Supporting Information

First-principles prediction of a room-temperature ferromagnetic and ferroelastic 2D multiferroic MnNX (X=F, Cl, Br, I)

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Table S1 The calculated elastic constants C_{ij} (N/m) and total energy (eV) of FM and collinear AFMi (i=1, 2, 3, 4) configurations for MnNX (X= F, Cl, Br, I) monolayers using a 2×2×1 supercell.

Monolayer	C_{11}	C_{12}	C_{22}	C_{44}	E_{FM}	E_{AFM1}	E_{AFM2}	E_{AFM3}	E_{AFM4}
MnNF	183	25	162	51	0	1.02	0.65	1.10	1.35
MnNCl	172	24	131	46	0	0.93	0.69	1.05	1.37
MnNBr	155	17	108	43	0	1.12	0.80	1.16	1.45
MnNI	145	18	81	39	0	0.73	0.62	0.96	0.95

Table S2 The structural parameters (Å) of initial state (*IS*) and transition state (*TS*), ferroelastic energy barriers (meV/atom) and reversible ferroelastic strain.

Monolayer	Initial state		Transition state		Barrier	Strain
	a_{IS}	b_{TS}	a_{TS}	b_{TS}		
MnNF	3.88	2.93	3.53	3.53	336	32.3%
MnNCl	3.86	3.15	3.64	3.64	145	22.6%
MnNBr	3.88	3.31	3.70	3.70	87	17.2%
MnNI	3.90	3.52	3.75	3.75	31	10.8%

