

TEM Imaging and Electron Diffraction of Vertically Stacked Graphene/h-BN with Fine Control of Twist Angle

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Vertical heterostructures incorporating two-dimensional (2D) atomically-thin crystals have attracted significant research interest owing to their novel physical properties arising from interlayer interaction [1]. Relatively weak van der Waals force is a main interaction between layers, which enables preparation of isolate layers and fabrication of vertical heterostructure through various bottom-up approaches. The twist angle between neighboring layers in a vertical stack is a degree of freedom which strongly influences the various properties. In particular, a vertical heterostructure can display in-plane atomic-scale reconstruction associated with inhomogeneous strain due to a competition between the in-plane strain energy and inter-layer stacking energy, especially for near-commensurate interlayer stacking condition. The process to build vertical structure with fine control of twist angle is therefore of critically importance.

We fabricated graphene/hexa-boron nitride (graphene/h-BN) vertical heterostructures with fine tuning of interlayer twist angle. For demonstration purpose, we prepared graphene/h-BN stacks with 0.0° interlayer twist angle by mechanical exfoliation and transfer. (Figure 1) During this stacking process we utilized decorated microwires on 2D crystals as rotational alignment markers. Our previous study demonstrated that atomic chain molecules, AuCN, can assemble to form nanowires on graphene and the axis of nanowires is aligned along the zigzag lattice direction of graphene [2,3]. Similarly, AgCN chains can assemble on various 2D crystals via simple drop-cast method to form microwires. We found that the axis of microwires are also aligned along zigzag lattice directions of hexagonal 2D crystals. The aligned microwires can be observed by optical microscope and serve as rotational alignment markers during the mechanical stacking process. Twist-angle-controlled graphene/h-BN heterostructures were prepared on holey Si₃N₄ TEM grids for TEM analysis.

Figure 2 shows selected area electron diffraction (SAED) and phase-contrast TEM image of a graphene/h-BN heterostructure. Strongly overlapped diffraction spots from graphene and h-BN indicate that we achieved the twist angle of nearly zero degree through our stacking process as shown in Figure 2(a). High-index diffraction spots of graphene/h-BN exhibit the original h-BN and graphene spots together with satellite peaks. The graphene/h-BN moiré pattern was also observed using phase-contrast TEM imaging as shown in Figure 2(c). The experimentally measured moiré periodicity was 14.2 nm, which is in agreement with an expected value with 0.0° twist angle between graphene and h-BN. Furthermore, the uniform moiré pattern over the observed region indicates that 2D interface between graphene and h-BN is pristine. (Figure 2(c)). We believe that our process of angle-controlled stacking is reliable and can be extended to build heterostructures using various 2D crystals, including transition metal dichalcogenides (TMDCs).

References:

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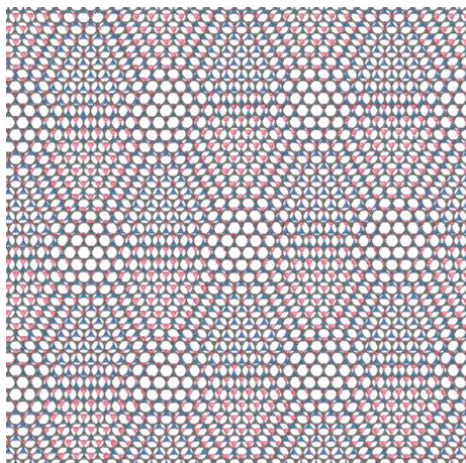


Figure 1. Atomic structure of graphene/h-BN heterostructure with 0.0° interlayer twist angle. The mismatch between two lattices is exaggerated ($\sim 9\%$)

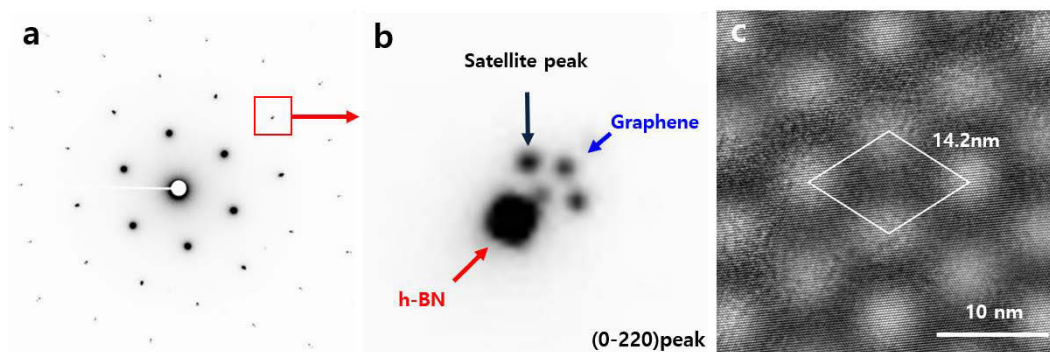


Figure 2. Electron diffraction pattern and phase-contrast TEM image of graphene/h-BN stack. (a) SAED of graphene/h-BN heterostructure with zero twist angle. (b) Zoomed-in diffraction signal in red box. (c) Phase-contrast TEM images of the graphene/h-BN heterostructure. The experimentally measured moiré wavelength was consistent with moiré wavelength of zero twist angle alignment.