Supplemental Materials

for

Pressure-stabilized MnB_6 that exhibits high-temperature ferromagnetism and high ductility at ambient pressure

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Hubbard U parameter calculations

We have tested the Hubbard values (1 - 6 eV) by comparing the calculated and experimental lattice parameters of the known MnB. As tabulated in the following Table, we found that the derived structure parameters are in better agreement with experiments at U = 2.3 eV. We therefore recalculate the electronic structures and magnetic properties with the choice of U = 2.3 eV.

Table I. A comparation of the calculated structure parameters of MnB using the Hubbard U values chosen from 1 to 6 eV with experimental data [Sci. Rep., 7, 43759 (2017)].

Hubbard U (eV)		**1**	**2**	**2.3**	**2.5**	**3**	**4**	**5**	**6**	**Exp.**
Lattice parameters (Å)	***a***	5.5052	5.5820	5.6922	5.8248	5.9301	6.2681	6.3209	6.3672	5.6377
	b	2.9754	2.9684	2.9572	2.9615	2.9767	3.0696	3.0786	3.0812	2.9945
	c	4.1272	4.1432	4.1891	4.2673	4.3324	4.5751	4.6605	4.7654	4.1795
Volume ($Å^3$)	**V**	67.604	68.651	70.515	73.612	76.477	88.029	90.691	93.489	70.5587

Curie temperature calculations

We have calculated the Curie temperature of MnB_6 basing on 6×6×6, 8×8×8, 10×10×10, 12×12×12 and

14×14×14 supercells. As shown in the following figure, the results indicate that the Curie temperature becomes convergence with the using of the 10×10×10 supercell. Therefore, the Curie temperature of MnB_6 is estimated by classical Metropolis MC simulations with a 10×10×10 spin−lattice containing 1000 spin sites with periodic boundary conditions. Details of the tests on the supercell can be found in the Supplemental Materials. During the simulation steps, each spin is rotated randomly in all directions. The average magnetization per site is taken after the system reaches the equilibrium (with at least 10^5 simulation steps) state at a given temperature. The Curie temperature is taken as the critical point of the specific heat, defined as $Cv = (\langle E^2 \rangle - \langle E \rangle^2) / k_B T^2$.

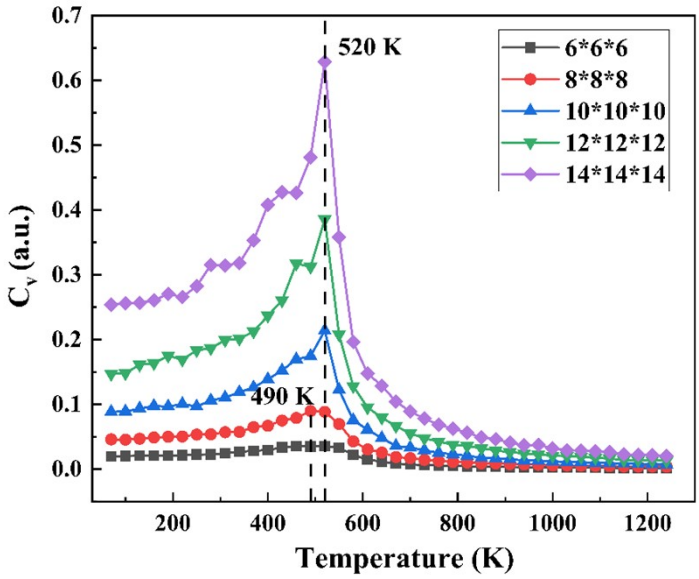


Figure 1. Temperature variation of the heat capacity for MnB_6.

Table SI. The bulk moduli (*B*, GPa), shear moduli (*G*, GPa), and the Vickers' hardness (*H*, GPa).