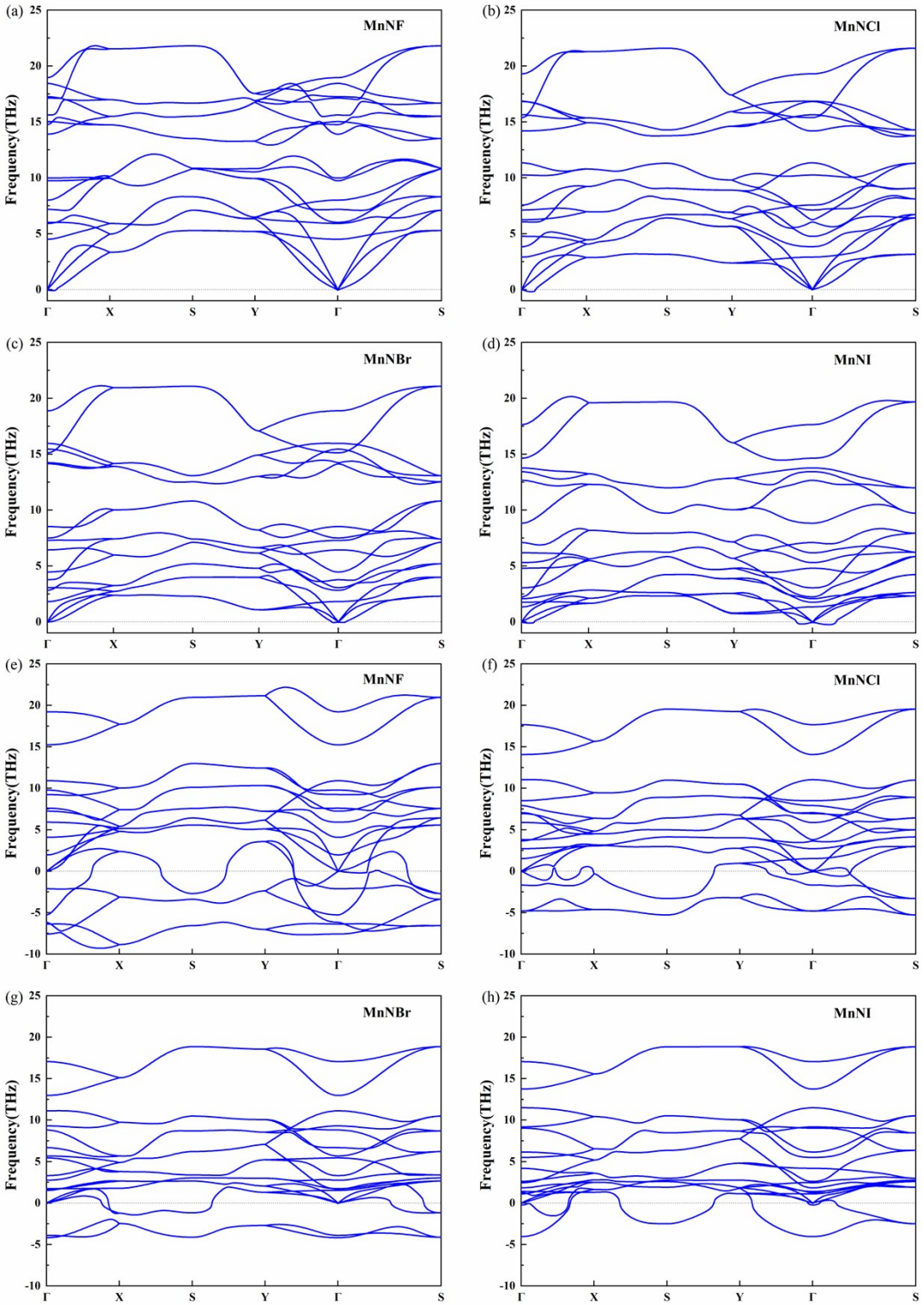

Fig. S1 The full phonon dispersions of (a)-(d) initial ferroelastic states and (e)-(h) transition states for MnNX monolayers.

