

Electronic Supplementary Information for

**Synthesis of In-plane and Stacked Graphene/Hexagonal Boron Nitride
Heterostructures by Combining with Ion Beam Sputtering Deposition
and Chemical Vapor Deposition**

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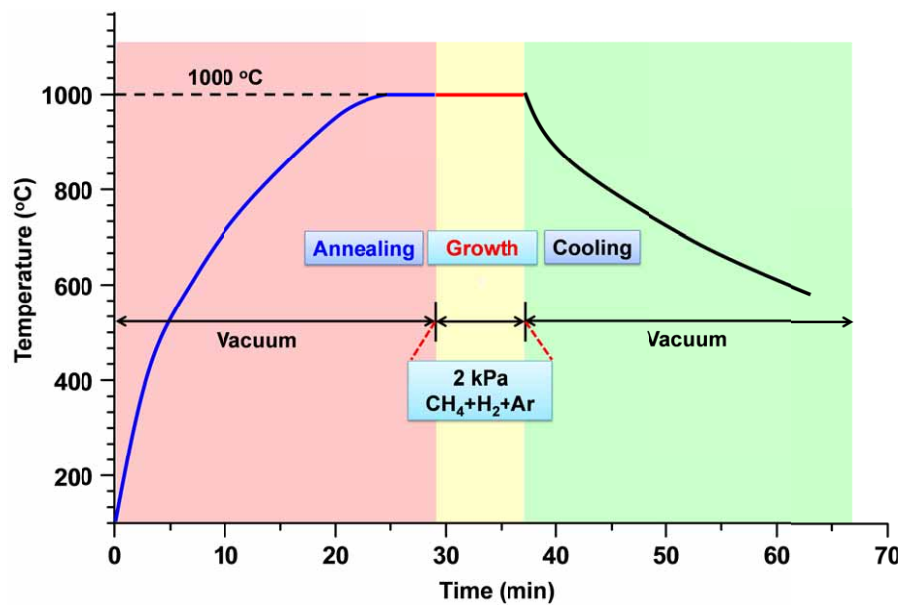


Figure S1. The growth protocol of graphene on the IBSD-grown h-BN/Cu substrates by CVD.

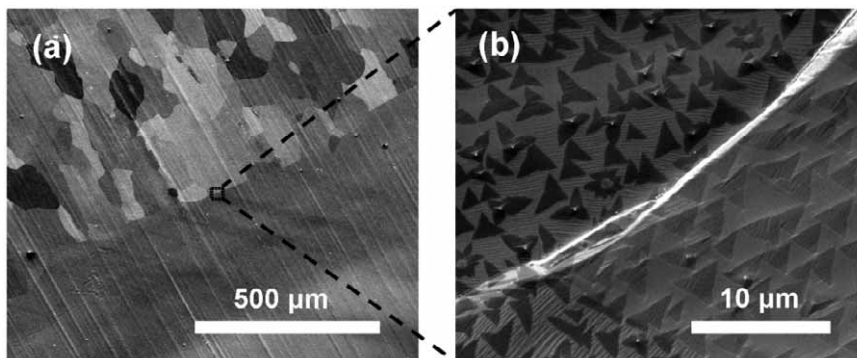


Figure S2. SEM images of the h-BN domains on Cu foils. (a) A large-scale SEM image showing the boundary between the melted/resolidified and the unmelted Cu surface. (b) The enlarged region of (a) marked with the dotted lines. The self-aligned h-BN domains were formed on the melted/resolidified Cu surface, while the randomly oriented h-BN grains were observed on the un-melted Cu substrate.

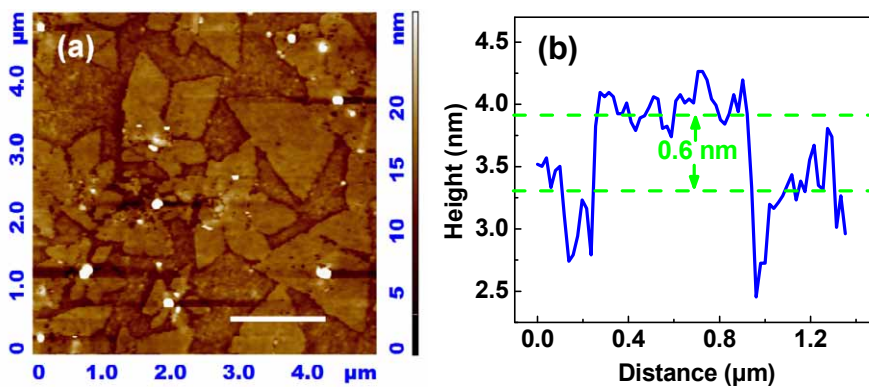


Figure S3. (a) Typical AFM image of h-BN domains transferred onto SiO₂/Si. (b) The height profile along the white line drawn in (a), showing a thickness of 0.6 nm for the h-BN flake. The result confirms the monolayer nature of the pre-grown h-BN.

