Supplementary Materials for
Influence of hexagonal boron nitride on electronic structure of graphene

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Contents:

1. The supplementary results calculated by VASP neglecting primitive lattice mismatch

Vienna Ab-initio Simulation Package (VASP) [1-5] is used to produce the detailed results for Gr/BN-21.8°, Gr/BN-13.1°, Gr/BN-9.4°, Gr/BN-5.1°, and Gr/BN-3.48°, shown in Figs. S1-S5, respectively. For discussion, please see the main text.

Gr/BN-21.8°

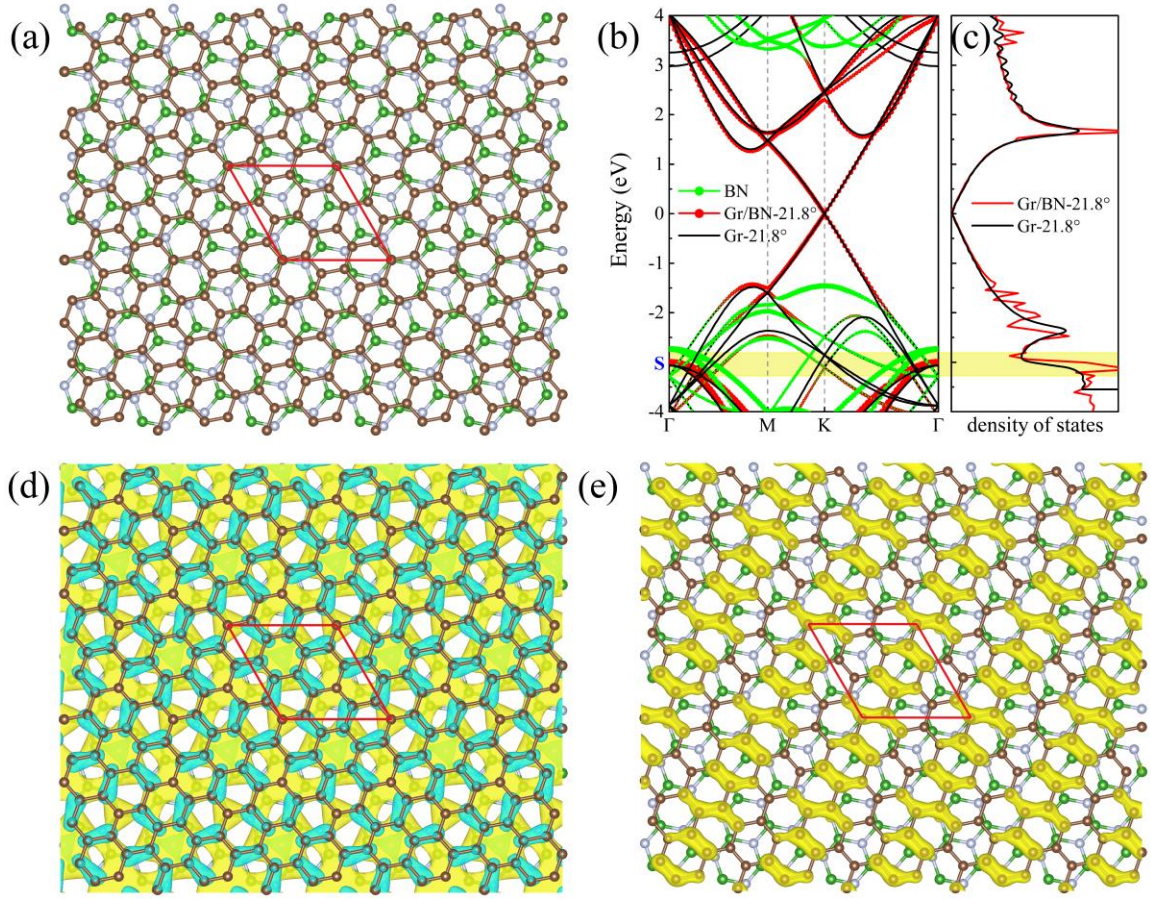

Figure S1. (a)Top view of the atomic structure of Gr/BN-21.8°. Brown, green and grey spheres denote C, B and N atoms, respectively. (b) Band structure of Gr/BN-21.8° in red dots and that of Gr-21.8° in black lines. Green dots denote contributions of BN layer. (c) DOS of Gr/BN-21.8° in red lines and that of perfect graphene in black line, calculated on a Γ-centered 31×31×1 k-mesh. (d) Differential total charge density. Green color indicates loss of electrons. (e) Charge density of the state with respect to an energy region marked by the yellow color bar in (b). Red diamond denotes a moiré superlattice induced by a 21.8° rotation between the upper and lower layers.