Phases	C_{11}	C_{22}	C_{33}	C_{44}	C_{55}	C_{66}	C_{12}	C_{13}	C_{23}	*B*	*G*	H_{vc}	H_{vt}
P-6*m*2 MnB_6	602.2	602.2	495.2	215.8	215.8	222.7	156.9	150.9	150.9	289.5	213.1	29.2	28.9
*C*2/*m* Mn_3B_4	553.6	493.7	463.4	180.2	214.1	220.4	195.8	240.0	189.5	305.5	177.8	19.0	19.5
*P*21/*c* MnB_4	549.3	957.6	531.4	238.9	243.6	170.8	58.3	121.8	90.0	279.8	240.9	38.5	37.7
*C*2/*m* MnB_3	333.9	455.1	468.3	164.8	305.5	251.7	42.0	176.2	175.4	218.0	185.9	32.3	31.0
Imma MnB_2	481.4	210.0	470.1	112.6	125.6	110.4	235.6	206.2	238.5	240.3	79.30	4.1	5.8
Pnma MnB_2	519.8	243.9	487.3	97.92	206.4	118.5	133.3	233.4	159.9	235.0	125.4	13.2	13.8
Pnma MnB	414.4	527.7	504.8	215.8	218.4	175.8	171.1	147.5	127.7	259.1	186.0	25.9	25.5
Cmcm MnB	504.1	504.1	496.1	158.4	149.7	26.46	95.63	199.8	199.7	269.6	92.25	5.0	6.7

*I*4/*mcm* Mn_2B	534.6	534.6	519.6	223.9	223,9	168.0	230.9	211.0	211.0	321.5	183.0	18.8	19.4

Table SII. The optimized structural parameters of predicted MnB, Mn_3B_4 and MnB_2 and MnB_6 compounds.

Compounds	**Space group**	**Pressure**	***a*, *b*, *c* (Å, deg)**	**Atomic positions**
MnB_6	*P*-6*m*2	0 GPa	a=4.3777 b=4.3777 c=2.8550 *α*=*β*=90 *γ*= 120	B1 (3j) (0.134 0.865 -0.000) B2 (3k) (0.463 0.536 0.500) Mn1 (1e) (0.667 0.333 0.000)
Mn_3B_4	*C*2/*m*	0 GPa	a= 7.3888 b= 2.8752 c= 5.5346 *α*=*γ*=90 *β*= 112.3838	B1 (4i) (0.967 0.000 0.147) B2 (4i) (0.189 -0.000 0.430) Mn1 (4i) (0.185 0.500 0.158) Mn3 (2d) (0.000 0.500 0.500)
MnB	*Cmcm*	0 GPa	a=2.6461 b=7.2404 c=2.8459 *α*=*γ*=*β*=90	B1 (4c) (-1.500 -0.430 0.250) Mn1 (4c) (-1.000 -0.355 0.750)
MnB_2	*Imma*	150 GPa	a=2.8181 b=4.5725 c=5.0489 *α*=*β*=*γ*=90	B1 (2c) (-0.667 -0.333 -0.000) Mn1 (1b) (-1.000 -1.000 -0.500)

