Rotational Alignment of Epitaxially-grown hBN on Macrostepped Graphene/SiC(0001) Single-Crystal Substrates

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Many of the intriguing properties of 2D devices rely on the relative rotational alignment between layers. For instance in the graphene/hBN system, band structure modulation can occur at specific alignments [1], but a misalignment may be beneficial if innate graphene properties are to be examined. To allow for scalable graphene/hBN heterostructure formation, this work investigates hBN growth on single-crystal epitaxial graphene (EG) on macrostepped SiC(0001) substrates. The presented results suggest these macrosteps may influence the hBN epitaxial relation such that a metastable, 30° in-plane hBN/EG alignment is more favorable with certain growth conditions than the direct 0° alignment between hBN/graphene, despite their similar crystal structures.

Plasma-enhanced chemical beam epitaxy (PE-CBE), an ultra-high vacuum (UHV) compatible process, was utilized to provide a clean environment for examination of the hBN structural, electrical, and chemical properties via *in-situ* and *in-vacuo* characterization methods. To determine the effect of substrate macrostep morphology, EG on SiC (0001) substrates with no offcut and with a 4° offcut toward $<11\text{-}20>_{SiC}$ were tested. The alignment of the hBN/EG/SiC(0001) heterostructure was studied by relating *in situ* electron diffraction to nuclei edge directions. In addition, cross-sectional transmission electron microscopy (TEM) confirmed registry of the hBN to the EG/SiC substrate, while plan-view TEM showed in-plane alignment and uniformity. The macrostep-directed epitaxy of hBN on EG highlighted in this work highlights the possibility of various rotational alignment during van der Waals epitaxy, a promising feature for direct growth of 2D heterostructures.

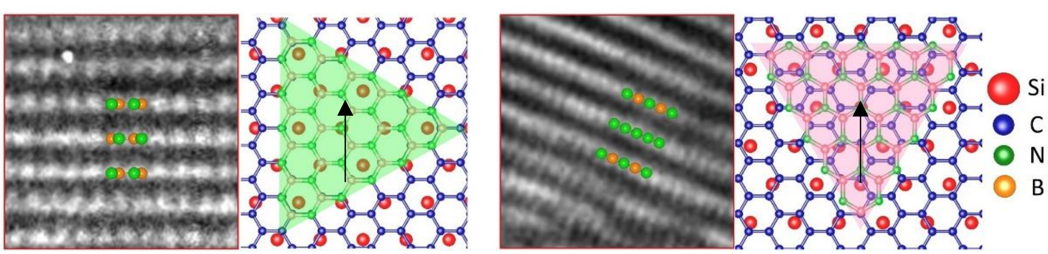

Figure 1: Cross-sectional HRTEM of hBN grown by PE-CBE on EG/SiC(0001) substrates imaged in the <10-10> zone axis of SiC and associated schematic of the plan-view hBN/EG/SiC crystalline orientation for the “high-flux” condition (left) and “low-flux” condition (right). Arrow in schematic indicates the zone axis of TEM images.

[1] M. Yankowitz, *et al.*, Nat. Phys. **8**, 382 (2012).

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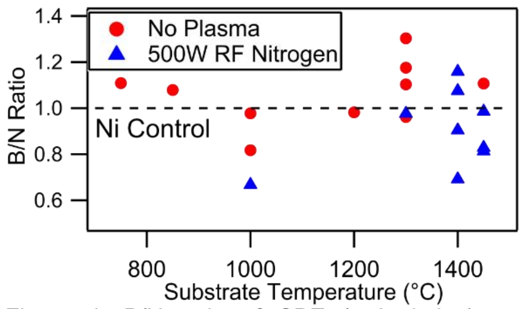

Figure 1: B/N ratio of CBE (red circles) and plasma-enhanced CBE (blue triangles) hBN as determined by XPS peak area ratios. Stoichiometry is relative to hBN films grown on nickel substrates without plasma.

