Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary materials for

Enabling triferroics coupling in breathing kagome lattice Nb_3X_8 (X = Cl, Br, I) monolayers

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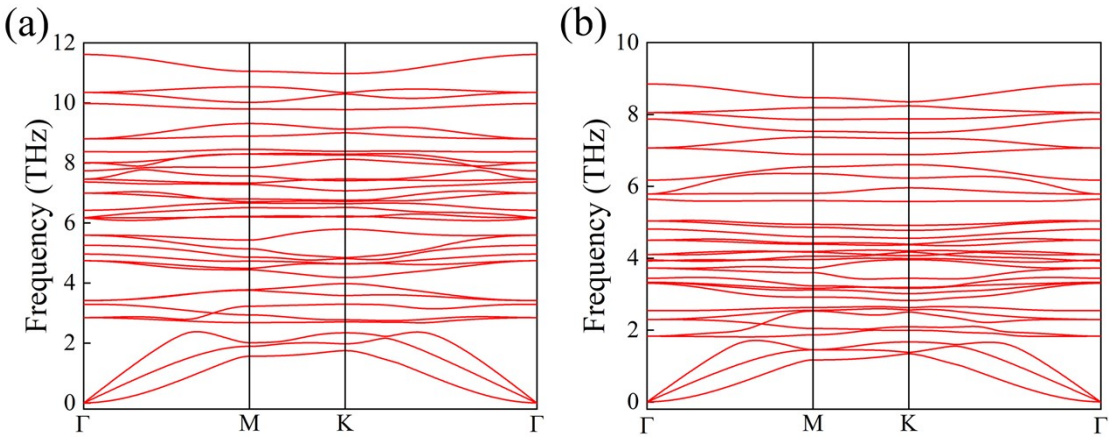

Fig. S1 Phonon dispersion spectrums for the State I and State II of (a) Nb_3Cl_8 and (b) Nb_3Br_8 monolayers.

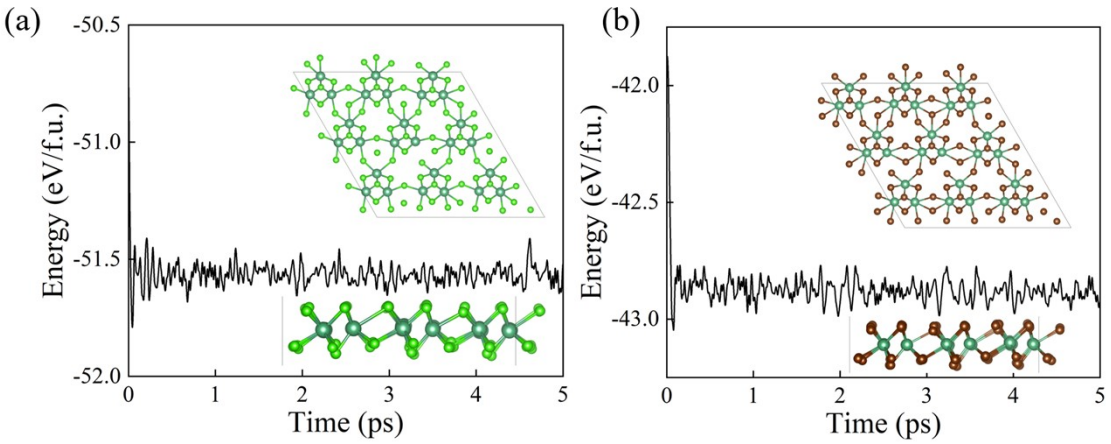

Fig. S2 AIMD simulations of (a) Nb_3Cl_8 and (b) Nb_3Br_8 monolayers. The top and side views of final structure are in the picture.

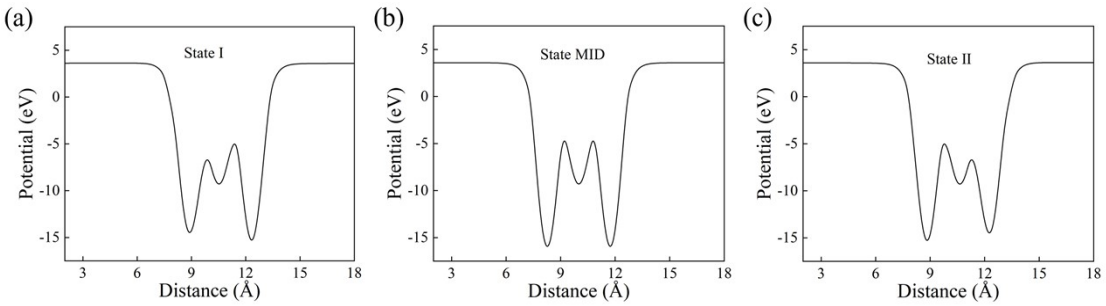

Fig. S3 Work functions along the *z* axis of ferroelectric and paraelectric phases: (a) State I; (b) State MID; (c) State II.