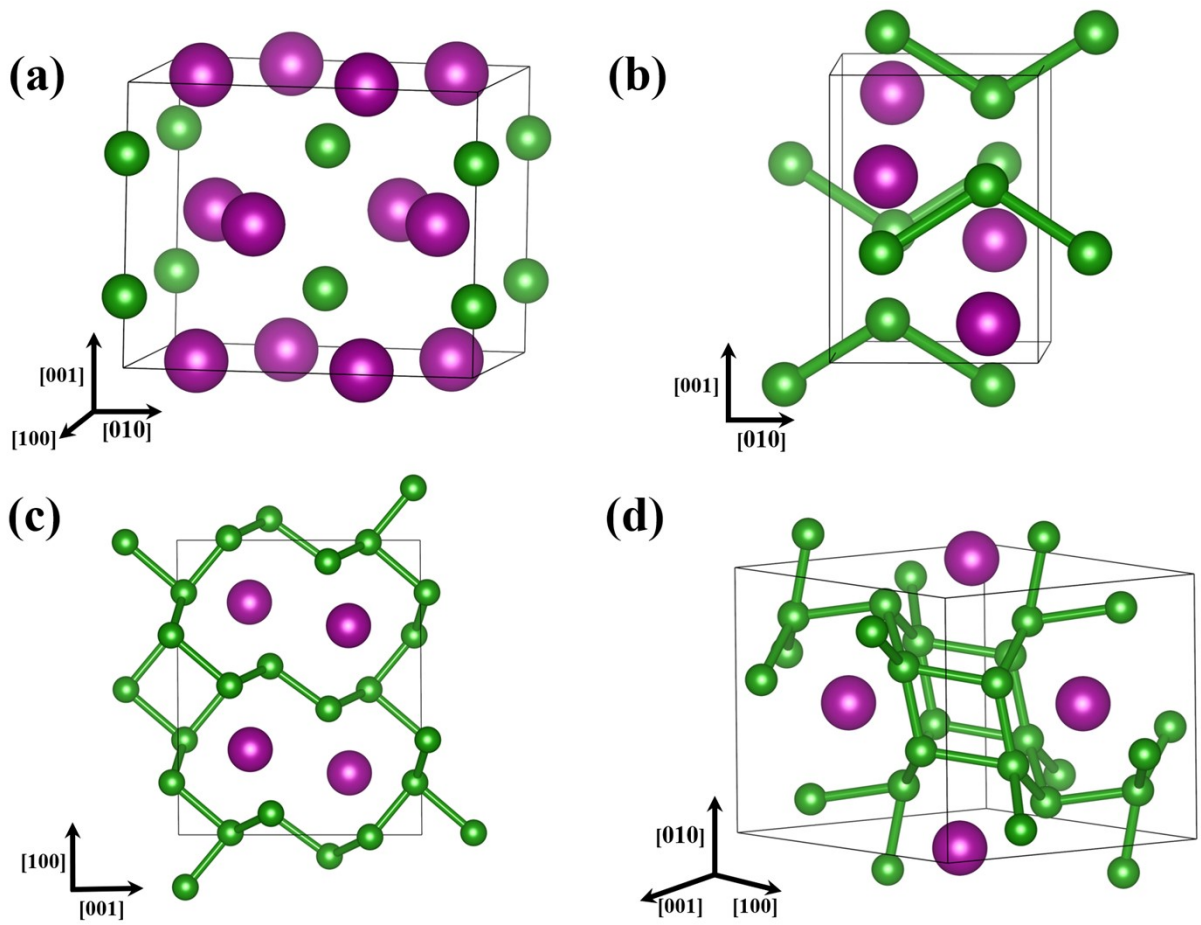


Fig. S1 Crystal structures of (a) the Al_2Cu-type structure of Mn_2B, (b) the FeB-type structure of MnB, (c) the C2/m structure of MnB_3 and (d) the $P2_1/c$ structure of MnB_4.

