Hexagonal Boron Nitride on Single-Crystal Epitaxial Graphene and SiC(0001) Substrates by Plasma-Enhanced CBE Deposition

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A unique high temperature, plasma assisted chemical beam epitaxial (CBE) deposition of hexagonal boron nitride (h-BN) is explored as a versatile method to grow thin films on large-area single crystal substrates in an effort to produce wafer-scale, epitaxial h-BN. Epitaxial graphene on SiC(0001) and reconstructed SiC(0001) surfaces are examined as candidate substrates for this growth method as single-crystal alternatives to transition metal foils typically used as h-BN substrates. Borazine dosing with the addition of RF nitrogen plasma exposure and substrate temperatures up to 1450°C, achieved with a custom modified manipulator, produced films with increased crystallinity and reduced polyaminoborane particulates on epitaxial graphene substrates. Samples were transferred *in-vacuo* for X-ray photoelectron spectroscopy (XPS) to examine stoichiometry and chemical environment of the sub-to-few monolayer BN films. Films grown with nitrogen plasma exhibited 10% higher nitrogen incorporation relative to those without plasma, showing active nitrogen is beneficial in maintaining stoichiometry at these growth temperatures in ultra-high vacuum. Use of plasma aids single-phase growth as indicated by the absence of shoulder peaks in B 1s and N 1s XPS spectra (Fig. 1). Crystallinity and orientation of nuclei was examined by *in-vacuo* and *ex-situ* scanning probe microscopy.

Growth of single-phase, stoichiometric h-BN was also achieved on SiC(0001) substrates. *In-situ* reflective high-energy electron diffraction during h-BN deposition shows nucleation differs between the Si-rich (3x3) and the C-rich $\left(6\sqrt{3}\times 6\sqrt{3}\right)R30°$ SiC surface reconstructions: the (3x3) reconstruction quickly transitioned to a (1x1) reconstruction upon precursor dosing, but the C-rich reconstruction persists. Instead of triangular nuclei seen on graphene surfaces, h-BN growth on reconstructed SiC surfaces followed the stepped morphology of the SiC surface.

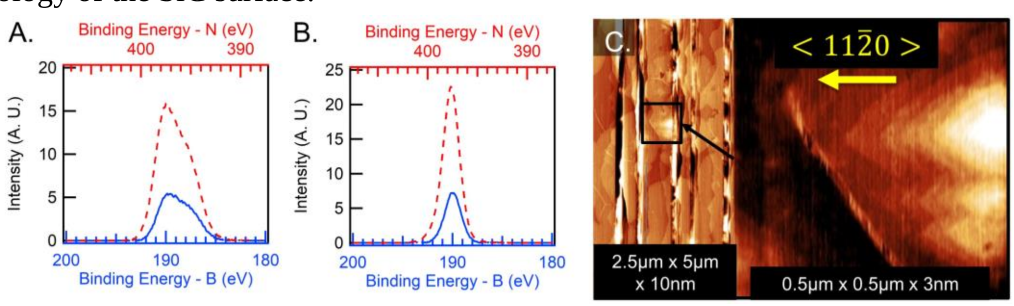

Fig. 1: XPS spectra of the B 1s (solid blue) and N 1s (dashed red) core levels from CBE growth (A) and plasma-assisted CBE growth (B) using borazine at 1450°C on epitaxial graphene substrates. The lack of lower binding-energy peaks in Fig. 1B indicate phase-pure films. Fig. 1C shows atomic force micrographs of h-BN on epitaxial graphene after a 1450°C growth, with a closeup of a multilayer h-BN domain. The underlying SiC substrate <11-20> direction is indicated.

