Supporting information

TM_2B_6: A New designed Ferromagnetic 2D Metal-Boride with High Curie Temperature

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Table S1. The distance (h) between transition meal layers, TM-B bond lengths (TM-B-1 is the bond length between TM with the six boron atoms in the near layer, TM-B-2 is the bond length between TM with the four boron atoms in the other layer), B-B bond length and electrons transferred from TM atoms to boron framework (TE).

TM	h (Å)	TM-B-1 (Å)	TM-B-2 (Å)	B-B (Å)	TE(e)
Ti	2.32	2.35,2.52	4.25,4.35	1.65,1.67,1.70	1.19,089
V	2.28	2.36,2.45,2.47	4.15,4.20	1.68,1.69	0.54
Cr	2.14	2.35,2.46	4.02,4.07	1.68,1.70	0.49
Mn	1.92	2.41,2.48	3.82,3.89	1.70,1.72	0.54,0.53
Fe	0.75	2.20,2.29	2.42,2.50	1.75,1.79	0.48,0.47
Co	0.79	2.00,2.15,2.17	2.25,2.35	1.73,1.75,1.76	0.17,0.15
Ni	0.81	2.00,2.20	2.34,2.38	1.70,1.74,1.75	0.06,0.08

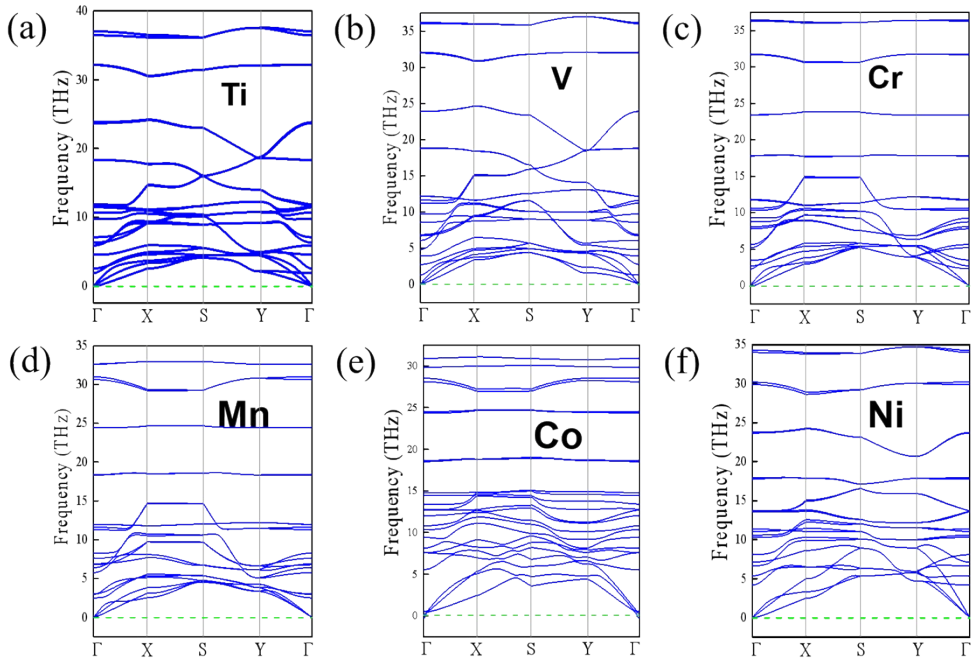

Figure S1. The phonon spectra of TM_2B_6, TM=Ti, V,Cr, Mn,Co and Ni.