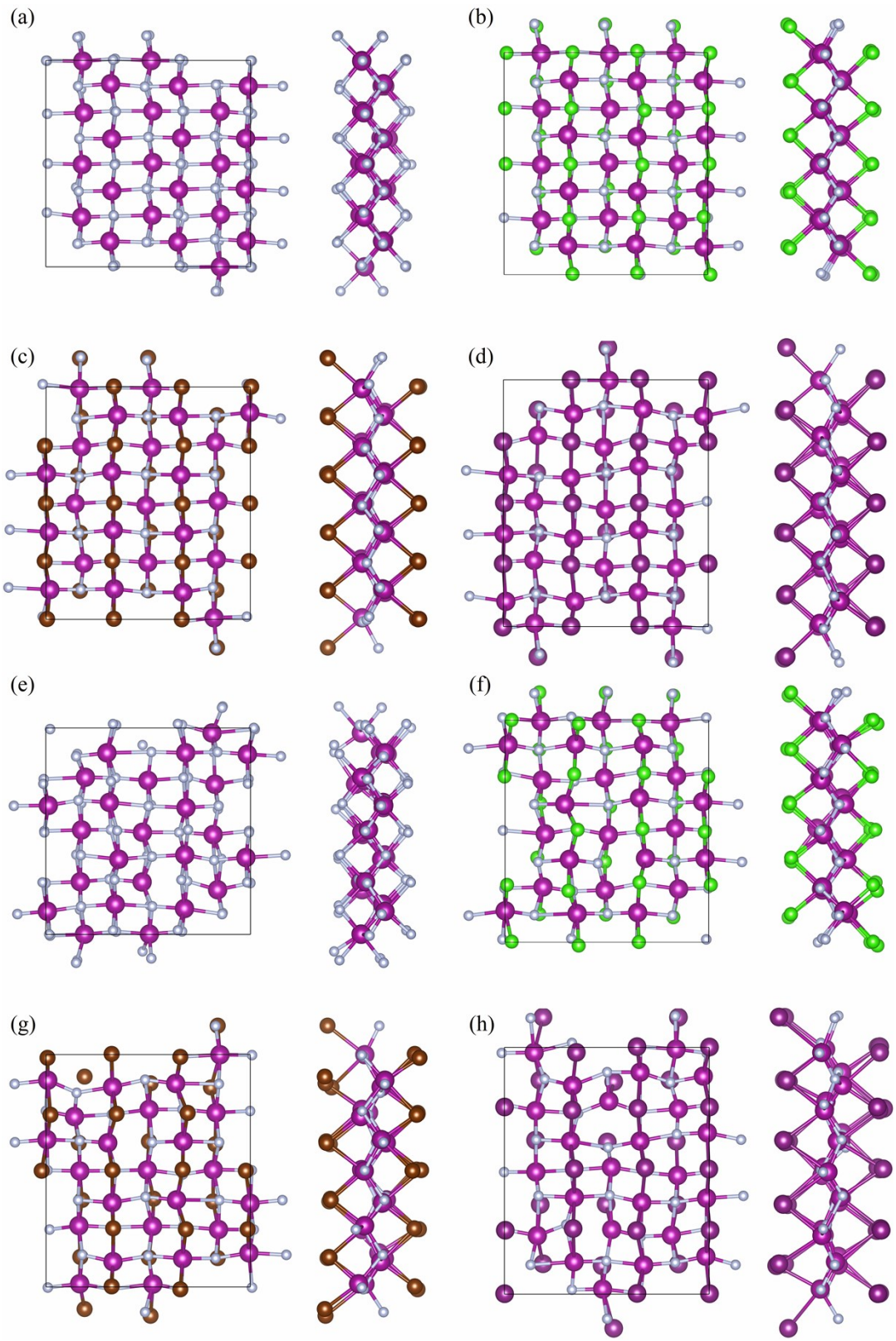

Fig. S2 Snapshots of MnNF, MnNCl, MnNBr, and MnNI monolayers after 10 ps AIMD simulations (a)-(d) at 300K, and (e)-(h) at 1000K.

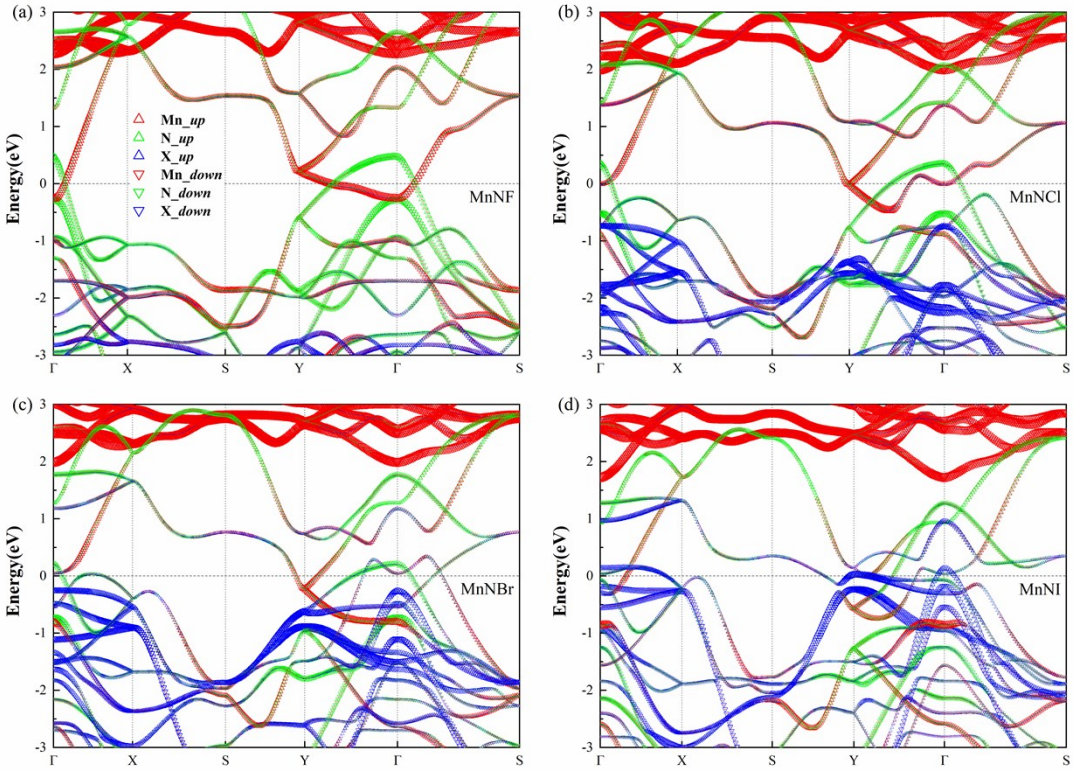

Fig. S3 The projected electronic energy band structure of MnNX monolayers calculated with the GGA+U functional (U=3.9 eV).

