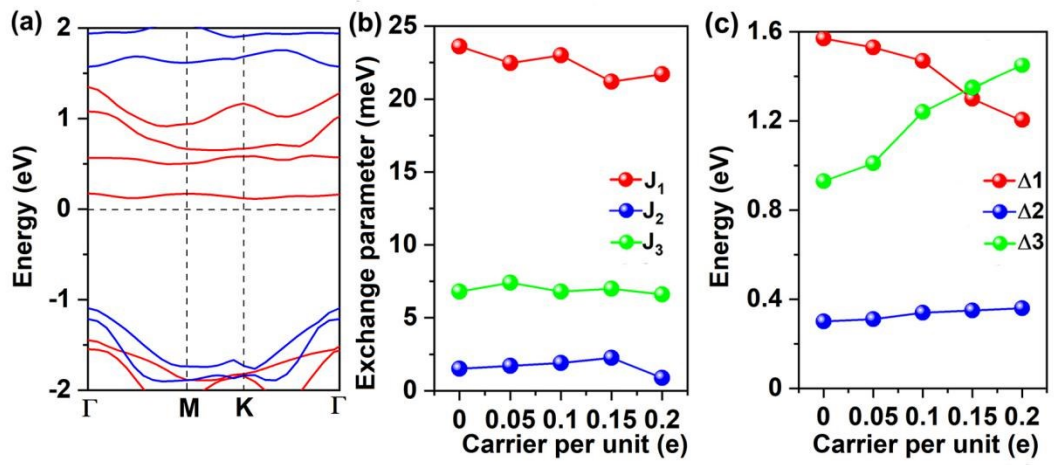

Fig. S7 Spin-resolved electronic band structure of $FeI_{1.5}Cl_{1.5}$ monolayer with (a) 0.2e per unit cell of carrier doping, the Fermi level is set as 0 eV. (b) Nearest (J_1), next-nearest (J_2), next next-nearest exchange parameter (J_3), (c) $\Delta 1$, $\Delta 2$, and $\Delta 3$ of $FeI_{1.5}Cl_{1.5}$ monolayer with carrier doping ranging from 0e to 0.2e per unit cell. Positive and negative values represent the electron and hole doping, respectively.

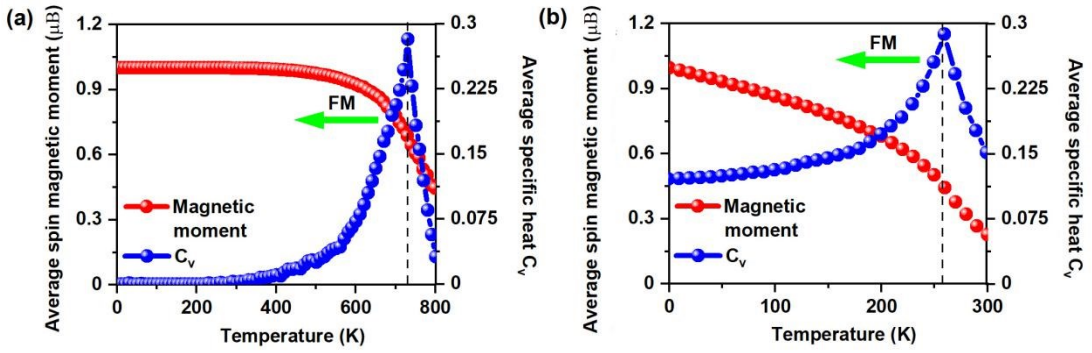

Fig. S8 On-site magnetic moments of Fe atoms and the specific heat C_v as function of temperature based on (a) Ising model and (b) Heisenberg model for $FeI_{1.5}Cl_{1.5}$ monolayer with 0.1e electron doping per unit.