Gr/BN-13.1°

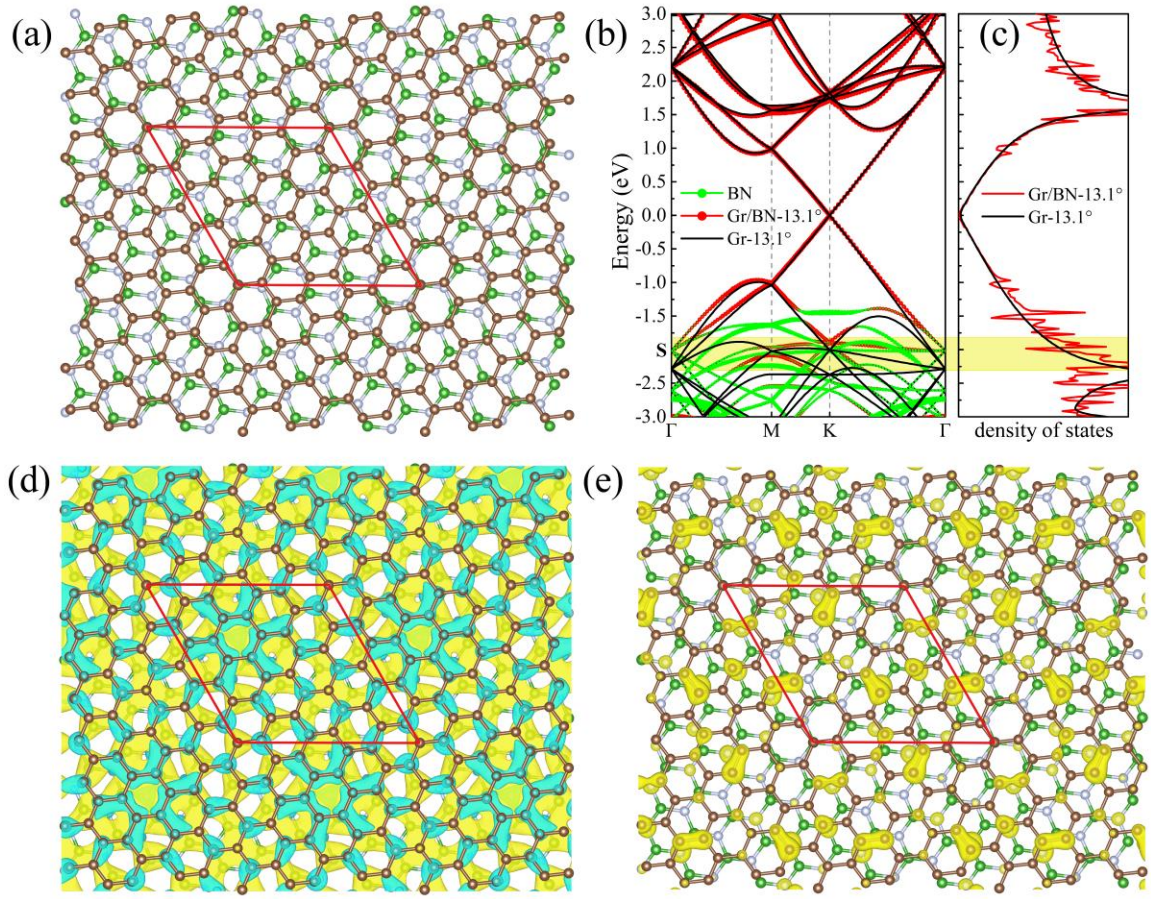

Figure S2. (a)Top view of the atomic structure of Gr/BN-13.1°. Brown, green and grey spheres denote C, B and N atoms, respectively. (b) Band structure of Gr/BN-13.1° in red dots and that of Gr-13.1° in black lines. Green dots denote contributions of BN layer. (c) DOS of Gr/BN-13.1° in red lines and that of perfect graphene in black line, calculated on a Γ-centered 21×21×1 k-mesh. (d) Differential total charge density. Green color indicates loss of electrons. (e) Charge density of the state with respect to an energy region marked by the yellow color bar in (b). Red diamond denotes a moiré superlattice induced by a 13.1° rotation between the upper and lower layers.