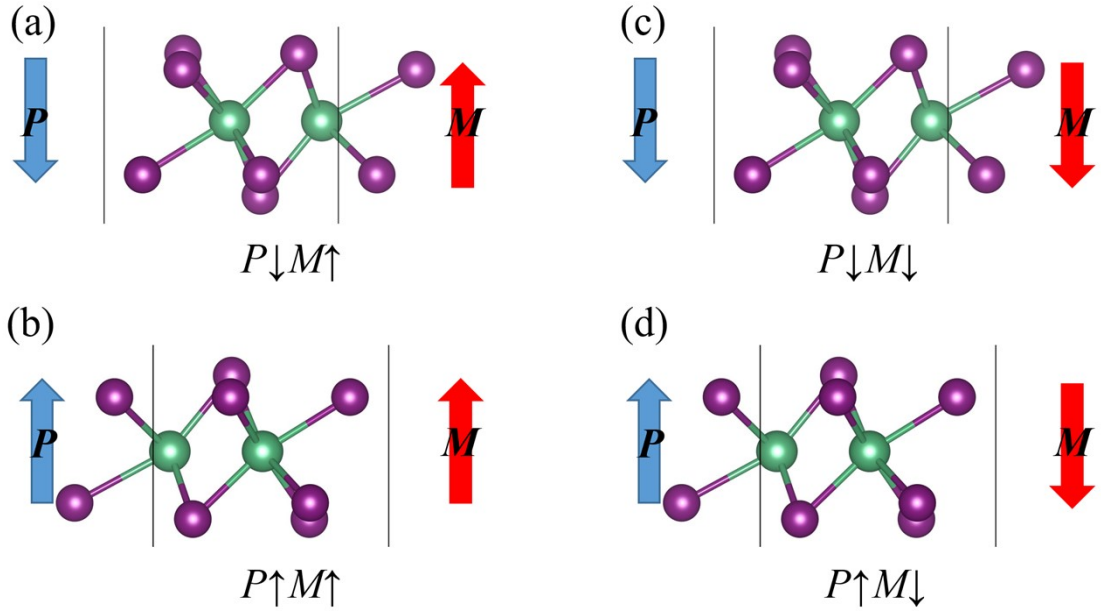

Fig. S4 Four different FE/FM configurations of the Nb_3I_8 monolayer: (a) $P\downarrow M\uparrow$ configuration; (b) $P\uparrow M\uparrow$ configuration; (c) $P\downarrow M\downarrow$ configuration; (d) $P\uparrow M\downarrow$ configuration. Blue and red arrows represent FE polarization and spin polarization directions, respectively.

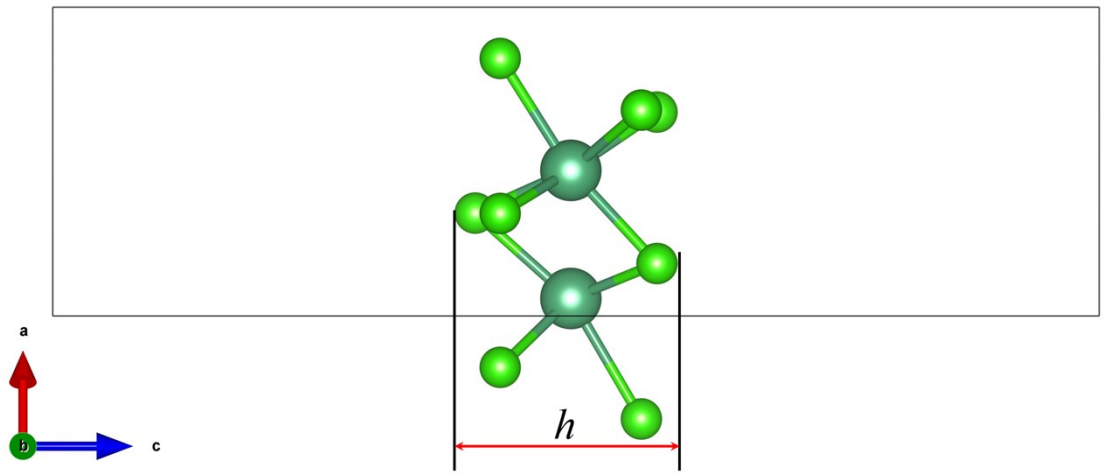

Fig. S5 Side view of the Nb_3Cl_8 monolayer with periodic boundaries indicated by a black rectangle. *h* is defined as the thickness of the monolayer and the vacuum region of 20 Å along *c* direction is

introduced in our calculations.