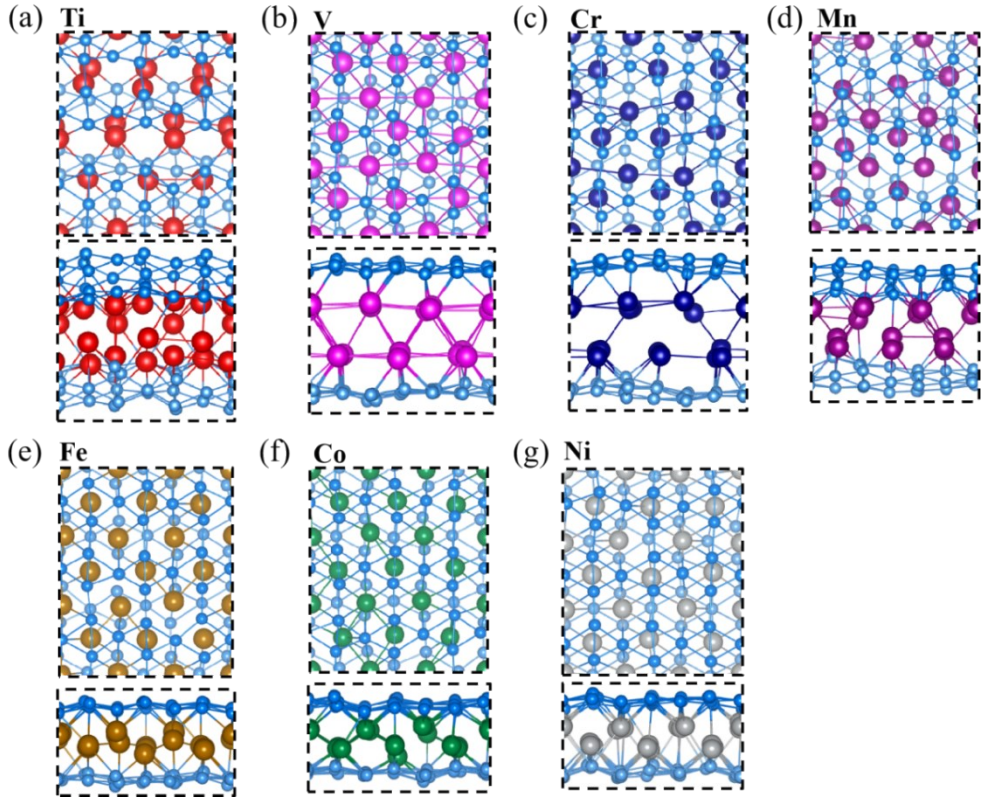

Figure S2. Snapshots of the TM_2B_6 structures at 1000 K at the end of 6 ps AIMD simulations.

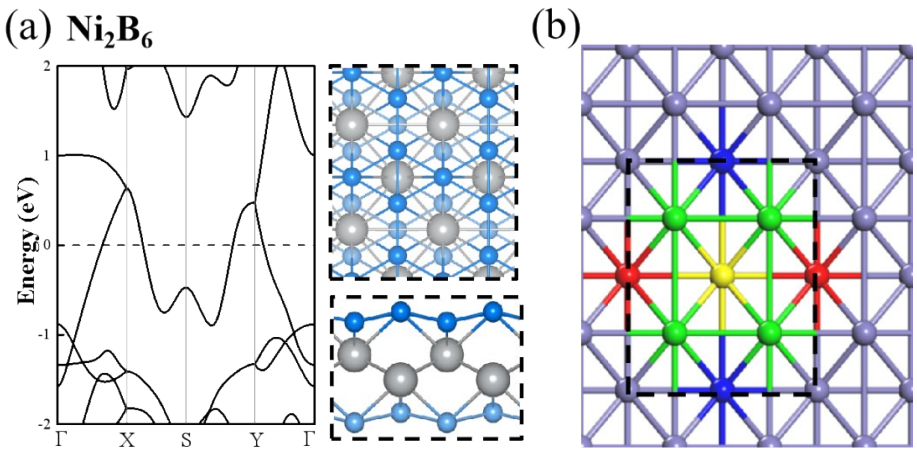

Figure S3. (a) Band structures and spin densities of Ni_2B_6, yellow and blue colors represent spin up and spin down electrons, respectively. (b) The schematic of FM Fe_2B_6 (Co_2B_6). The green, red and blue atoms represent the nearest, second-nearest, and third-nearest Fe (Co) atoms, respectively.

Table S2. The exchange parameters J_1, J_2 and J_3 of ferromagnetic Fe_2B_6, Co_2B_6 and H functionalized Fe_2B_6. The unit is meV.

	J_1	J_2	J_3
Fe_2B_6	39.7	22.9	4.1
Co_2B_6	77.0	89.0	34.9
H-Fe_2B_6(η=1/3)	29.6	6.1	6.3
H-Fe_2B_6(η=1/2)	7.6	12.7	3.3

Table S3. Summary of the relative energies along different directions in meV for TM_2B_6 (TM=Fe, Co) and H functionalized Fe_2B_6.