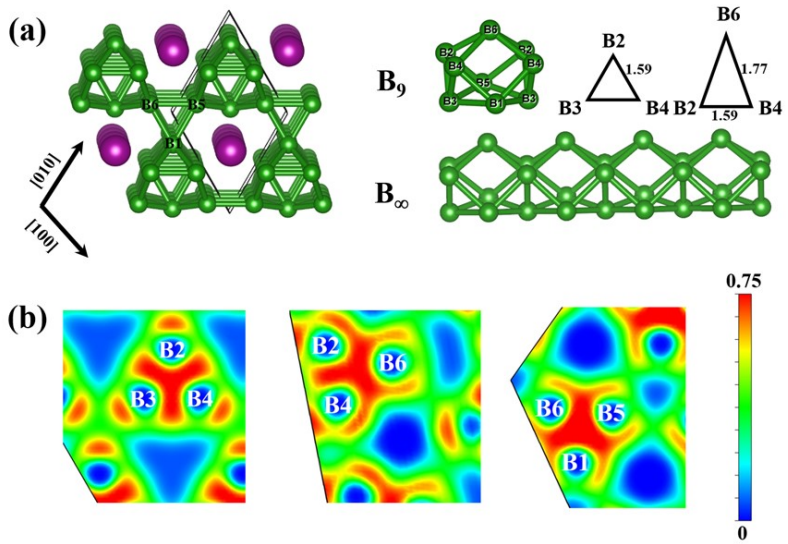
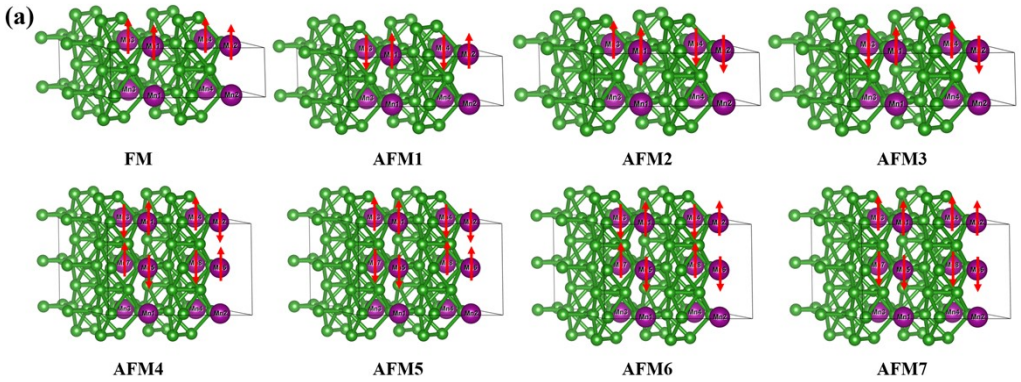


Fig. S2 (a) Crystal structures and (b) two-dimensional electron localization function (ELF) of MnB_6 at 127 GPa. Large purple and small green spheres represent Mn and B atoms, respectively.



(b)

Type	ΔH (eV/f.u.)
FM	0
AFM1	0.0931
AFM2	0.0931
AFM3	0.0931
AFM4	0.0932
AFM5	0.0932
AFM6	0.0932
AFM7	0.0932

Fig. S3 (a) The possible magnetic configurations of MnB_6 at 127 GPa. (b) The calculated enthalpies of different magnetic configurations with respect to FM with GGA+U methods at 127 GPa.

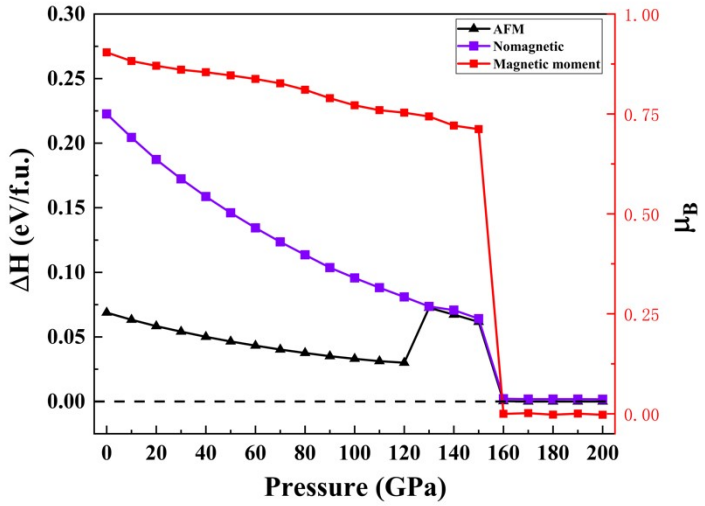


Fig. S4 The enthalpies of MnB_6 in different magnetic orders relative to ferromagnetic order.

