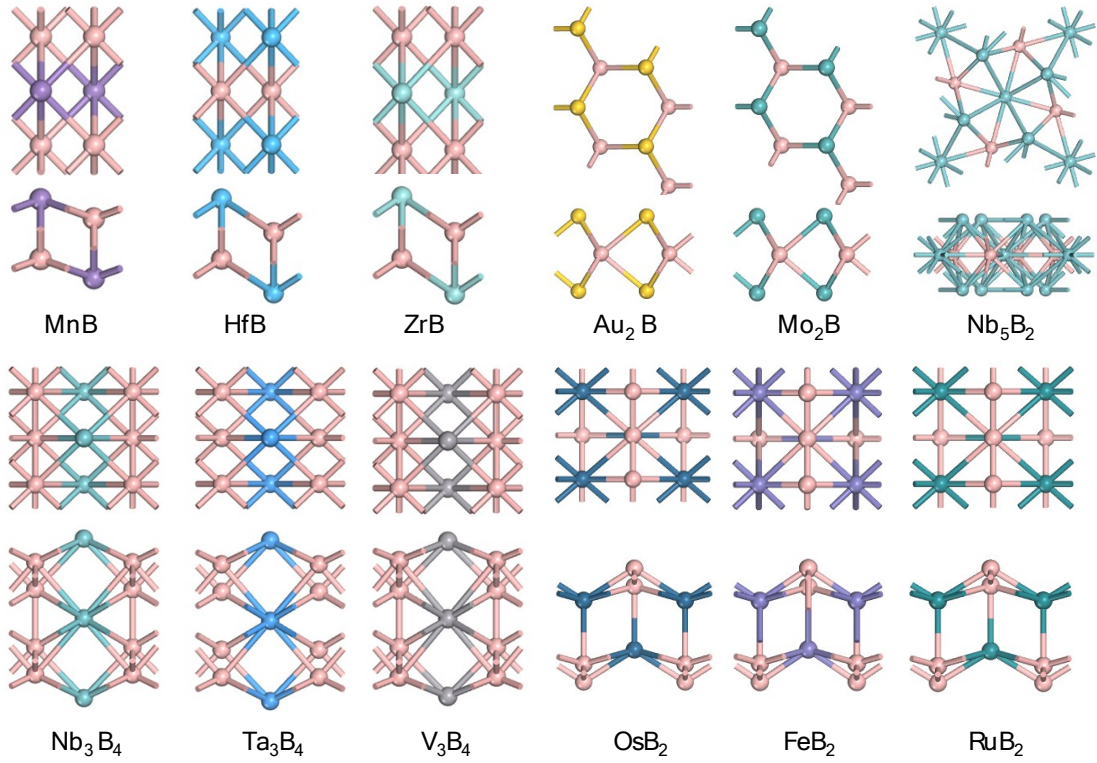**Table S1.** Tests of different correlation (U) from 3 to 5 eV for Mn_2AlB_2, Mn_4B_4. The front and the last column are the lattice parameters from simulations and experiments, respectively. The selection criterion is that the difference of lattice parameters between simulation and experiment is less than 0.1 Å for both systems.
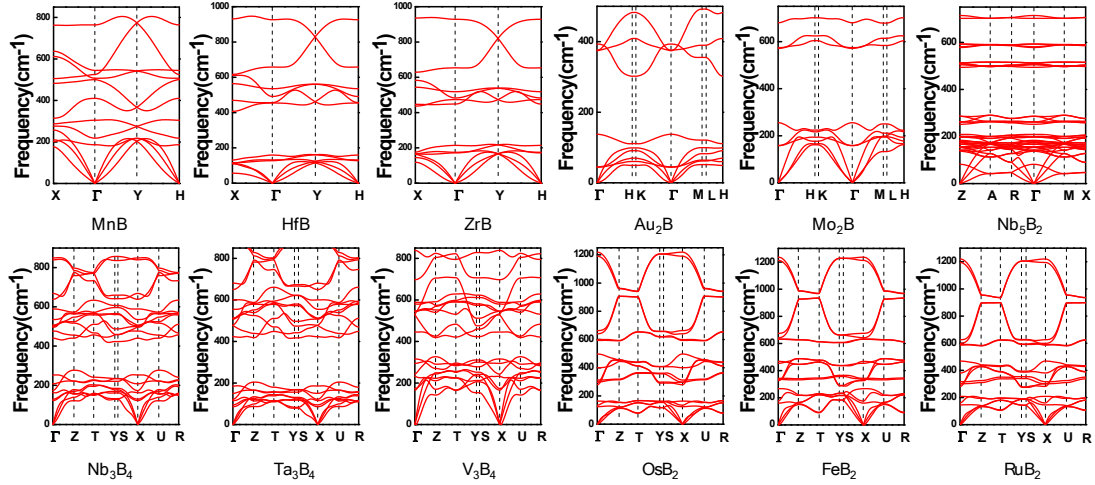
	U	0	3	3.5	4	4.5	5	Experiment
	a / Å	2.895	2.894	2.898	2.900	2.931	2.917	2.918
Mn_2AlB_2	b / Å	11.073	11.090	11.081	11.074	11.248	11.037	11.038
	c / Å	2.831	2.836	2.838	2.839	2.855	2.893	2.893
	a / Å	5.447	5.583	5.827	5.941	6.205	6.273	5.56
Mn_4B_4	b / Å	2.981	2.970	2.960	2.980	3.056	3.071	2.977
	c / Å	4.119	4.146	4.267	4.340	4.499	4.569	4.145

