Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information for

Ferromagnetic Dirac Half-Metallicity in Transition Metal-Embedded Honeycomb Borophene

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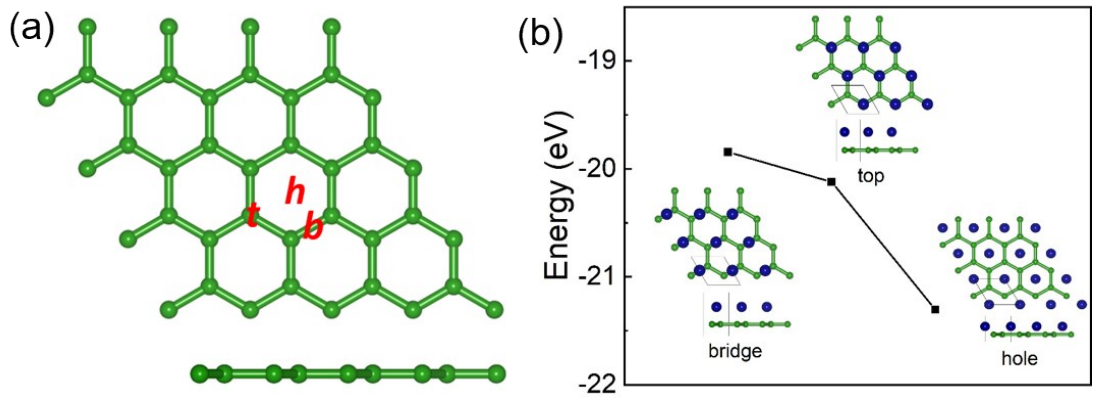

Figure S1. (a) Possible adsorption sites (*b* = bridge, *t* = top, *h* = hole) of transition metal atoms on the honeycomb borophene surface. (b) Energy profile for the three corresponding TMB_2 structures, also showing the structures corresponding to the *t*, *b*, and *h* configurations.

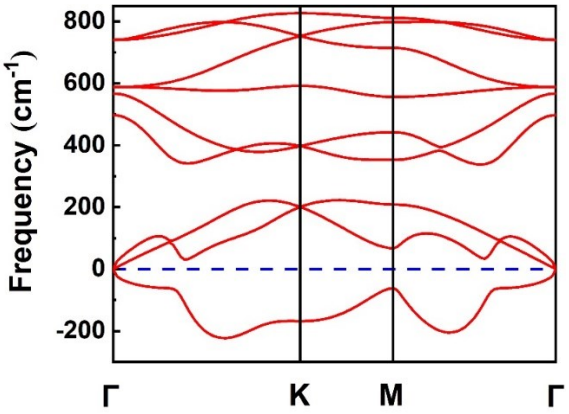

Figure S2. Phonon dispersion spectrum of MnB_2 monolayer.

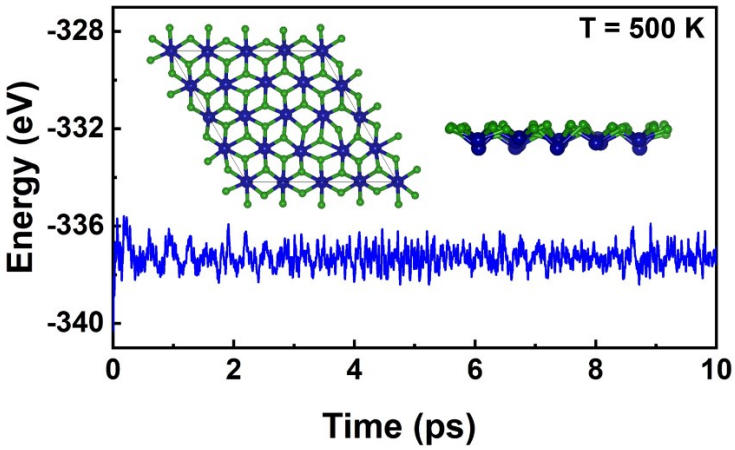

Figure S3. Top and side views of snapshots extracted from the MD simulations of 2D CrB_2 monolayers at the temperature of 500 K at the end of 10 ps, along with time evolutions of total energy.