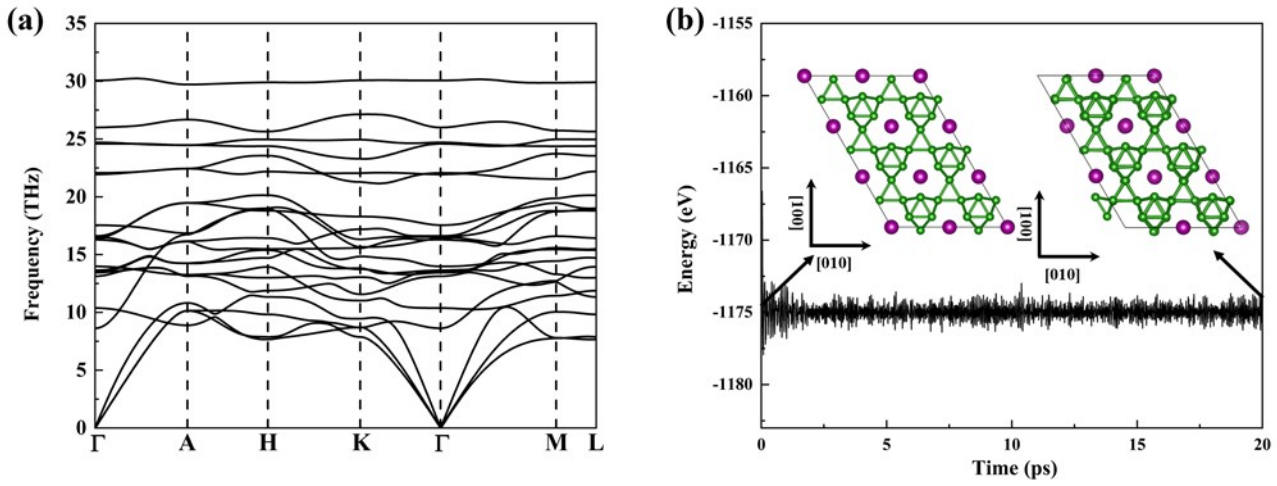


Fig. S5 (a) Phonon spectra of MnB_6 at 0 GPa. (b) Energy fluctuations during AIMD simulations up to 20 ps for MnB_6 structure at 0 GPa with 300 K.

