Functional Materials

MS.3.036

Cross sectional imaging of high mobility graphene heterostructure devices

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Keywords: graphene, FIB, TEM

Recent progress in combining different 2D crystals in a single electronic device has produced new structures with unique properties. Multilayer heterostructures combining separate layers of graphene with hexagonal boron nitride are of particular interest with potential application in next generation high speed electronics [1-3]. Imaging of suspended 2D crystals using transmission electron microscopy (TEM) and scanning transmission electron microscopy (STEM) has provided fundamental scientific insights into defect formation, edge structure, doping and plasmonic properties [4,5]. However, when many separate 2D crystals are superimposed in projection, these plan view TEM images become difficult to interpret. Scanning tunnelling microscopy of 2D materials on surfaces has demonstrated that their behaviour is critically dependant on the presence of neighbouring atomic species [6]. This technique allows correlation of atomic surface structure with spectroscopic measurement of the local electronic density of states for 2D crystals on different substrates but is not suited to the study of encapsulated interfaces. Consequently little is known about the behaviour of 2D crystals when covered by additional layers and encapsulated deep below the surface. The increasing complexity of the latest devices requires a new approach to characterising two dimensional materials when layered into multi-component systems.

We have studied multilayer graphene/BN heterostructures where single layer and bilayer graphene sheets are individually contacted and encapsulated between insulating spacer layers. The multilayer stack begins with a thick (~ 50 nm) BN flake deposited on top of an oxidised Si wafer. Each subsequent layer is prepared on a separate wafer, de-bonded and transferred on top of the target device. The freshly deposited layer can be then shaped by reactive plasma etching and annealed (300°C in Ar/H_2) in order to remove processing residues. Because the graphene layers are separated by insulating BN spacers, each of them can be connected with Au/Ti contacts and the electrical properties studied independently [1]. Charge transport properties of these devices depend critically on the separation of the graphene layers and transport measurements therefore provide a route for estimation of the spacer layer thickness. However, more detailed structural data is highly desirable.

Here we report on recent work using a Focused Ion Beam (FIB) Scanning Electron Microscope (SEM) to extract thin cross sectional slices from the active region of several multi-layered graphene – based heterostructure devices with the desired location identified using secondary electron SEM imaging. The extracted cross sections have been imaged at high resolution using aberration corrected STEM combined with chemical analysis using electron energy loss spectroscopy and energy dispersive x-ray spectroscopy. Our side view observations (Fig 1) confirm the location of individual graphene sheets encapsulated between the separate BN layers and also allow precise measurement of interlayer spacings and interface roughness. Our side view imaging approach also allows comparison to the electrical device characteristics of individual layers measured before slice extraction. Through analysis of the local interlayer separation and layer roughness we observe a clear correlation between interface roughness and the electronic quality of encapsulated graphene [7]. We also report on the cross sectional imaging of devices that incorporate other 2D crystal such as tungsten disulphide and which have recently demonstrated significant improvements in device characteristics [8]. This work proves the concept of heterostructures assembled with atomic layer precision and reports their first side view TEM images. This novel approach offers the potential for correlation of measured transport properties for individual layers with local structural data and therefore provides a route to better understanding of the unique properties of 2D crystals when integrated into complex devices.

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9. The authors gratefully acknowledge funding from the EPSRC under grant number EP/G035954/1 and from the Defense Threat Reduction Agency.

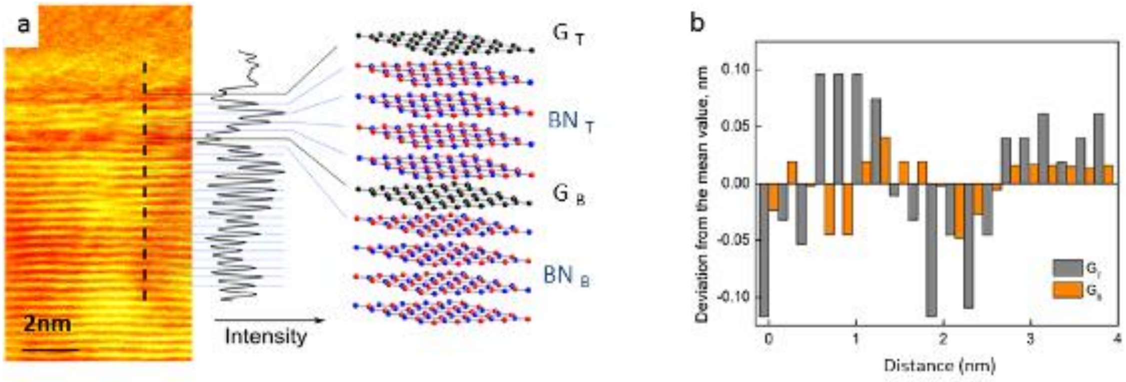

Figure 1. High resolution side view imaging of graphene/BN heterostructure devices. (a) Aberration corrected STEM image and schematic structure for one of our graphene heterostructured devices in which two independently contacted graphene flakes (G_T and G_B) are separated by a four layer boron nitride spacer (BN_T) and supported on a 20nm BN substrate (BN_B). (b) Estimate of the atomic roughness for each graphene layer using the deviation of layer position from horizontal.

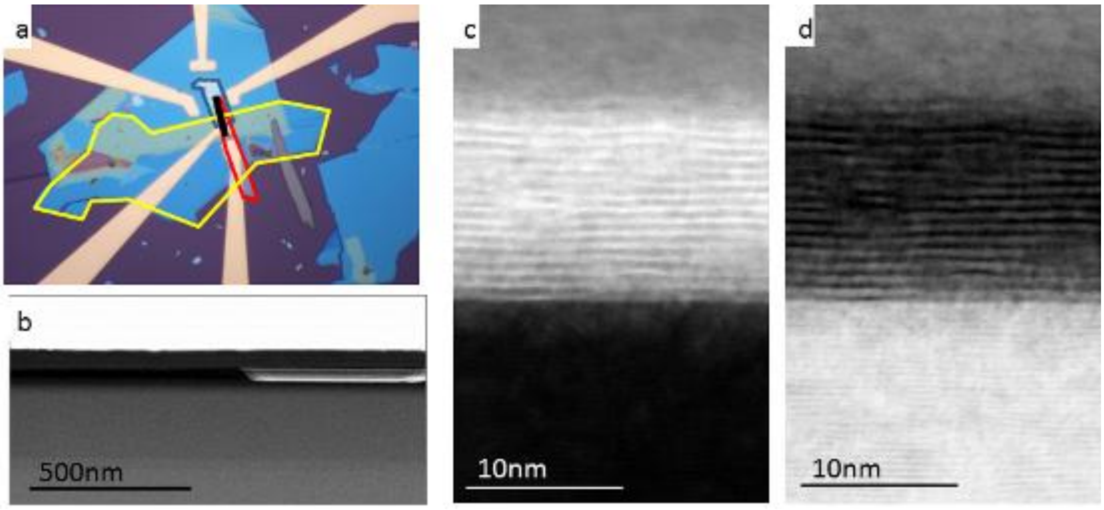

Figure 2. Imaging the encapsulated tungsten disulphide tunnelling barrier between two graphene sheets. (a) Optical image of device before extracting the cross section. High angle annular dark field STEM images showing (b) termination of tungsten disulphide layer within the slice and (c) lattice planes in barrier layer. (d) The complementary bright field STEM image for (c)