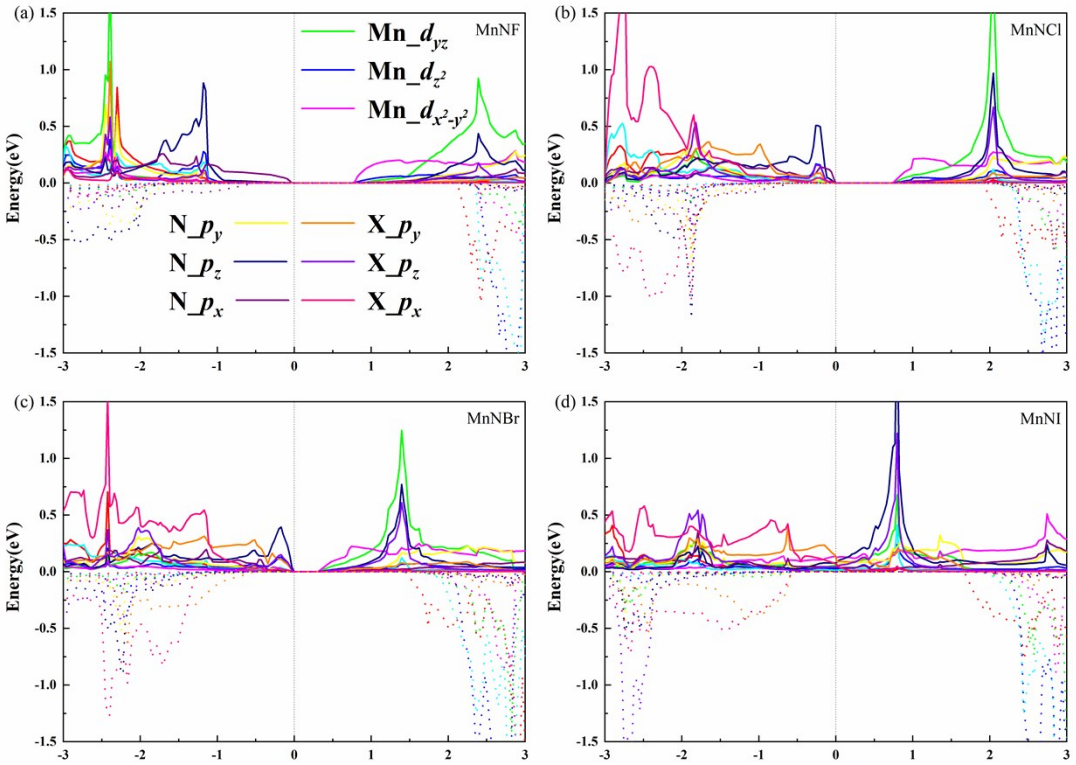

Fig. S4 The projected density of states (DOS) of MnNX monolayers calculated with the HSE06 functional.

