SUPPORTING INFORMATION FOR

Continuous Growth of Hexagonal Graphene and Boron Nitride In-Plane Heterostructures by Atmospheric Pressure Chemical Vapor Deposition

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Supplementary Figure 1. Preparation of ammonia-borane (AB) source 4 mg of ammonia-borane (AB) (Sigma-Aldrich, 682098) source was prepared as a tablet (~ 2mm tall x 1.5 mm diameter).



Supplementary Figure 2. Home-built holder for ammonia-borane (AB) source. (a-b) The AB source was placed in quartz tube that was laid on a home-built Teflon holder that had a magnet embedded in it. The AB source could be moved within the furnace tube using an external handheld magnet. (c) During graphene growth, the holder was placed away from the end of the heated furnace region to avoid undesired sublimation. (d) During the BN growth phase, the AB holder was placed closer to the chamber.



Supplementary Figure 3. Growth of boron nitride from graphene and bare Cu observed with optical microscopy (a) Triangular-shaped boron nitride flakes on copper were not observed by optical microscopy after 2 min preheating and 7 min growth at 1030 °C of the thin boron nitride hoop around the hexagonal graphene template. (b) 2 min preheating and 13 min BN growth sample. Small white dots, presumably very small BN flakes, are visible around the graphene flake. (c) 2 min preheating and 15 min BN growth sample. Triangular flakes of BN are clearly visible. This is the same image used in fig 1(b-c).
(d) BN flakes (triangle) start to grow at t=t₀ with ribbon growth around graphene at the same time. (e) BN flakes (triangle) start to grow at t=t₁ or t₂ (after BN ribbon growth from the graphene edge) (f) the image was from the black square in fig S3(b). Figure S2(e) is a more likely growth scenario than S2(d).



Supplementary Figure 4. AFM image for annealed Gr-BN layer The Gr-BN layer was annealed at 500 $^{\circ}$ C for 3 h in ambient¹ to remove the graphene region, while leaving the boron nitride region intact. Graphene was etched while boron nitride still remained compared with the image at Fig. 2e in the main manuscript. Height of boron nitride in profile is ~ 0.5 nm. Scale bar, 5 µm.



Supplementary Figure 5. Substrate background and BN peak (a) As a baseline, we examined the Raman spectrum from the oxidized silicon substrate, which shows a characteristic peak near 1450 cm⁻¹ due to the third TO-phonon mode of silicon². **(b)** The Raman spectrum of BN on an oxidized silicon substrate shows the BN peak at 1375 cm⁻¹ as well as the silicon peak².



Supplementary Figure 6. Electron energy loss spectroscopy (EELS) from boron nitride ribbon and electron diffraction (ED) pattern from graphene (a) Electron energy loss spectra show the expected K-edge peaks for boron (188 eV) and nitrogen (400 eV). Carbon peak (283 eV) is also observed due to carbon contamination, which may be caused by growth/transfer process or carbon TEM grid. (b) The images comprise 16 μ m² of graphene over a c-flat TEM grid. Electron diffraction analysis shows that in this area no grain boundaries are observable. Diffraction patterns taken from areas i, ii, iii and iv does not show appreciable rotation. (Compare the dashed line which has the same orientation). Selected area electron diffraction patterns were acquired with a JEOL 2010F operating at 200 kV equipped with a Gatan Orius CCD camera. A SAED aperture corresponding to approximately 700 nm was used for choosing the area from which the diffraction pattern was taken. Scale bar, 1 μ m.



Supplemental Figure 7. Cooling-induced tearing of the interface between graphene and boron nitride We observed occasional tearing at the interface between graphene and boron nitride, most likely due to strain caused by the cooling process³. Scale bar, 2 um.

References

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