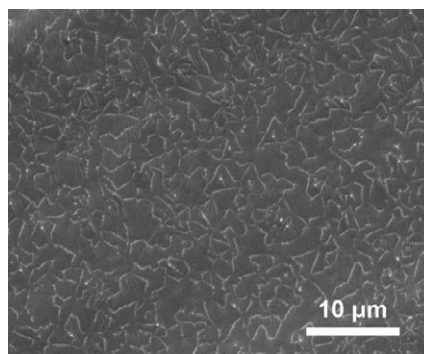


Figure S4. Typical SEM image of the continuous monolayer h-BN film. A large number of grain boundaries are observed in the h-BN film.

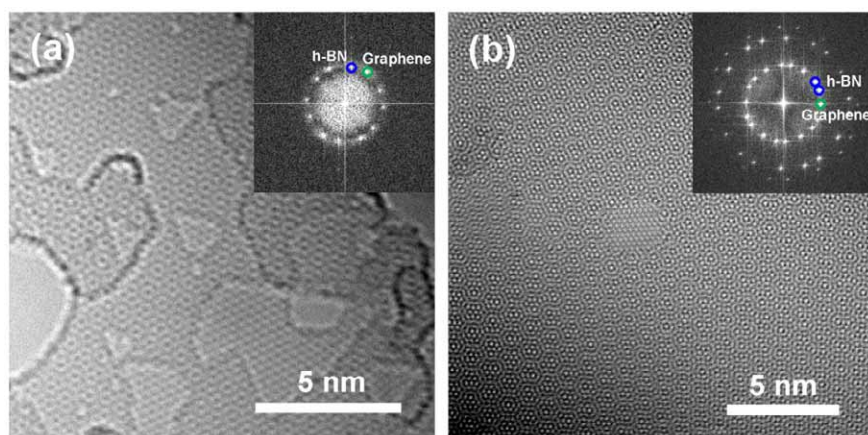


Figure S5. HRTEM images for the stacked graphene/h-BN heterostructures with different Moiré patterns. (a) Moiré pattern with some triangular defects. These defects are attributed to the irradiation damage of h-BN layer from electron beam. The corresponding FFT in the inset in (a) shows two separate spots, the blue one with the smaller reciprocal lattice from h-BN and the other one (green) from graphene. (b) Moiré pattern obtained from a monolayer graphene on a bilayer h-BN, the inset in (b) shows three separate FFT spots, two from h-BN and the other from graphene.

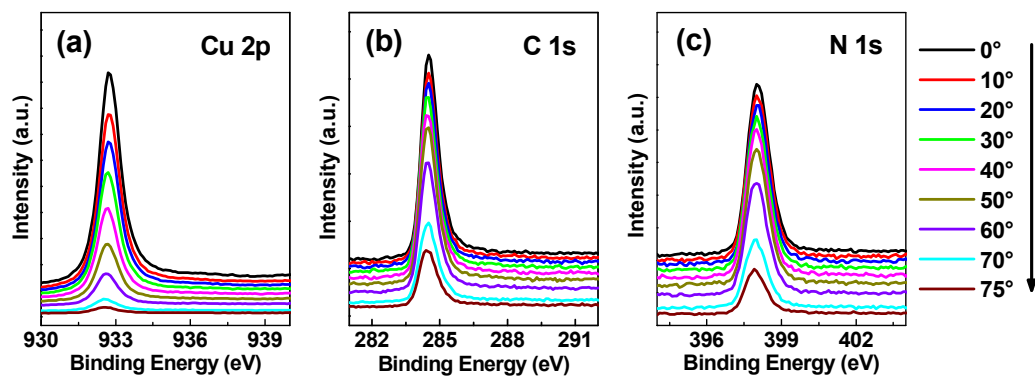


Figure S6. The raw XPS core level spectra of (a) Cu 2p, (b) C 1s, and (c) N 1s for the 12-min-grown graphene/h-BN film on Cu.