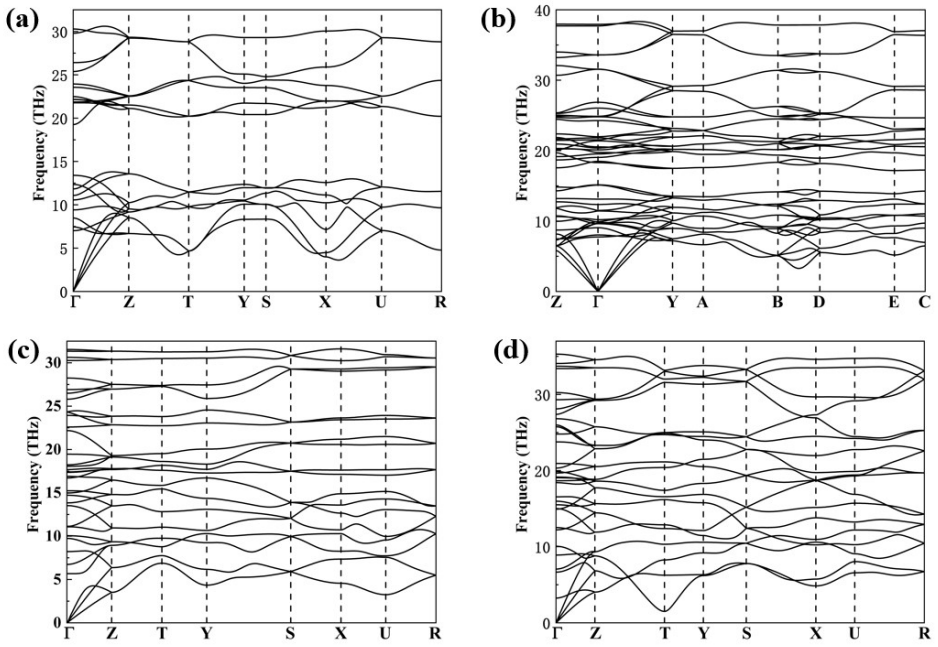


Fig. S6 Phonon spectra of (a) *Cmcm* MnB (100 GPa), (b) *C*2/*m* Mn_3B_4 (100 GPa), (c) *Pnma* MnB_2 (100 GPa) and (d) *Imma* MnB_2 (150 GPa)

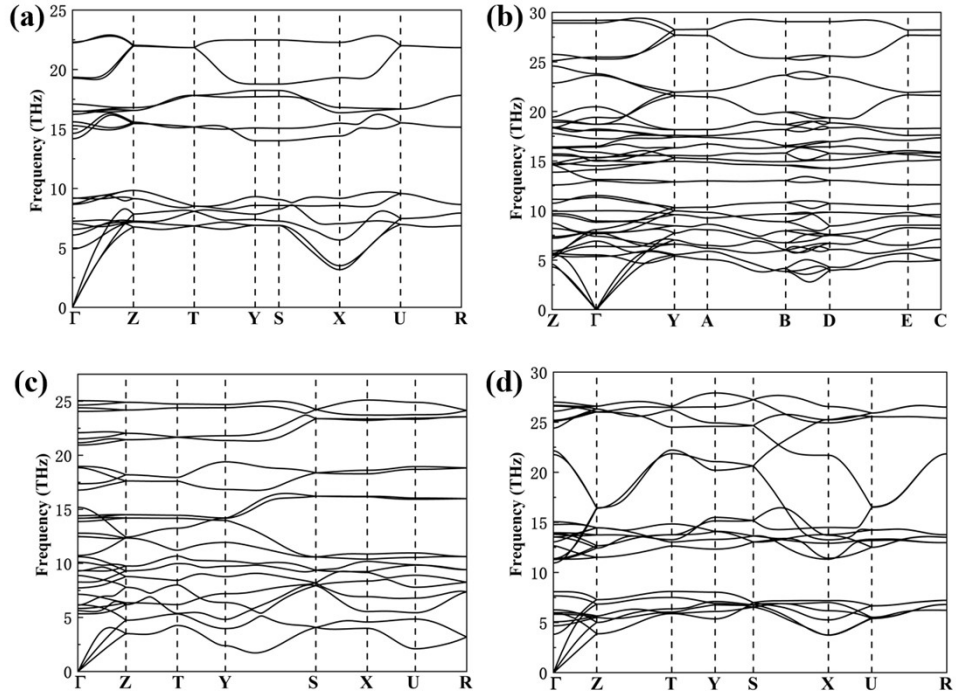


Fig. S7 Ambient-pressure phonon spectra of (a) *Cmcm* MnB, (b) *C*2/*m* Mn_3B_4, (c) *Pnma* MnB_2 and (d) *Imma* MnB_2.