Table S2. The cohesive energies, magnetic moments M (PBE), bond lengths and slice thicknesses of MnB, HfB, ZrB, Au_2B, Mo_2B, Nb_5B_2, Nb_3B_4, Ta_3B_4, V_3B_4, OsB_2, FeB_2, and RuB_2.

	MnB	HfB	ZrB	Au_2B	Mo_2B	Nb_5B_2	Nb_3B_4	Ta_3B_4	V_3B_4	OsB_2	FeB_2	RuB_2
Cohesive energy / eV	4.8	6.69	6.67	3.30	5.58	6.83	7.01	7.37	6.31	6.64	5.57	6.26
Slice thickness / Å	0.125	0.135	0.138	0.14	0.138	0.155	0.253	0.252	0.237	0.222	0.219	0.219
Bond length / Å	2.119	2.4	2.435	2.271	2.176	2.439	2.333	2.334	2.159	2.085	1.912	2.056
	2.176	2.439	2.484	2,273			2.362	2.344	2.199	2.38	2.046	2.179
							2.425	2.406	2.291	2.203	2.238	2.389
							2.461	2.459	2.363			
M / μ_B	4.71	0	-0.01	0	0	0	0	0.02	2.30	0	0	0

Figure S1. (a) The structures and (b) calculated phonon band structures for MnB, HfB, ZrB, Au_2B, Mo_2B, Nb_5B_2, Nb_3B_4, Ta_3B_4, V_3B_4, OsB_2, FeB_2, and RuB_2. Upper plots: top view, lower plots: side view.
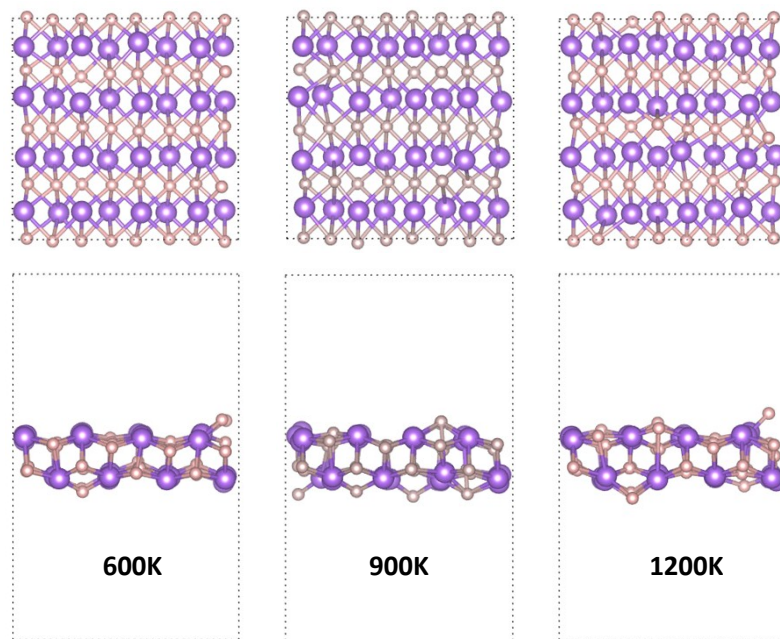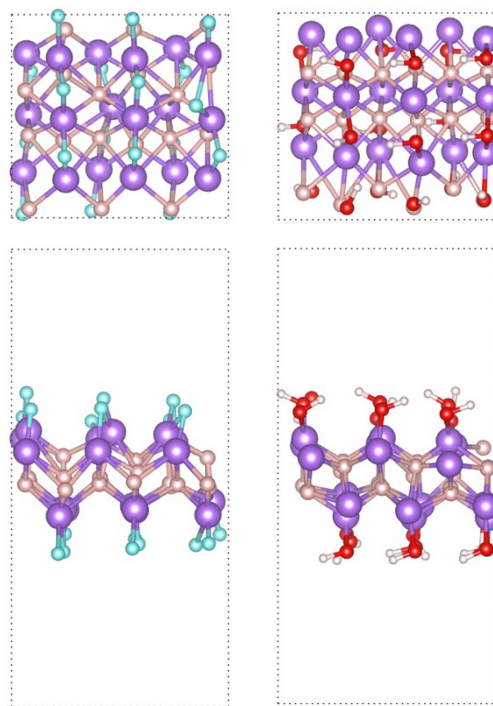
Figure S2. (a) Structure snapshots of (4×4×1) supercell for MnB sheet during AIMD simulations at 600K, 900K and 1200K, respectively. (b) Structure snapshots of (3×3×1) supercell for MnBF and MnBOH sheets during AIMD at 600K after 20 ps. Upper plots: top view, lower plots: side view.
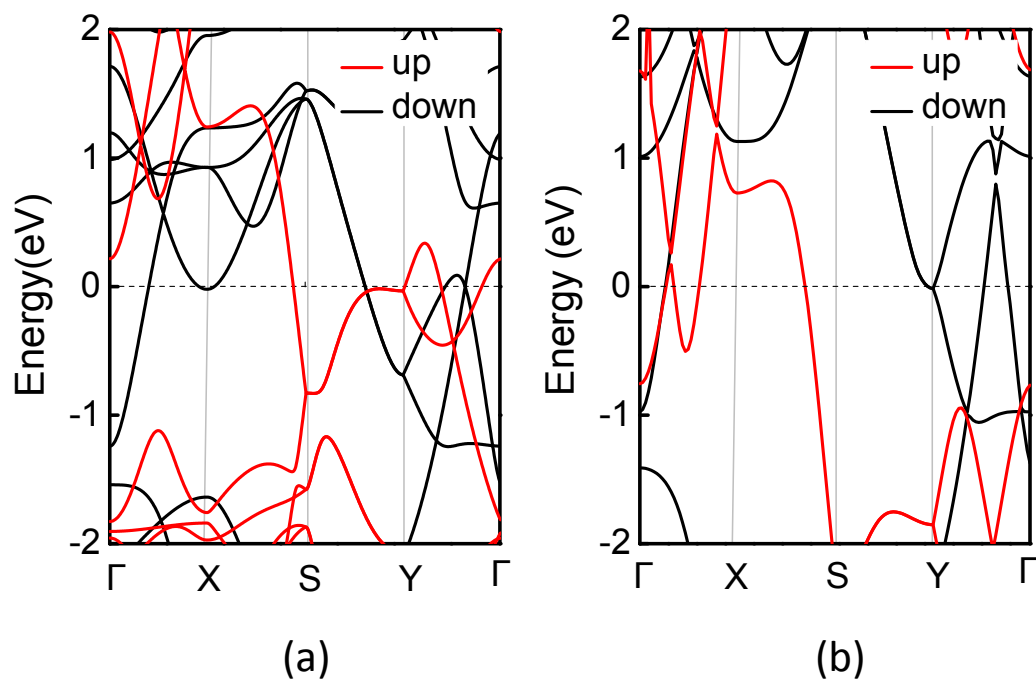
Figure S3. Spin-polarized band structure of MnB sheet calculated from (a) GGA-PBE and (b) hybrid functional (HSE06), and the Fermi levels are all set to zero.