Gr/BN-9.4°

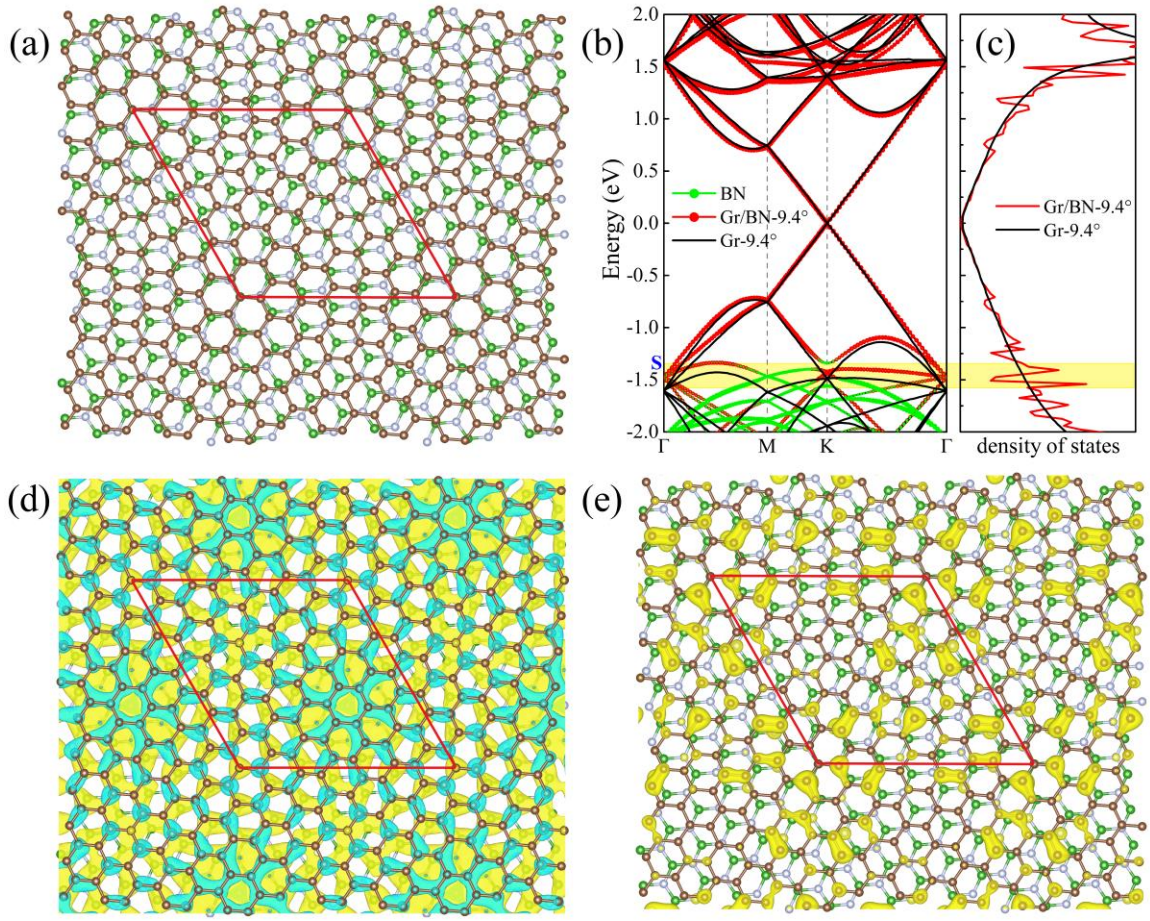

Figure S3. (a)Top view of the atomic structure of Gr/BN-9.4°. Brown, green and grey spheres denote C, B and N atoms, respectively. (b) Band structure of Gr/BN-9.4° in red dots and that of Gr-9.4° in black lines. Green dots denote contributions of BN layer. (c) DOS of Gr/BN-9.4° in red lines and that of perfect graphene in black line, calculated on a Γ-centered15×15×1 k-mesh. (d) Differential total charge density. Green color indicates loss of electrons. (e) Charge density of the state with respect to an energy region marked by the yellow color bar in (b). Red diamond denotes a moiré superlattice induced by a 9.4° rotation between the upper and lower layers.

