

Imaging and Spectroscopy of Graphene/Hexagonal Boron Nitride Lateral Heterostructure Interfaces

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Recently, we demonstrated heteroepitaxy in two-dimensional space with a 1D sharp interface in a prototypical material system: graphene and hexagonal boron nitride (BN) [1]. Using microscopy probes on samples staying on the growth substrate, we established lattice coherence between the graphene seed and the BN monolayer grown from the edge of the seed. Atomic-resolution, element-contrast (Z-contrast) scanning transmission electron microscope (STEM) imaging of the boundary is desirable to reveal the presumably atomically sharp graphene-BN interface, but is not easily obtainable because contaminations introduced during the transfer process preferably reside over boundaries.

In this study, we examined the contaminant-covered graphene-BN boundary using electron energy-loss spectroscopy (EELS) in an aberration-corrected STEM, a Nion UltraSTEMTM 100, equipped with a cold field emission electron source, a corrector of third and fifth order aberrations, and a Gatan Enfina spectrometer [2]. The microscope was operated at 60 kV acceleration voltage to avoid knock-on damage of the graphene and BN lattices. A semi-convergence angle of 30 mrad, and 54 to 200 mrad semi-collection angles were used to obtain the medium angle annular dark field (MAADF) images. The EEL spectrum map were collected with an energy resolution of ~350 meV and with an energy dispersion of 40 meV/pixel.

Figure 1 (A) shows an experimental MAADF image of the graphene-BN boundary. Fast Fourier Transform of the areas shown in (A) indicates that the BN is aligned with the graphene monolayer. Evidence of a sharp interface is provided by the chemical map shown in (E), where the boron K-edge clearly defines a sharp graphene-BN interface, establishing EELS as a powerful “see-through” tool.

Fig 2 shows intensity profiles along the yellow lines of Fig 1. The left panel of Fig 2 shows that the graphene-BN boundary is composed of monolayer graphene and monolayer BN. The right panel shows a transition width of 0.5 nm between graphene and BN as determined from the boron K-edge signal.

A direct observation of the boundary at atomic resolution requires a reliable method to free the graphene-BN interface of contaminants. We will also discuss our effort to clean the buried interface by in-situ annealing. This method produces regions of thousands of nanometer squares of clean graphene and could be applied to other two dimensional (2D) materials.

References:

[1] L Liu *et al*, *Science* **343** (2014), p. 163.

[2] OL Krivanek *et al*, *Ultramicroscopy* **108** (2008), p. 179.

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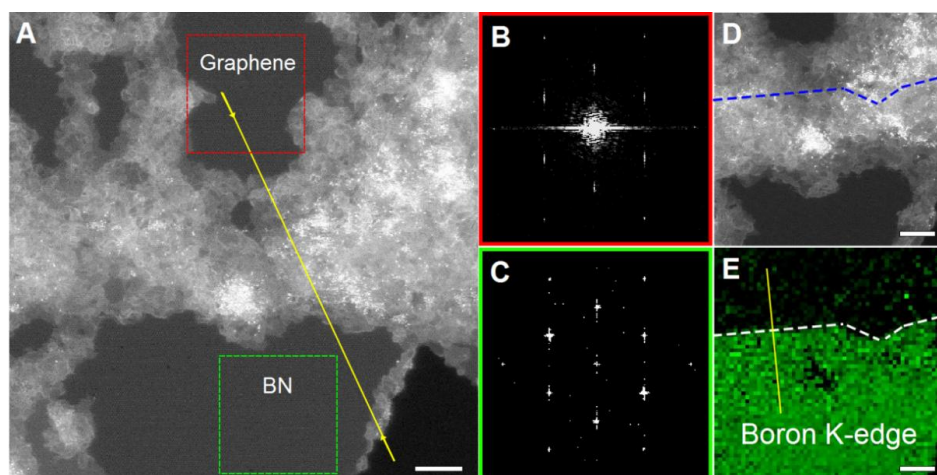


Figure 1. STEM-MAADF image and EELS. (A) MAADF image of a graphene-BN boundary. (B) and (C) fast Fourier transforms (FFT) of the highlighted areas shown in (A). (D) and (E) simultaneously acquired STEM image and electron energy loss spectrum map (boron K-edge) of a region containing a buried graphene-BN boundary, respectively. The blue/white dashed lines indicate the boundary between the graphene and the BN. Scale bars are 5nm. Profiles in Fig. 2 are taken along yellow lines in (A) and (E). Adapted from Ref. [1].

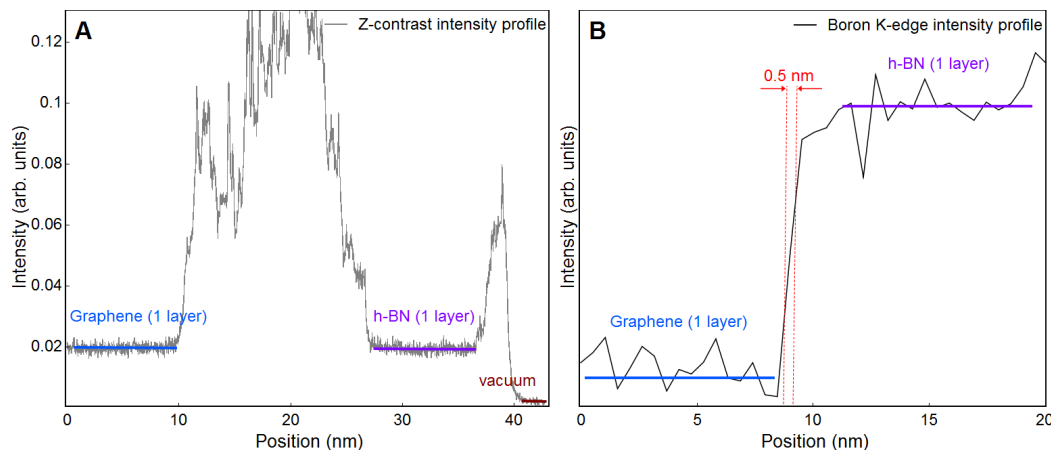


Figure 2. Intensity and EELS profiles. (A) Intensity profile along the yellow line in Fig. 1. The intensity profile indicates that the boundary is composed of monolayer graphene and BN. The intensity profile was obtained by integrating the signal across the interface within a width of 1.2 nm. (B) Boron K-edge intensity profile along the yellow line in Fig. 1. The boundary is sharp with a transition width of 0.5 nm. The transition width is defined from 25% to 75% of the values of the boron K-edge signal across the graphene-BN interface. The intensity profile was obtained by integrating the signal across the interface within a width of 4.8 nm. The spatial resolution is 0.5 nm. Adapted from Ref. [1].