Supporting Information

Room Temperature Spontaneous Valley Polarization in Two-dimensional FeClBr Monolayer

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The Hamiltonian for the spin of Fe atom, which including the DMI, can be expressed as

$$H = -\sum_{\langle i,j \rangle} D_{ij} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j) - J\sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - K\sum_i (S_i^z)^2 \tag{S1}$$

where $\boldsymbol{S}_i$ is the three-dimensional unit vector representing the orientation of the spin of ith Fe atom, $\langle i, j \rangle$ is the nearest-neighbor Fe atom pairs. The three magnetic interaction terms are DMI, Heisenberg isotropic exchange and easy axis single ion anisotropy, respectively. The result is shown in Fig. S1. It can be seen that the orientation of magnetization is parallel to the z-axis direction, which is consistent with the magnetocrystalline anisotropy.

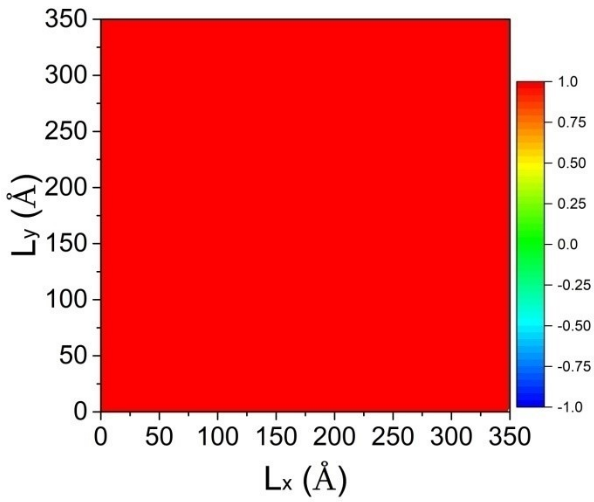

Fig. S1 Magnetic states for FeClBr monolayer in real-space.

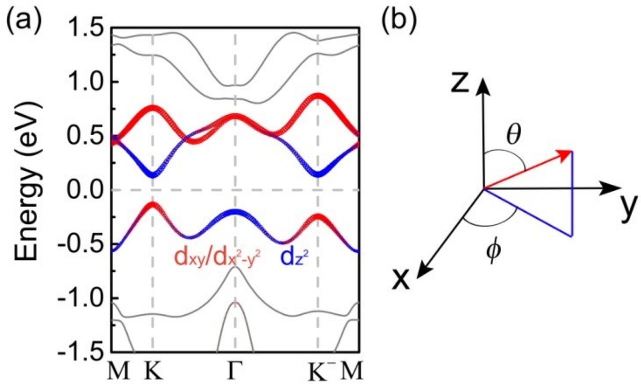

Fig. S2(a) The orbital projected band of FeClBr monolayer, the red and blue represent the contribution of $d_{x^2-y^2}/d_{xy}$ and d_{z^2}. (b) Schematic of polar angles of spin orientation.

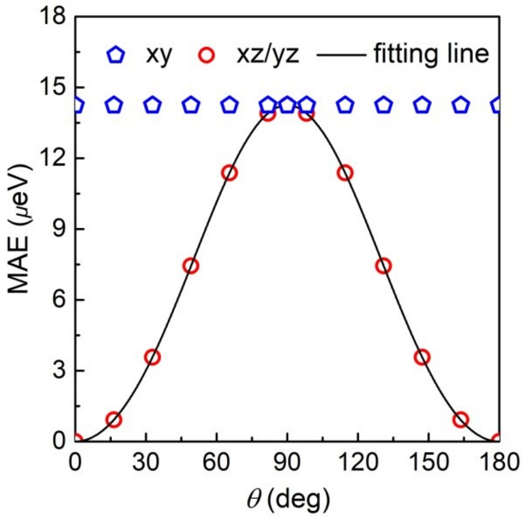

Fig. S3 Angular dependence MAE of FeClBr monolayer with the direction of magnetization lying on three different planes: *xy*, *xz* and *yz*.