Gr/BN-5.1°

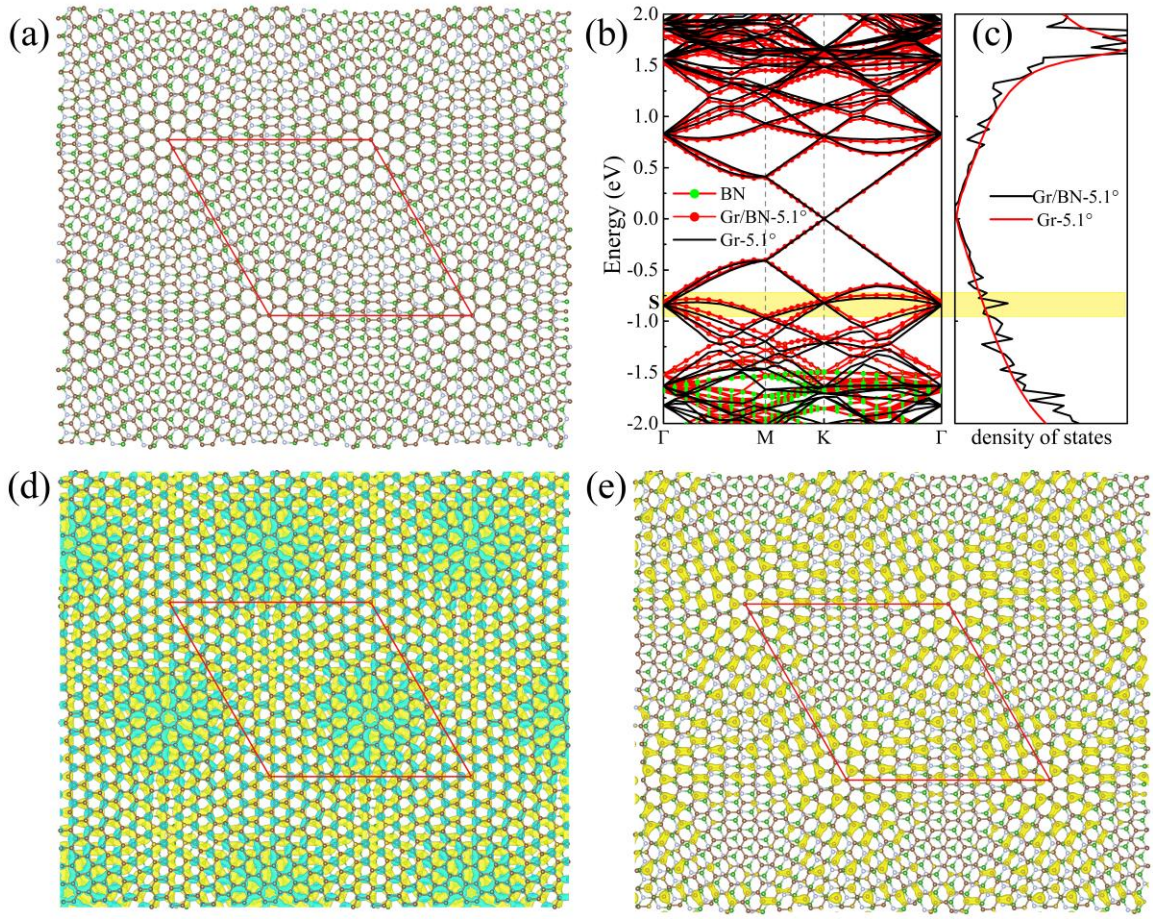

Figure S4. (a)Top view of the atomic structure of Gr/BN-5.1°. Brown, green and grey spheres denote C, B and N atoms, respectively. (b) Band structure of Gr/BN-5.1° in red dots and that of Gr-5.1° in black lines. Green dots denote contributions of BN layer. (c) DOS of Gr/BN-5.1° in red lines and that of perfect graphene in black line, calculated on a Γ-centered11×11×1 k-mesh. (d) Differential total charge density. Green color indicates loss of electrons. (e) Charge density of the state with respect to an energy region marked by the yellow color bar in (b). Red diamond denotes a moiré superlattice induced by a 5.1° rotation between the upper and lower layers.

