S/TEM Characterization of Vertical Heterostructures Formed by Mono- to Multi-layer Graphene and WSe2

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Two-dimensional (2D) transition metal dichalcogenides (TMD) are emerging semiconductor materials in the field of nano optoelectronics owing to their desirable physical and electronic properties [1]. They are touted to replace silicon in the sub-5 nm regime where silicon suffers from quantum confinement effects [2]. Hence, there is a great interest in growing large area defect-free electronic grade TMD materials towards target optoelectronic applications. A recent development in the synthesis of TMD materials is the use of 2D growth substrates such as graphene and hBN as alternatives to traditional sapphire and silicon. Compared to the traditional substrates, 2D substrates offer potential advantages such as step edge free flat surfaces, absence of dangling bonds etc. [3]. Moreover, 2D substrates could enable true van der Waals epitaxy with the as-grown TMD with relaxed lattice matching conditions [4]. In addition, usage of 2D substrates could lead to the formation of 2D vertical heterostructures with enhanced optoelectronic properties and exciting physical phenomena [5, 6]. For example, H. Buch et al. have observed superlubricity in epitaxial graphene-WS2 vertical heterostructures [6].

In this work, we investigate the nucleation of WSe2 synthesized on mono- to multi-layer graphene via MOCVD process. The mono- to multi-layer graphene was first prepared using chemical vapor deposition (CVD) technique on Cu substrate and then transferred to sapphire substrate prior to the WSe2 growth. As-grown heterostructures are characterized using scanning/transmission electron microscopy (S/TEM) imaging to understand the atomic structure of the heterostructures, epitaxy between graphene and WSe2 layers and the role of defects in graphene as the underlying mechanism behind the nucleation of WSe2 on Graphene.

Figure 1a shows a scanning electron microscope (SEM) image of WSe2 triangles (~ 500 nm) synthesized on CVD grown multilayer graphene after the MOCVD step. Layer numbers of graphene are labeled in the image. It can be observed that the nucleation density of WSe2 triangles is increasing with the graphene layer number. Selected area diffraction (SAD) pattern obtained from the as-grown heterostructures in monolayer graphene area is presented in Figures 1b. It is evident that WSe2 triangles (green hexagon) maintain an epitaxial relationship with the graphene layer (blue hexagon). Figures 2 shows the atomic resolution images of graphene and WSe2. While Figure 2a is a monochromated high-resolution TEM image taken from monolayer graphene region, Figure 2b is an atomic resolution high-angle annular dark-field (HAADF)-STEM image taken from a WSe2 triangle. The presentation will further uncover the role of the substrate and epitaxy, graphene layer number, and substrate defects on the nucleation density of WSe2 [7].
Figure 1. (a) SEM image of WSe$_2$ triangles grown on multilayer CVD graphene; the labels indicate the number of graphene layers and (b) SAD pattern obtained from heterostructures grown on the monolayer graphene area showing epitaxy between graphene and WSe$_2$.

Figure 2. (a) Monochromated HRTEM image from monolayer graphene region; the numbers in the image indicate the number of graphene layers and (b) atomic resolution HAADF-STEM image of WSe$_2$ grown on the monolayer graphene region.

References
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