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Gap openings in graphene regarding interfacial interaction from substrates

—Supplementary Information

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1. Methods to calculate atomic cohesive energy of graphene on the substrate

About $E_{C=C}$ in GLs/BN, it can be given by $E_{C=C} = (E_{total} - E_C - E_{system})/3$, where E_{total}, E_C and E_{system} denote the total energy of the whole system, the energy of a C atom removed from the graphene layer, and the energy of the system after removing the C atom, respectively. In the case of E_{C-C}, $E_{C=C}$ and E_{C-Si} with $n = 1$ for GLs/SiC, $E_{C-Si} = (E_{total} - E_{graphene} - E_{SiC})/2$, $E_{C-C} = (E_{graphene} - E_{C(-Si)})/3$, and $E_{C=C} = (E_{graphene} - E_C - E_{C-C})/2$, where E_{total} denotes the total energy of the whole system, $E_{graphene}$ the energy of graphene layer, E_{SiC} the energy of SiC part, $E_{C(-Si)}$ the energy of the C atom bonded to the Si atom of the substrate, and E_C the energy of the free C atom not bonded to the Si atom. In virtue of these, the values of E_{total}, E_C and E_{system} for GLs/BN and E_{total}, $E_{graphene}$, E_{SiC}, $E_{C(-Si)}$ and E_C for GLs/SiC should be given in advance. When $n = 2$, the way to have $E_{C=C}$ is similar to the above BN case. About GL/BN, the most stable stacking orders of graphene and BN will be considered, where one C is over B, while the other C is centered above an *h*-BN hexagon. As to GL/SiC, the SiC(0001) surface with the Si-termination is adopted. To explore them, the spin-polarized DFT calculations are performed using the Cambridge sequential total energy package.[1] The local density approximation with Ceperley and Alder-Perdew and Zunger[2] is employed to describe the exchange correlation effects.

2. Results of convergence between k-points

Table S1 Results of convergence between k-points

k-points		6 × 6 × 1	7 × 7 × 1	8 × 8 × 1
graphene on h-BN substrate	$d_{graphene-BN}$ (Å)	3.398	3.398	3.398
	band gap (meV)	36	36	36
graphene on 4H-SiC (0001) with Si termination	d_{Si-C} (Å)	1.973	1.973	1.973
	gap (eV)	1.73	1.73	1.72

In light of Table S1, the bandgap values and the interlayer distances between graphene and *h*-BN or 4H-SiC (0001) substrate keep unchanged with further increasing the *k*-points.

1. M. D. Segall, P. J. D. Lindan, M. J. Probert, C. J. Pickard, P. J. Hasnip, S. J. Clark and M. C. Payne, *J Phys-Condens Mat*, 2002, **14**, 2717-2744.
2. J. P. Perdew and A. Zunger, *Phys. Rev. B*, 1981, **23**, 5048-5079.