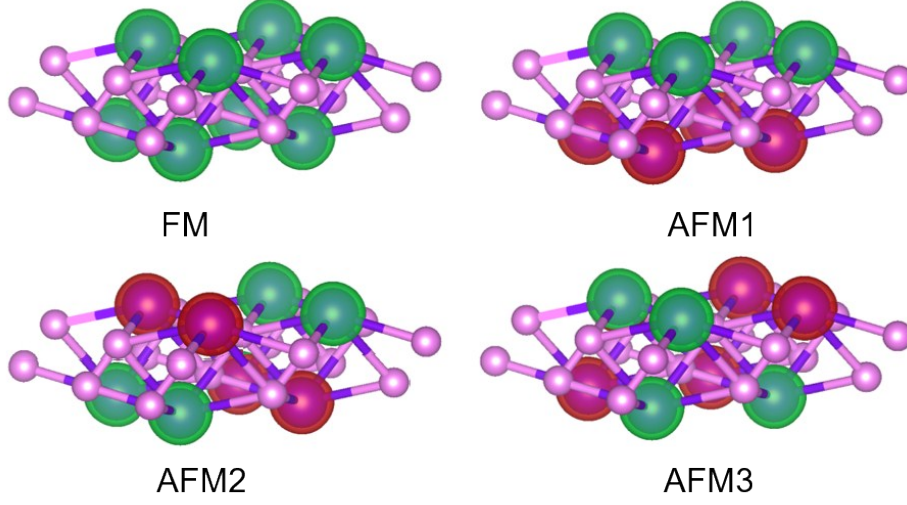**S4 Estimation of J_1, J_2, and J_3**

Figure S4. The 2×2 supercell for MnB sheet in FM and three AFM magnetic configurations (the green and red atoms represent the Mn atoms in spin-up and spin-down states, respectively).

The Hamiltonian of classical Heisenberg model can be written as equation (1)

$$H = -\sum_{i,j} J_1 M_i M_j - \sum_{k,l} J_2 M_k M_l - \sum_{p,q} J_3 M_p M_q$$

(1)

where J_1, J_2, and J_3 are the first, second and third nearest-neighbor exchange parameters, respectively. From the Figure S4, we can estimate J1, J2 and J3 from the following equations.

$$E(FM) = -(8J_1 + 8J_2 + 8J_3)M^2$$

$$E(AFM1) = -(-8J_1 + 8J_2 + 8J_3)M^2$$

$$E(AFM2) = -(-8J_2 + 8J_3)M^2$$

$$E(AFM4) = 0$$

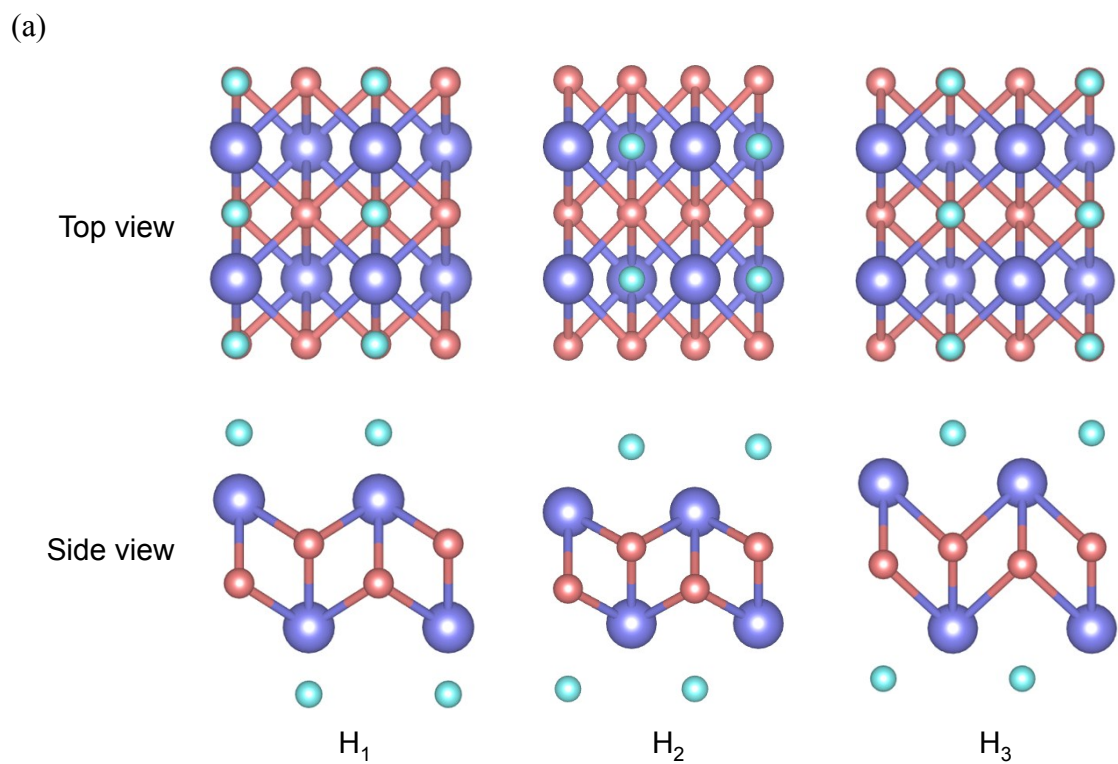$$\Delta E_1 = E(AFM1) - E(FM) = 16J_1 M^2$$