	(100)	(010)	(110)	(001)
Fe_2B_6	0	1.64	1.12	0.01
Co_2B_6	18.81	22.41	0.18	0
H-Fe_2B_6(η=1/3)	1.78	0	4.47	1.27
H-Fe_2B_6(η=1/2)	0	2.86	0.86	0.01

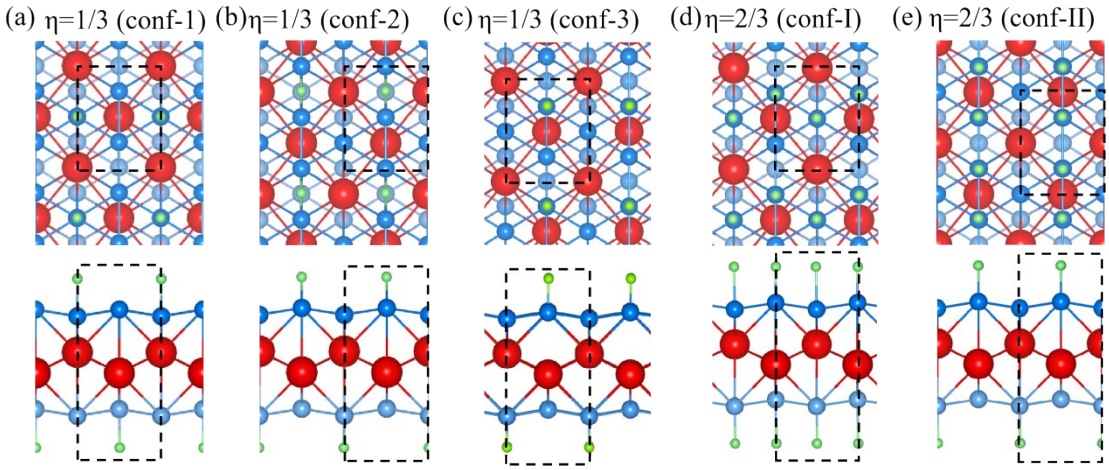

Figure S4. Possible configurations considered for H functionalized TM_2B_6 sheets with concentrations of η=1/3 and 2/3.

Table S4. Lattice Parameters (a,b), The distance (h) between transition meal layers, Energy Difference between FM and AFM states (ΔE), Magnetic Moments (MMs) on TM atoms, and Ground states (GS) of H-TM_2B_6(η=1/3) and H-TM_2B_6(η=2/3). M, SC and HM represent metal, semiconductor and half-metal, respectively.

TM	H-TM_2B_6(η=1/3)					H-TM_2B_6(η=2/3)				
	(a,b) (Å)	h (Å)	ΔE (eV)	MM (μ_B)	GS	(a,b) (Å)	h (Å)	ΔE (eV)	MM (μ_B)	GS

Ti	(2.98, 4.86)	1.50	--	0	NM	M	(3.08, 3.20)	1.79	--	0	NM	M
V	(2.92, 4.18)	1.31	-0.19	±2.35	AFM-1	SC	(3.00, 3.12)	1.62	-0.01	±0.72，±0.69	AFM-1	SC
Cr	(2.98, 4.44)	1.58	-0.25	±3.61	AFM-0	M	(3.08, 3.30)	1.66	-0.14	±3.53	AFM-0	M
Mn	(2.94, 4.48)	1.64	-0.17	±3.83，±3.95	AFM-2	M	(2.98, 3.22)	1.67	-0.07	±3.86	AFM-1	M
Fe	(2.94, 3.76)	1.09	0.16	2.29, 2.29	FM	HM	(2.94, 3.22)	1.53	-0.06	±2.28	AFM-0	SC
Co	(2.90,3.64)	1.13	-0.04	±0.43，±0.88	AFM-2	M	(2.96,3.16)	1.58	--	0	NM	M
Ni	(2.90, 3.64)	1.17	--	0	NM	SC	(3.00, 3.18)	1.62	--	0	NM	M