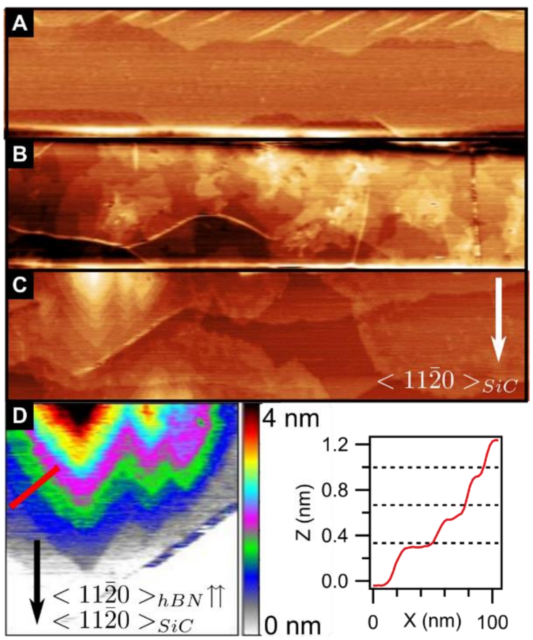

Figure 2: AFM of hBN grown by PE-CBE on epitaxial graphene (EG) substrates. A) A micrograph of the as-received substrate. B) 4nm hBN grown at 1450°C showing homogeneously nucleated hBN domains with rotational disorder in the middle of an (EG) plateau. C) A slower hBN deposition of hBN at 1450°C with multilayer hBN domain shown. D) A zoom-in of highlighted region of (C), with the direction of the SiC(0001) and proposed hBN nuclei lattice orientation. Line scan with dashed lines corresponding to expected hBN monolayer spacing. Image scales are 5μm x 0.2μm x 5nm for A-C and 0.5μm x 0.5μm x 4nm for D.

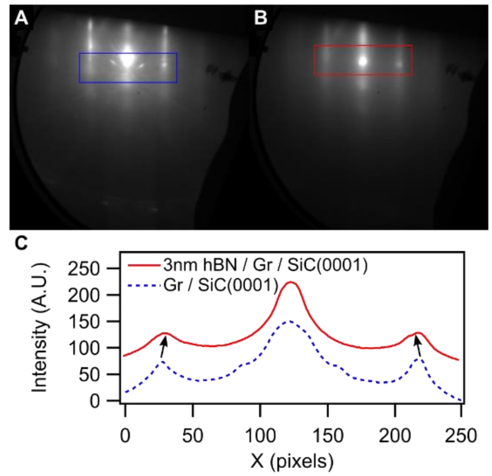

Figure 3: RHEED images of EG/SiC(0001) in the <10-10> direction (A) and the same sample with ~3nm hBN deposited via PE-CBE (B). Averaged line scans across the first-order streaks in (C).

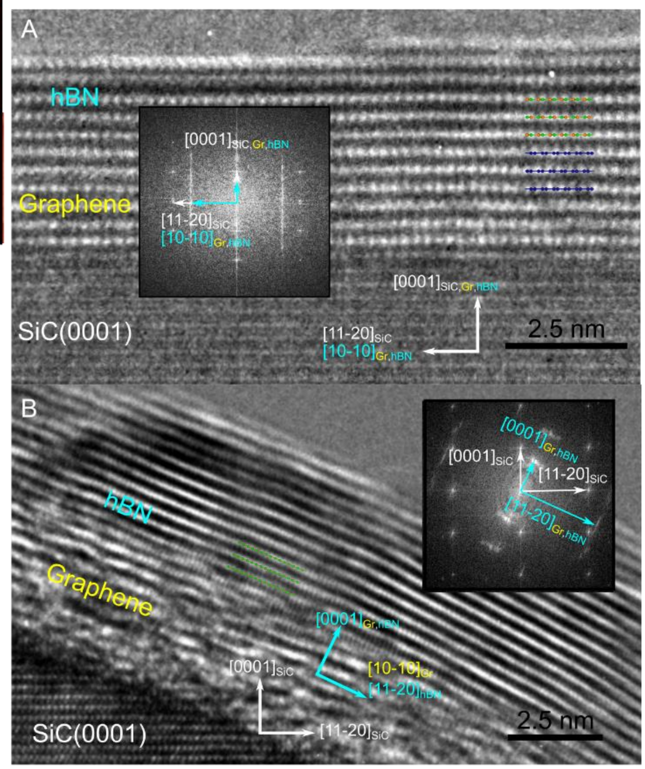

Figure 4: HRTEM of EG/SiC(0001) in the <10-10>SiC zone axis for "high-flux" growth, resulting in hBN aligned to the EG (A), and "low-flux" growth, resulting in hBN 30° rotated from the EG (B). Insets show the FFT of the images to confirm the different orientations of hBN.