Gr/BN-3.48°

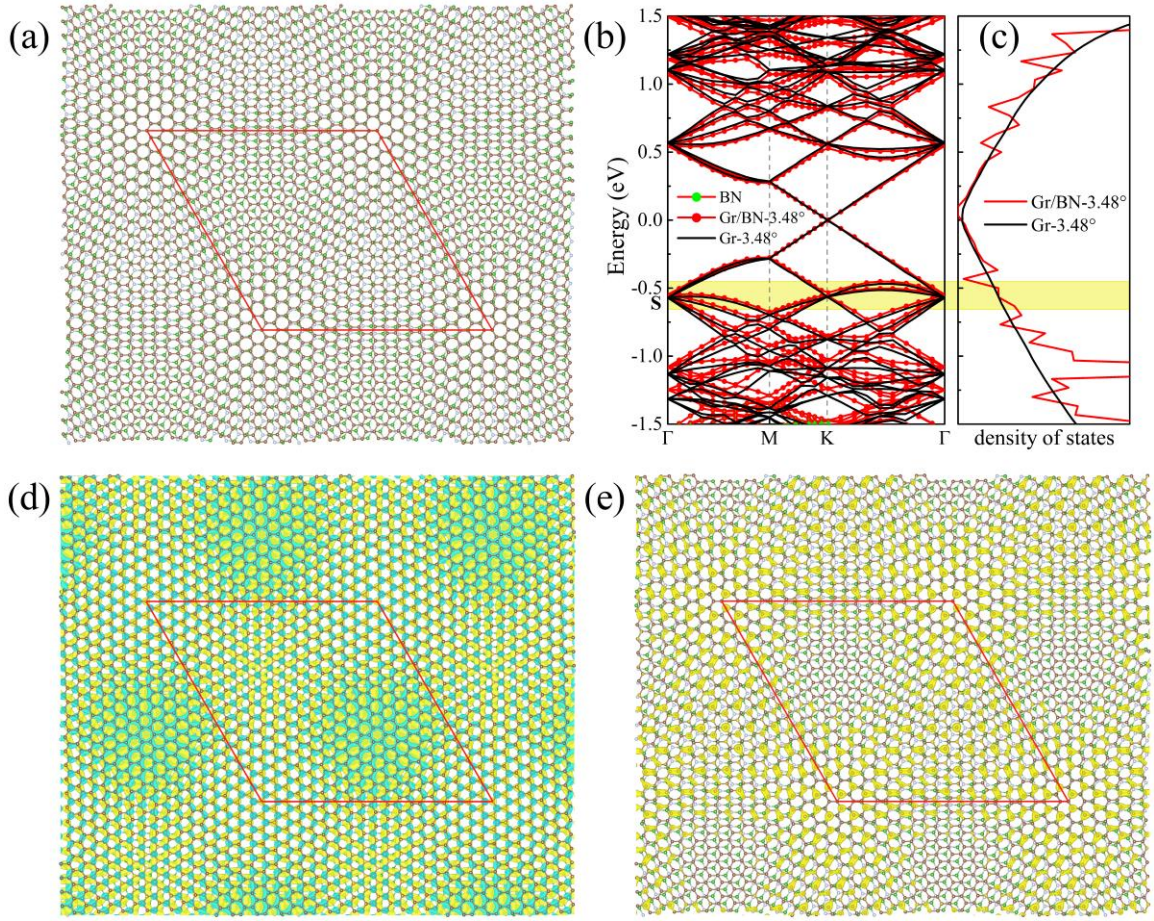

Figure S5. (a)Top view of the atomic structure of Gr/BN-3.48°. Brown, green and grey spheres denote C, B and N atoms, respectively. (b) Band structure of Gr/BN-3.48° in red dots and that of Gr-3.48° in black lines. Green dots denote contributions of BN layer. (c) DOS of Gr/BN-3.48° in red lines and that of perfect graphene in black line, calculated on a Γ-centered 6×6×1 k-mesh. (d) Differential total charge density. Green color indicates loss of electrons. (e) Charge density of the state with respect to an energy region marked by the yellow color bar in (b). Red diamond denotes a moiré superlattice induced by a 3.48° rotation between the upper and lower layers.

2. The supplementary results calculated by DFTB+ considering primitive lattice mismatch

The following calculated Gr/BN systems are very large, containing 3026 and 2654 atoms, respectively. The computational cost is increasing dramatically if the atom number grows larger. Comparing with VASP [1-4,6,7], the Density Functional based Tight Binding (DFTB+) model [8] is computationally affordable for large sized Gr/BN moiré patterns. The matsci-0-3 DFTB basis set is utilized in construction of the Hamiltonians [9]. For discussion of the figures below, please see the main text.