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Supplementary Page

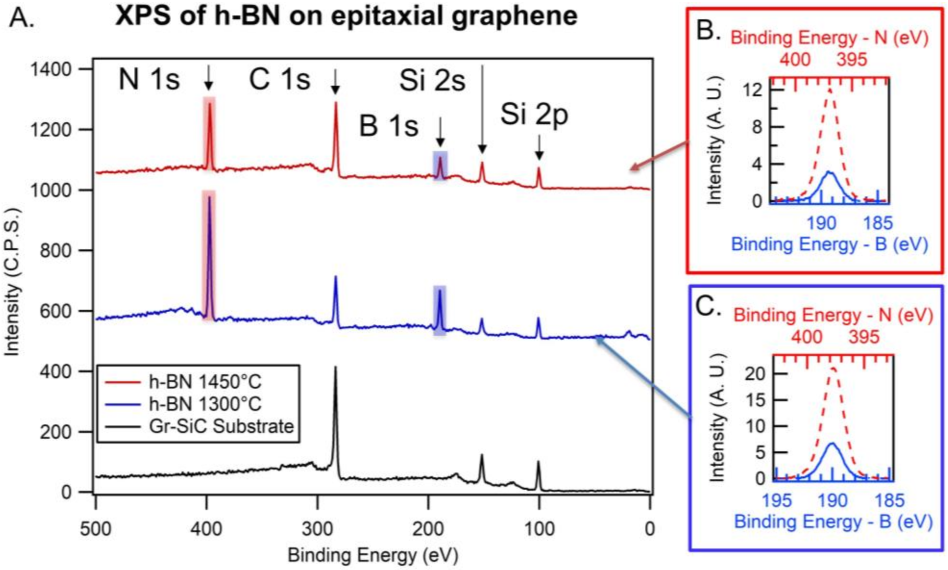

Fig. S1: A) X-ray photoemission spectra showing survey scans of the as-loaded epitaxial graphene on 4H-SiC(0001) substrate (black) and the CBE deposited BN films at 1300°C (blue) and 1450°C (red). The insets show the B1s and N1s core level peaks, corresponding to a ratio of 84% B:N (nitrogen rich) for the 1300°C deposition and 70% B:N for the 1450°C deposition as referenced to a CVD-grown control. According to XPS substrate peak attenuation, the film is approximately 9Å or ~3 monolayers thick.

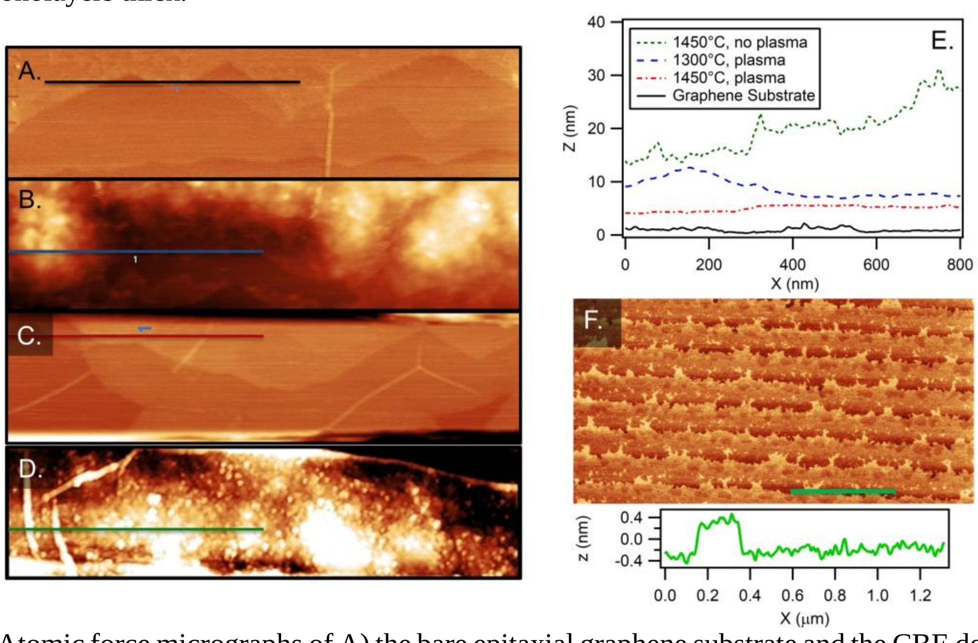

Fig. S2: Atomic force micrographs of A) the bare epitaxial graphene substrate and the CBE deposited h-BN at B) 1300°C with nitrogen plasma, C) 1450°C with nitrogen plasma, and D) 1450°C without plasma. Each micrograph is 2µm×0.5µm×10nm. Line scans in E) show the topography roughens due to polyaminoborane particles in growths without plasma or at lower growth temperatures. The surface steps of the $(6\sqrt{3}\times 6\sqrt{3})R30°$ SiC(0001) surface coated in h-BN from a 1450°C plasma-assisted CBE deposition are shown in F) with a line scan showing surface step height.