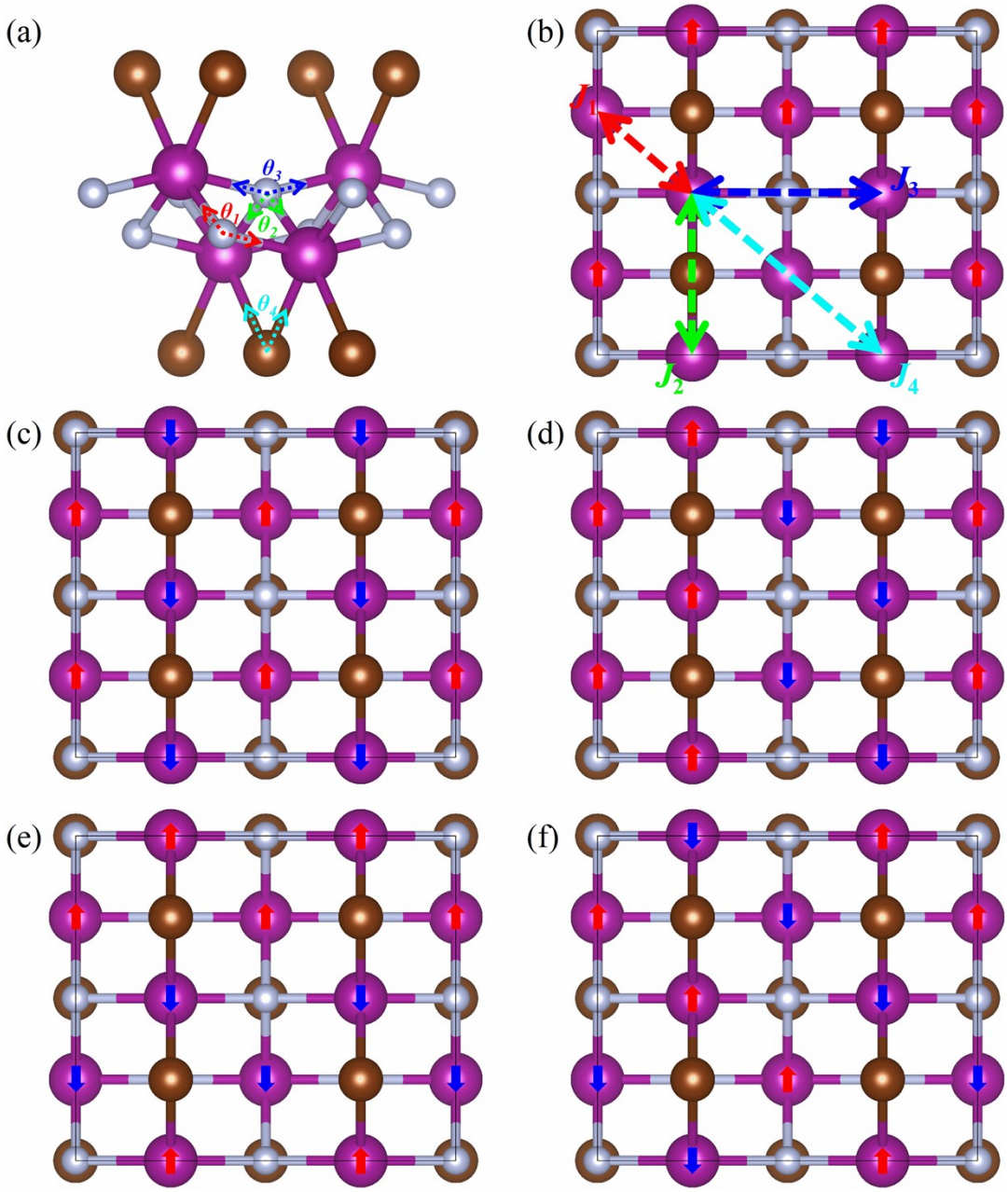

Fig. S5 (a) The schematic structures of Mn−N(Br)−Mn bridges, with the four bond angles labeled as θ_1, θ_2, θ_3 and θ_4. (b) The top view shows J_1, J_2, J_3, and J_4 representing the first, second, third and fourth nearest couplings of Mn, respectively. The initial magnetic configurations of FM and collinear AFMi (i=1, 2, 3, 4) for MnNX monolayers within a 2×2×1 supercell are shown in (b)-(f).

We employ the Ising model and the anisotropic Heisenberg model to predict the Curie temperature (T_C). Our Ising model simulation results give quite high T_C values from 486 to 758 K, however, the anisotropy is greatly overestimated which results in the T_C value being about twice the anisotropic Heisenberg result. We calculate the spin exchange parameters according to the total energies of the five magnetic configurations in Table S1 which are schematically shown in the Fig. S5. The magnetic energy contributions of these magnetic

configurations in each magnetic unit cell is written as:

$$E_0 - 9J_1 - 4.5J_2 - 4.5J_3 - 9J_4 - 2.25A = E_{FM},$$
$$E_0 + 9J_1 - 4.5J_2 - 4.5J_3 - 9J_4 - 2.25A = E_{AFM1},$$
$$E_0 - 0J_1 + 4.5J_2 - 4.5J_3 + 9J_4 - 2.25A = E_{AFM2},$$
$$E_0 - 0J_1 - 4.5J_2 + 4.5J_3 + 9J_4 - 2.25A = E_{AFM3},$$
$$E_0 - 0J_1 + 4.5J_2 + 4.5J_3 - 9J_4 - 2.25A = E_{AFM4},$$

where E_0 is the energy of the nonmagnetic state, A is the anisotropy energy parameter, J_1, J_2, J_2 and J_4 represent the first-, second-, third-, and fourth- nearest couplings, respectively, as shown in Fig. S5(a).