The untwisted Gr(28×28)/BN(27×27)

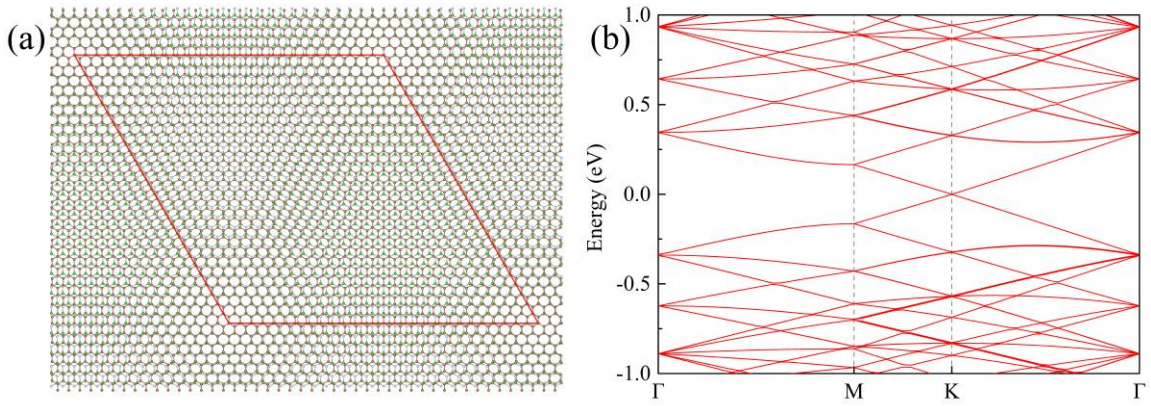

Figure S6. (a) the top view of the atomic structure of the untwisted Gr(28×28)/BN(27×27). Central regions of the upper left and lower right of the diamond have the quasi AB1 and AB2 stacking. Regions near the four vertices have the quasi-AA stacking. The red diamond denotes the moiré superlattice. (b) The energy bands of Gr(28×28)/BN(27×27).

Gr/BN-1.95° simultaneously considering the primitive lattice match and interlayer twisting

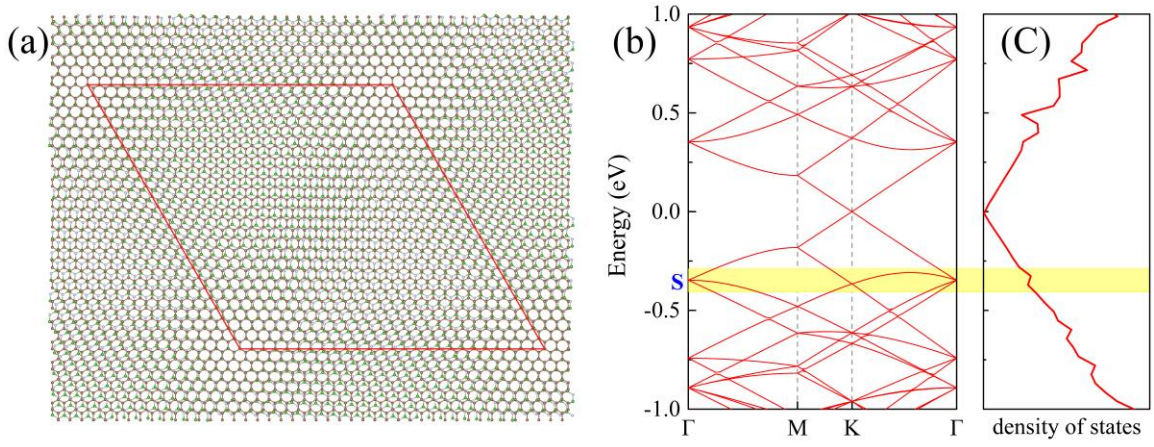

Figure S7. (a)Top view of the atomic structure of Gr/BN-1.95°, in which the super lattice constants of the unstrained graphene and BN are 63.96 and 63.89 Å, respectively, resulting a tiny super lattice mismatch of 0.11%. Brown, green and grey spheres denote C, B and N atoms, respectively. Red diamond denotes a moiré superlattice induced by a 1.95° rotation between the upper and lower layers. (b) The band structure of Gr/BN-1.95°. (c) The DOS of Gr/BN-1.95°, calculated on a Γ-centered 21×21×1 k-mesh. The first dip in DOS below fermi-level is marked by a yellow color bar, and its energy position correspond well to the S position in (a).

References for Supplementary Materials

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