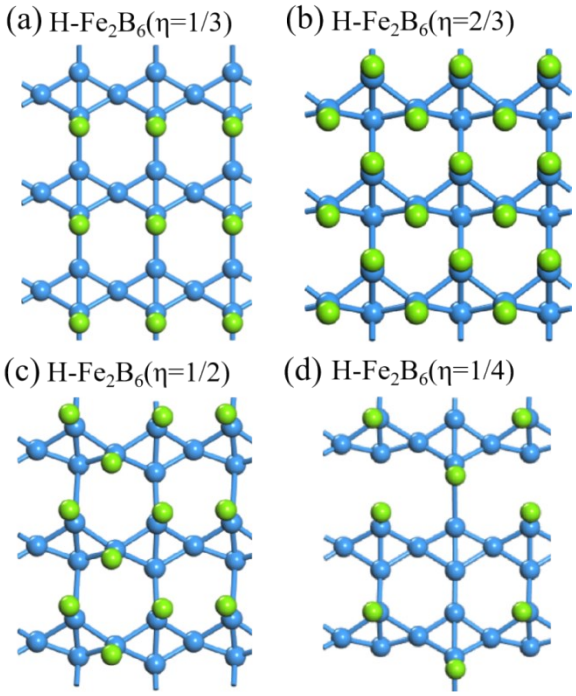

Figure S5. Top view of H functionalized Fe_2B_6 with H concentration of 1/3, 2/3, 1/2 and 1/4.

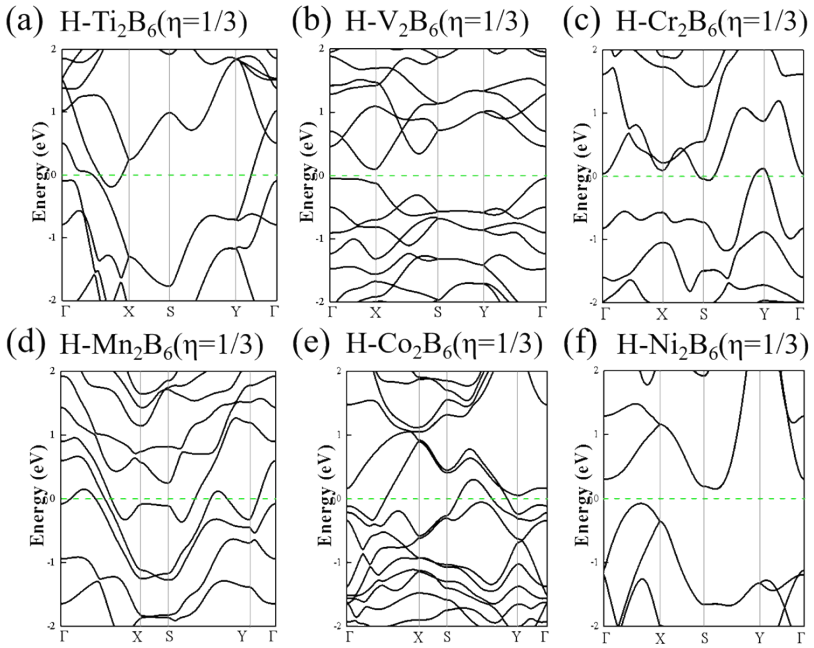

Figure S6. Band structures of H-TM_2B_6(η=1/3).

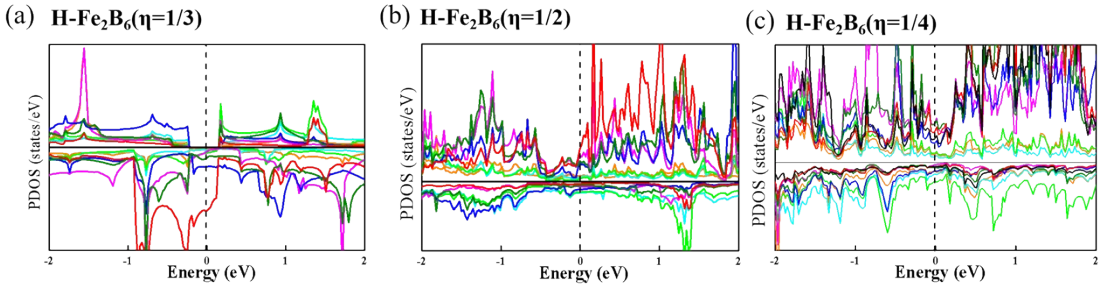

Figure S7. PDOS of H-Fe_2B_6 with H concentration of 1/3, 1/2 and 1/4.

