Supplementary information materials

Engineering of Hydrogenated Two-dimensional h-BN/C

Superlattices as Electrostatic Substrates

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Fig .S1. The optimized structures of hydrogenated *h*-BN/C zigzag edged SLs with unequal ratio of two phases (m = 3, and n = 1).



Fig. S2. The averaged electrostatic energy of two types of SLs with unequal ratio of two phases (m+n=4, $1 \le n \le 3$), $Z^4(B_2N_2H_4)^{4-n}(C_4H_4)^n d1$ SLs (a), and $Z^8(B_2N_2H_4)^{4-n}(C_4H_4)^n d2$ SLs (b). From top to bottom plane, the zigzag width *n* is 1, 2, and 3 respectively. The potential steps across the $(B_2N_2H_4)^{4-n}$ in $Z^4(B_2N_2H_4)^{4-n}(C_4H_4)^n d1$ SLs with $1 \le n \le 3$ are 0.84, 1.17 and 0.82 eV respectively, and that in $Z^8(B_2N_2H_4)^{4-n}(C_4H_4)^n d2$ SLs are 1.84, 1.87 and 1.43 eV respectively.

The potential steps in the SLs with n=1 could be reduced by the screening effects due to free charges at the edges of $(B_2N_2H_4)^{4-n}$.



Fig. S3. Full band structure along the Γ -X-S-Y- Γ of the whole 2D Brillouin zone (red dashed lines in c) for $Z^{8}(B_{2}N_{2}H_{4})^{4}(C_{4}H_{4})^{4}d1$ (a), and $Z^{8}(B_{2}N_{2}H_{4})^{2}(C_{4}H_{4})^{2}d2$ (b).



Fig. S4. The valance band splitting energy near the Γ point with the increasing of $Z^{4n}(B_2N_2H_4)^n(C_4H_4)^nd2$ SLs width.



Fig. S5. The plane averaged electrostatic energy of SLs, $Z^8(B_2N_2H_4)^4(C_4H_4)^4d1$ (a), and $Z^8(B_2N_2H_4)^2(C_4H_4)^2d2$ (b). The macroscopic averaged potentials are plotted by red lines.



Fig. S6. Local projected density of states (LDOS) of the pristine graphene 2pz orbitals.



Fig. S7. The 2D potential contour plots of the SLs $Z^8(B_2N_2H_4)^4(C_4H_4)^4d1$ (a), and $Z^8(B_2N_2H_4)^2(C_4H_4)^2d2$ (c); and plots of their heterostructures with graphene (b) and (d), respectively.



Fig. S8. The local projected density of states (LDOS) of the carbon $2p_z$ orbital in graphene under the vertical threshold electric fields for unipolar doping mode transition. (Left) Graphene on the SLs $Z^8(B_2N_2H_4)^4(C_4H_4)^4d1$ at electric field of 0.56 V/Å, and (right) on $Z^8(B_2N_2H_4)^2(C_4H_4)^2d2$ at 0.2 V/Å.



Fig. S9. Band structures of comparison between using the PBE and HSE06 functional (PBE results: black lines; HSE06 results: colored dots) for graphene on N-H (a), C-H (b), and B-H surfaces (c).



Fig. S10. The bc-plane (a-axis) averaged potential energy (up) and the band structure (down) of the armchair hydrogenated BN/C SLs (e.g. left: $A^8(B_2N_2H_4)^4(C_4H_4)^4dl$ and right: $A^8(B_2N_2H_4)^2(C_4H_4)^2d2$).



Optimized atomic model of the graphene on the armchair hydrogenated BN/C SLs $(Gr/A^{12}(B_2N_2H_4)^4(C_4H_4)^2d2)$



Fig. S11. Contour plots of 2D charge density difference of graphene adsorbed on top of armchair hydrogenated BN/C SLs.

Unit cell	a-axis (Å)	strain (%)	b-axis (Å)	strain (%)
$C_4(Gr)/C_4H_4$	2.49	0.81/-1.58	4.32	0.93/-1.37
C ₄ (Gr)	2.47		4.28	
C ₄ H ₄	2.53		4.38	

The calculated lattice constants and strains of C_4 (graphene), C_4H_4 , and $B_2N_2H_4$ in their heterostructures.

Unit cell	a-axis (Å)	strain (%)	b-axis (Å)	strain (%)
$C_4(Gr)/B_2N_2H_4$	2.51	1.62/-2.71	4.35	1.64/-2.68
C ₄ (Gr)	2.47		4.28	
$B_2N_2H_4$	2.58		4.47	

The results show that all of the strains from the lattice mismatch are less than 3%. Therefore we use the lattice commensurate model to calculate their doping properties. For the lattice incommensurate bilayer superlattices, the small mismatch would induces moir épatterns. Many new physicals would emerge under this moir épotential, such as novel VanHove singularities, Fermi velocity renormalization and Hofstadter butterflies (*Nature Physics 8, 382–386 (2012)*). These new phenomena in the lattice incommensurate bilayer superlattices are beyond the scope of this manuscript.