Supplementary Information for

Realizing Graphene-like Dirac Cones in Triangular Boron

Sheets by Chemical Functionalization

Shao-Gang Xu¹, Zhong-Jia Chen^{2,3}, Yu-Jun Zhao², Xiuwen Zhang^{1,*}, Hu Xu^{3,*} and Xiao-Bao Yang^{2,*}

¹Shenzhen Key Laboratory of Flexible Memory Materials and Devices; Key Laboratory of Optoelectronic Devices and Systems of Ministry of Education and Guangdong Province, College of Physics and Optoelectronic Engineering, Shenzhen University, Shenzhen 518060, P. R. China

²Department of Physics, South China University of Technology, Guangzhou 510640, P. R. China

³Department of Physics, Southern University of Science and Technology, Shenzhen 518055, P. R. China

E-mail: xiuwenzhang@szu.edu.cn; xuh@sustech.edu.cn; scxbyang@scut.edu.cn

The binding energy of the absorption systems is defined as $E_B = (E_{tot} - B_{sheet} - n \times \mu_X)/(m + n)$

where the E_{tot} represent the total energy of the 2D systems, B_{sheet} represents the total energy of the triangular B sheet, μ_X is the per atom energy of X₂ (X=H, F, Cl), and *m/n* represents the number of B/X atoms in the unit cell.

Table S1. The lattice constants, Binding energy (E_B) , Dirac cone and Fermi velocity $({}^{\nu}f)$ for all the B-X materials.

Phase (X= H, F, Cl)	a (Å)	b (Å)	E _B (eV/atom)	Dirac cone	v_f (10 ⁶ m/s)
B ₃ H	2.871	2.871	-0.280	Yes	0.492/0.511
B_6H_2	2.894	4.913	-0.351	Yes	0.291/0.390
B ₃ F	2.871	2.871	-1.099	Yes	0.419/0.469
B_6F_2	2.905	4.922	-1.176	Yes	0.379/0.427
B ₃ Cl	2.969	2.969	-0.388	Yes	0.441/0.471
B ₆ Cl ₂	2.998	4.882	-0.565	Yes	0.367/0.303

In order to measure the relative structural stability of the new structures, we have calculated the formation enthalpy $\Delta H(x)$ for the B-H systems,

$$\Delta H(x) = (E_{tot} - y \times \mu_B - x \times \mu_H)/(x + y)$$

where the E_{tot} represent the total energy of the 2D systems, μ_B is the referred to the per atom energy of α B sheet, μ_H is the per atom energy of H₂, and x/y represents the number of H/B atoms in the unit cell.

Table S2. The calculated formation enthalpy for the 2D B-H phases.

Phase	B_2H_2 (<i>Cmmm</i>)	B ₂ H ₂ (<i>Pmmn</i>)	B ₃ H	B ₆ H ₂
Δ <i>H</i> (eV)	-0.0145	-0.0061	-0.0381	-0.1095



Fig. S1. The phonon dispersions along the high symmetry lines of the new phases.



Fig. S2. The AIMD snapshots at the temperature of 500 K (10 ps) for $B_6H_2(a)$, $B_6F_2(b)$ and $B_6Cl_2(c)$, respectively.



Fig. S3. Orbital-resolved band structures and band decomposed charge density of the new phases. (a-c, d-f, g-i) represent the projected band structures and band decomposed charge density at Dirac point of B_6X_2 (X = H, F, Cl) monolayer, respectively. All the data in this figure are calculated using the PBE functional.



Fig. S4. The 2D electron localization function (ELF) at the honeycomb B planes of the new phases.



Fig. S5. (a-d) represent the band structures of the $p(\sqrt{3} \times \sqrt{3})$ supercell for graphene-(a), B₃H-(b), B₃F-(c) and B₃Cl-(d), respectively. (e-h) represent the band structures of the $p(2\times 2)$ supercell for graphene-(e), B₃H-(f), B₃F-(g) and B₃Cl-(h), respectively.



Fig. S6. (a) The hopping diagram of the TB model for B_6F_2 monolayer. (b) The band structure of B_6F_2 monolayer based on a tight-binding model. (c) The 1D energy bands for the B_6F_2 strip. (d) The black solid lines represent the PBE band, and the red dash lines represent the results of wannier fit.



Fig. S7. Orbital-resolved band structures and band decomposed charge density of the TMB_2 phases. (a-c, d-f, g-i) represent the projected band structures and band decomposed charge density at Dirac point of FeB₂, TiB₂ and ZrB₂ monolayer, respectively. All the data in this figure are calculated using the PBE functional.