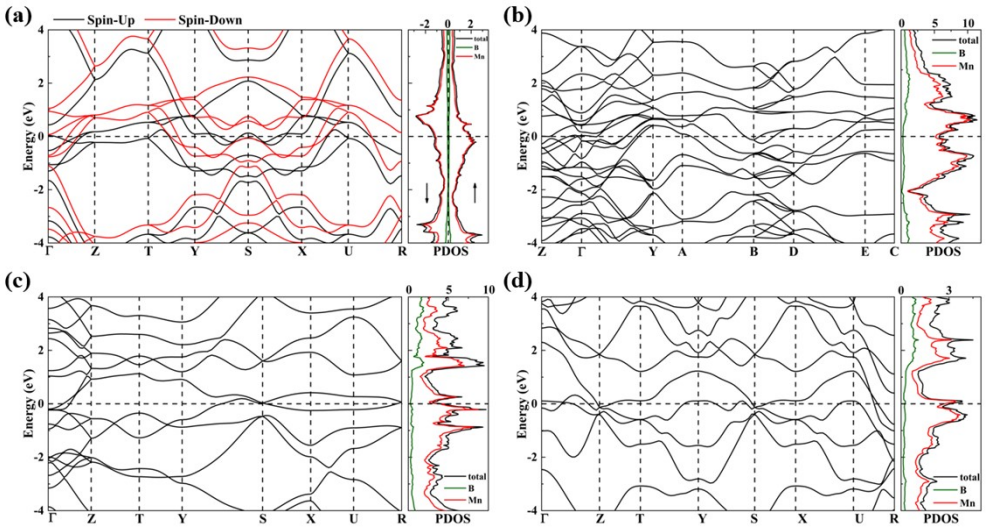


Fig. S8 Band structures and projected density of states (PDOS, in unit of states/eV/f.u.) of (a) *Cmcm* MnB at 100 GPa, (b) *C*2/*m* Mn_3B_4 at 100 GPa, (c) *Pnma* MnB_2 at 100 GPa and (d) *Imma* MnB_2 at 150 GPa. "↑" and "↓" represent spin-up and spin-down electronic density of states, respectively.

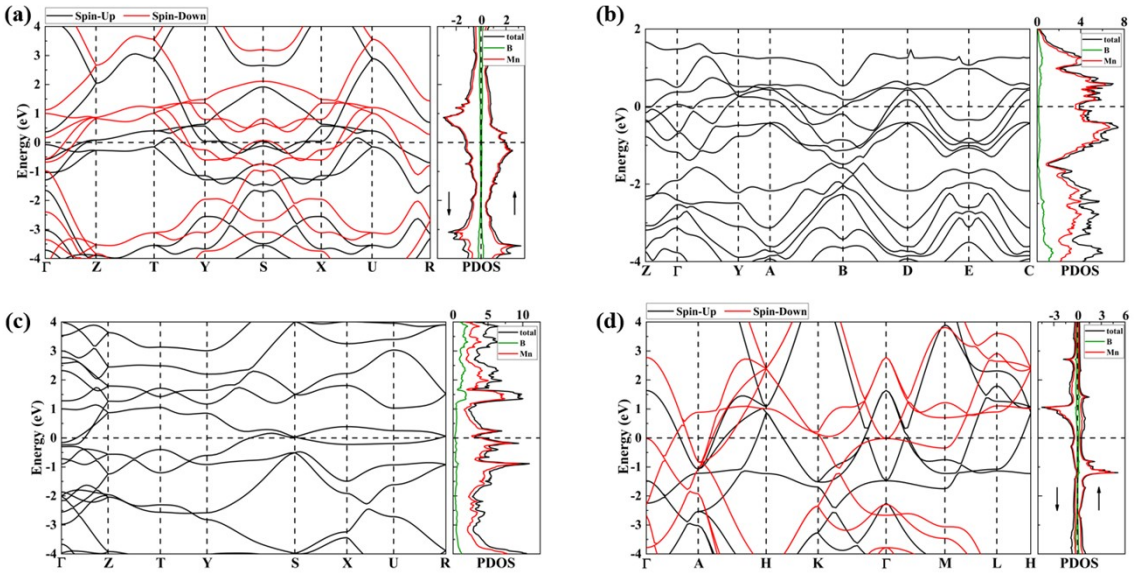


Fig. S9 Band structures and projected density of states (PDOS, in unit of states/eV/f.u.) of (a) *Cmcm* MnB, (b) *C*2/*m* Mn_3B_4, (c) *Pnma* MnB_2 and (d) *Imma* MnB_2. "↑" and "↓" represent spin-up and spin-down electronic density of states, respectively.