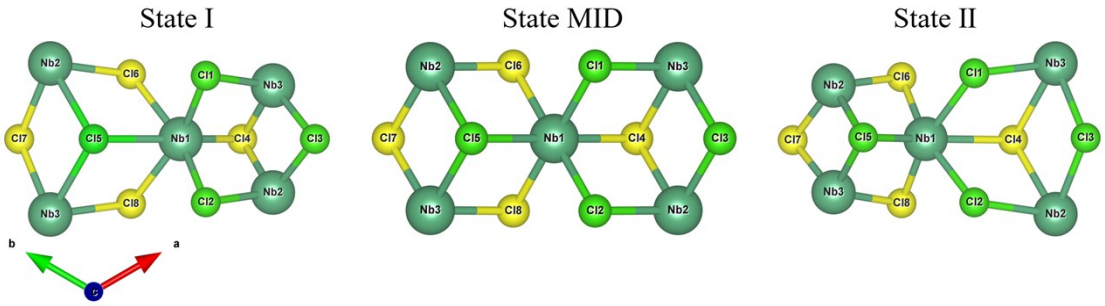

Fig. S6 Top views of the Nb_3Cl_8 monolayer for the State I, Stats MID and State II. The light green and yellow balls represent the Cl atoms at upper and bottom layers, respectively.

Table S1. Charge analysis of the Nb_3Cl_8 monolayer by Hirshfeld method. The unit is *e*.

Atoms	State I	State MID	State II
Cl_1,Cl_2, Cl_3	-0.1089	-0.1367	-0.1587
Cl_5	-0.1077	-0.0843	-0.0266
Cl_4	-0.0226	-0.0843	-0.1077
Cl_6,Cl_7,Cl_8	-0.1587	-0.1367	-0.1089

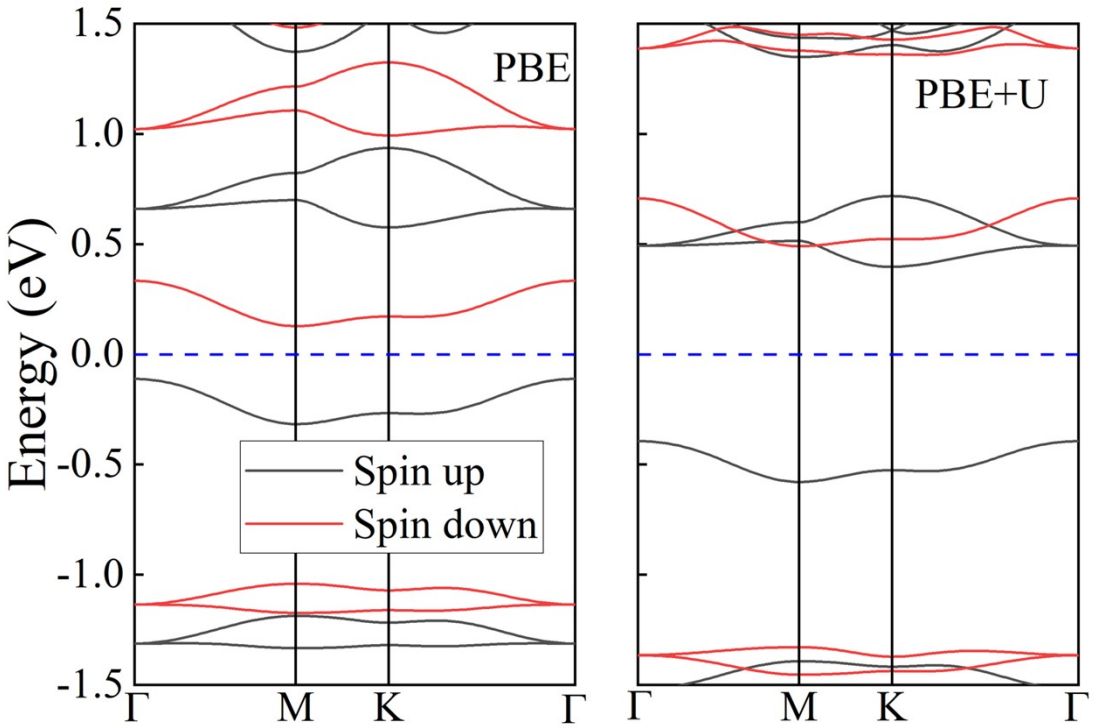

Fig. S7 Band structures of the Nb_3Cl_8 monolayers by PBE or PBE+U calculations.