$$\Delta E_2 = E(AFM2) - E(FM) = (8J_1 + 16J_2)M^2$$

$$\Delta E_3 = E(AFM3) - E(FM) = (8J_1 + 8J_2 + 8J_3)M^2$$

$$J_1 = \frac{\Delta E_1}{16M^2} = 3.1 meV$$

$$J_2 = \frac{2\Delta E_2 - \Delta E_1}{32M^2} = 1.4 meV$$

$$J_3 = \frac{-\Delta E_1 - 2\Delta E_2 + 4\Delta E_3}{32M^2} = -2.3 meV$$

(a)

Top view

Side view

H_1 H_2 H_3

(b)

Energy / eV	MnBF	MnB(OH)
H_1	-39.547	-51.296
H_2	-40.162	-52.112
H_3	-40.800	-52.324

Figure S5. Optimized geometries of freestanding MnB sheet's functionalized structural forms (take MnBF for example). (a) H_1 –H_2 are the top and side view of 2×2 supercell MnBX (X=F or OH) structural forms. Upper plots: top view, lower plots: side view. (b) The energy of the unit cell of MnBX in three forms respectively.
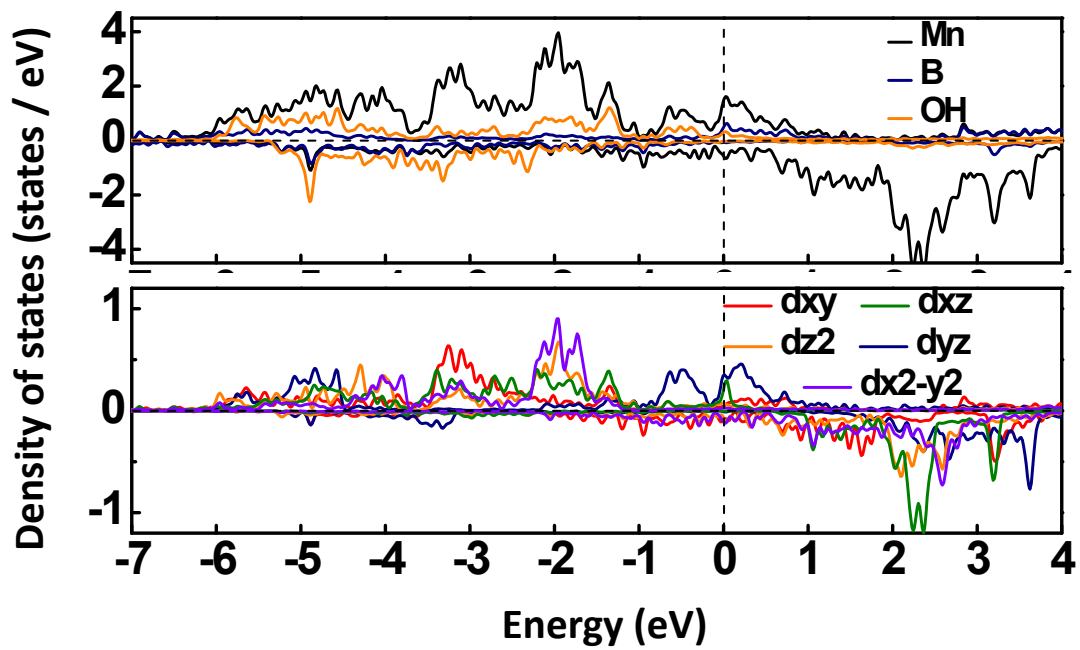
Figure S6. Atom-projected and orbital-projected density of states for MnOH sheet, and the Fermi levels are all set to zero.