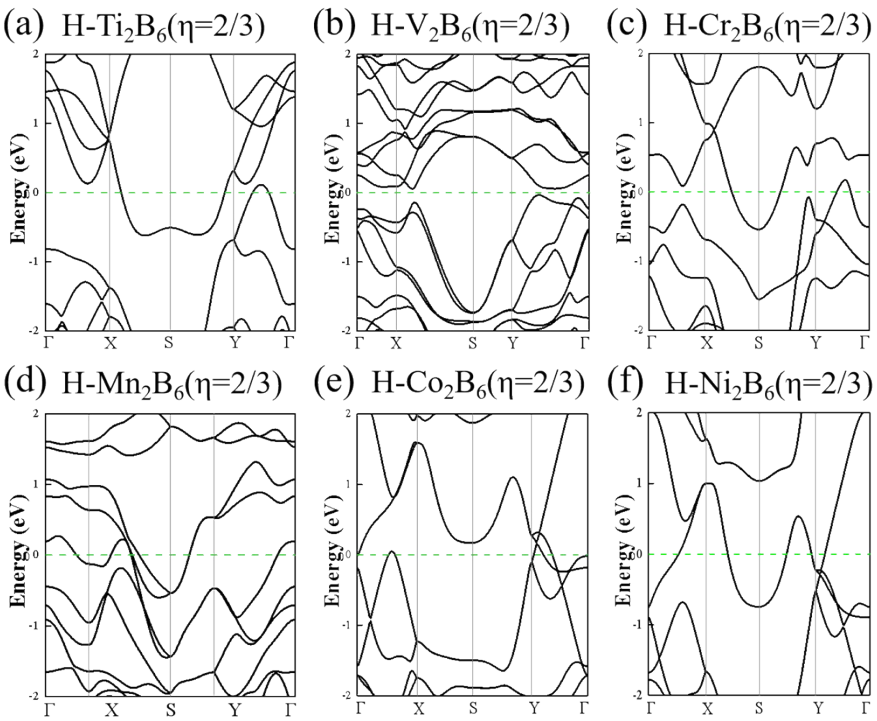

Figure S8. Band structures of H-TM_2B_6(η=2/3).

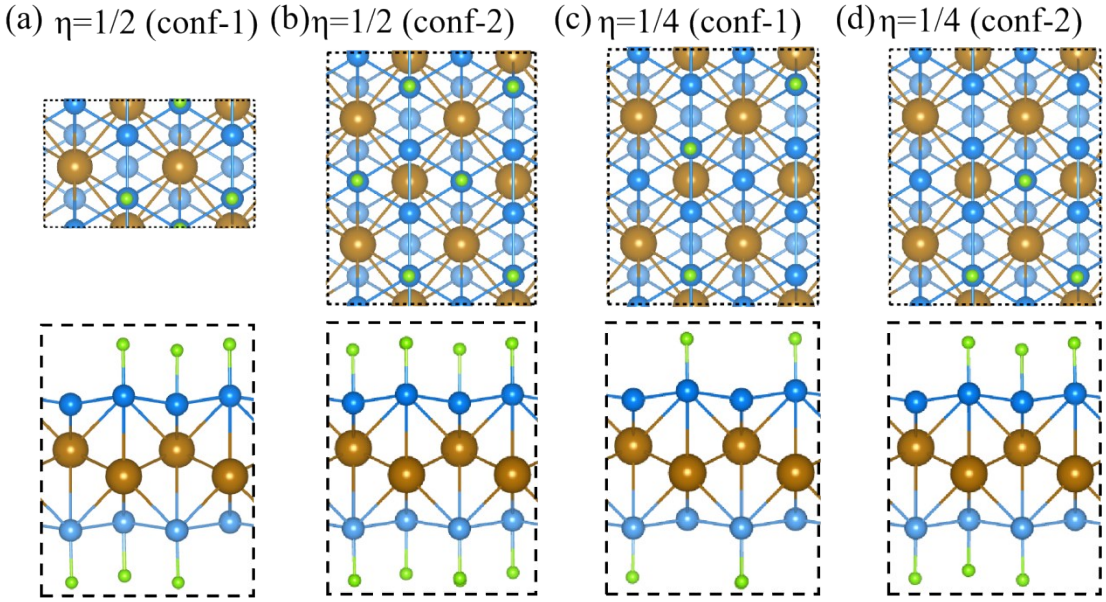

Figure S9. Possible configurations considered for H functionalized Fe_2B_6 sheets with η=1/2 and 1/4.