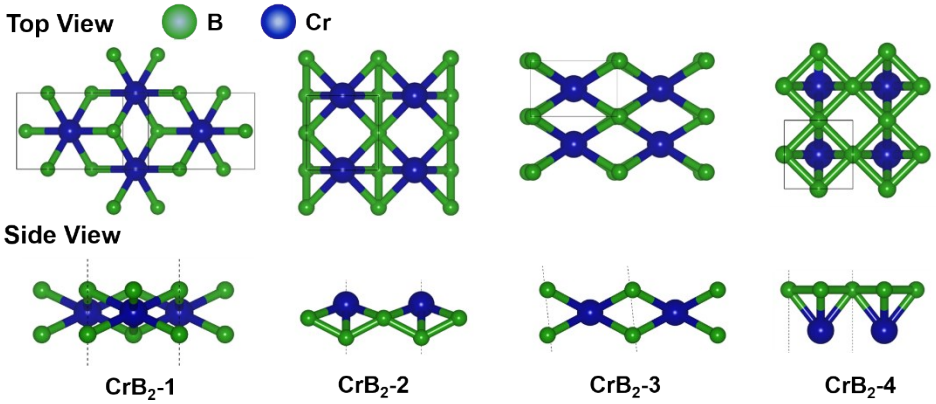

Figure S4. The other potential 2D CrB_2 allotropes were obtain in our global search. The CrB_2-1 is a monoclinic lattice in which the Cr atoms are sandwiched by the B atoms. The Cr atoms are six-coordinate with B atoms. The CrB_2-2 is an orthorhombic lattice and the Cr atoms are six-coordinate with B atoms. The CrB_2-3 is a monoclinic lattice. The Cr atoms are sandwiched by the B atoms and eight-coordinate with B atoms. The CrB_2-4 is a tetragonal lattice and the Cr atoms are four-coordinate with B atoms.

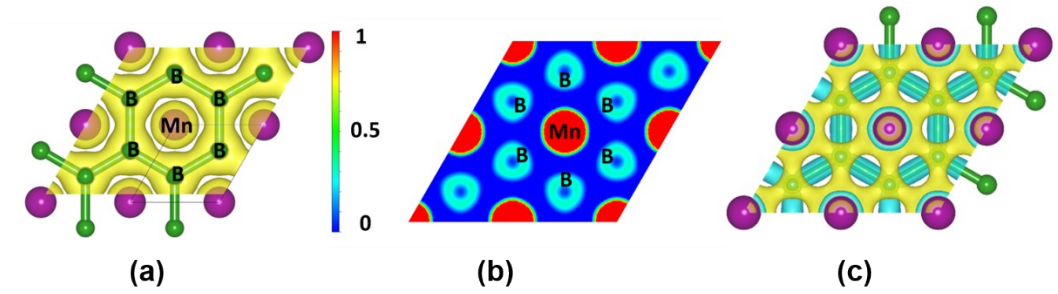

Figure S5. (a) Electron localization function (ELF) isosurface plotted at 0.09 e/Bohr^3 and (b) ELF map of CrB_2 monolayer in the plane perpendicular to the *z* direction. Red and blue colors correspond to the highest (1) and lowest (0) ELF values, respectively. (c) Differential charge density (isovalue: 0.01 e/Bohr^3) of monolayer MnB_2. Yellow and blue colors denote charge accumulation and depletion, respectively.

Table S1. Calculated energy (ΔE, eV/TMB_2) of magnetic states relative to that of non-magnetic states: $\Delta E = E_{magnetic} - E_{nonmagnetic}$. A positive ΔE value indicates that the magnetic state has lower energy.

TM	Ti	V	Cr	Mn	Fe	Co	Ni
ΔE (eV)	0	0	−0.17	−0.012	0	0	0

Table S2. Calculated energies of ferromagnetic (E_{FM}) and antiferromagnetic (E_{AFM}) states and energy differences (ΔE) between E_{AFM} and E_{FM} for Cr_6B_{12} formula unit.

	E_{Total} (eV)	ΔE (meV)
FM	-127.937	0
AFM1	-127.710	227
AFM2	-127.746	191
AFM3	-127.764	173

Table S3. *d*-projected density of states and net magnetic moment of Cr atoms.

	d_{z^2}	d_{xy}	$d_{x^2-y^2}$	d_{xz}	d_{yz}	Total
Spin up (*e*)	0.87	0.68	0.67	0.50	0.54	3.26
Spin down (*e*)	0.10	0.29	0.29	0.24	0.18	1.10
Net magnetic moment (μ_B)	0.77	0.39	0.38	0.26	0.36	2.16

S1. The Hamiltonian of the classical Heisenberg model can be written as:

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

where M_i is the spin magnetic moment per Cr atom, J_1, J_2, and J_3 are the first, second, and third nearest-neighbor exchange parameters, respectively. M (=2.16 μ_B per Cr atom) is the spin magnetic moment for CrB_2 monolayer. The exchange parameters J_1, J_2, and J_3 of the four magnetic configurations can be estimated by the following equations:

$$E(FM) = -\left(18J_1 + 18J_2 + 18J_3\right)M^2$$

$$E(AFM1) = -\left(-2J_1 - 6J_2 + 2J_3\right)M^2$$

$$E(AFM2) = -\left(-6J_1 + 2J_2 + 2J_3\right)M^2$$

$$E(AFM3) = -\left(-6J_1 - 6J_2 + 18J_3\right)M^2$$

$$\Delta E_1 = E(AFM1) - E(FM) = (20J_1 + 24J_2 + 16J_3)M^2$$

$$\Delta E_2 = E(AFM2) - E(FM) = \left(24J_1 + 16J_2 + 16J_3\right)M^2$$

$$\Delta E_3 = E(AFM3) - E(FM) = (24J_1 + 24J_2)M^2$$

$$J_1 = \frac{\Delta E_3 - 3(\Delta E_1 - \Delta E_2)}{48M^2}$$

$$J_2 = \frac{\Delta E_3 + 3(\Delta E_1 - \Delta E_2)}{48M^2}$$

$$J_3 = \frac{3\Delta E_1 + 3\Delta E_2 - 5\Delta E_3}{96M^2}$$