

## Monolayer-like Behavior of Bilayer Transition-Metal Dichalcogenides

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Following the emergence of two-dimensional (2D) materials with extraordinary properties, van der Waals (vdW) heterostructures have great attention, due to the novel properties and wide range of applications in electronic devices [1]. Understanding interfaces in the heterostructures is particularly important, because interfaces intrinsically play a crucial role in growth and microstructure of the epilayer and obtaining desirable properties. For example, during the WS<sub>2</sub> growth on graphene substrate, WS<sub>2</sub> grown on defective graphene indicates metallic property via W atomic bridges between WS<sub>2</sub> and graphene layers, despite semiconducting feature of WS<sub>2</sub> [2].

It is well known that when W(Mo)S<sub>2</sub> flakes are grown on a graphene substrate, the orientation of W(Mo)S<sub>2</sub> is predominantly oriented along the graphene, indicating vdW epitaxy growth [3]. Herein, we demonstrate interface-driven WS<sub>2</sub> AB/AC stacking boundaries (SBs) using the WS<sub>2</sub>/wrinkled graphene heterostructure. Surface defects in graphene, such as wrinkles and grain boundaries, play a role as active nucleation sites for WS<sub>2</sub> multilayers due to high chemical reactivity [4]. In the case of graphene wrinkles, they simultaneously impede lateral growth of epilayers due to the additional friction from the graphene wrinkles.[5] Additional stresses from wrinkle-driven friction induce Shockley partial dislocations for reducing stress, resulting in SBs in multilayer WS<sub>2</sub>. (Figure 1) For releasing partial dislocation-driven in-plane strain, out-of-plane buckling is facilitated with 1 nm height, energetically favorable process. (Figure 2a) Electron energy-loss spectroscopy (EELS) data shows red-shift of  $\pi+\sigma$  plasmon peak at the SB compared to the bilayer WS<sub>2</sub>, like the monolayer WS<sub>2</sub>. (Figure 2b,c) It indicates monolayer-like behavior at buckled SB due to the reduced interlayer interaction. These findings show the effect of substrate morphology even in vdW epitaxy growth and indicate that the potential for various applications with desirable manipulation of the materials using extended 1D defects.

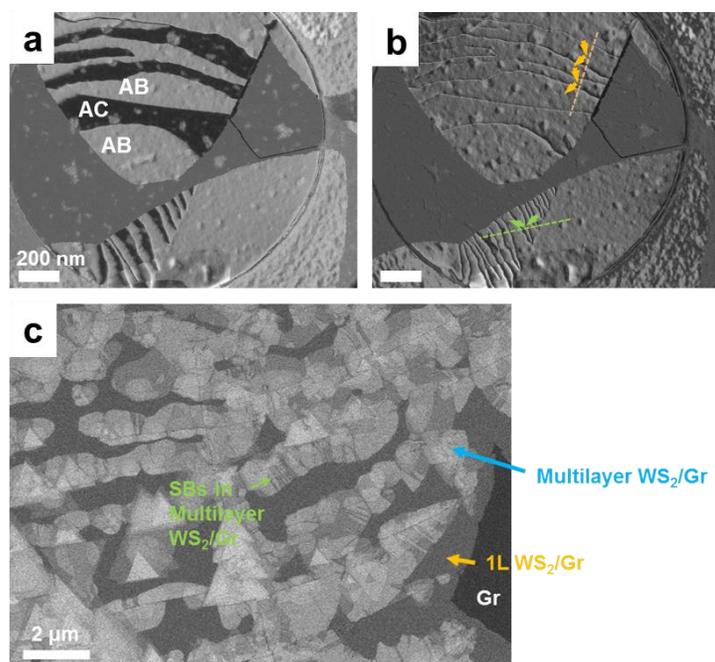
SBs are observed using scanning electron microscopy (SEM), transmission electron microscopy (TEM), and atomic force microscopy (AFM). Dark-field and atomic-resolution TEM modes are used for analyzing Burgers vectors and atomic structure, respectively. TEM images were obtained at an acceleration voltage of 80 kV using an FEI Titan cube G2 60-300, equipped with image- and probe-aberration correctors and a monochromator. EELS analysis was performed using Gatan Quantum 965 dual EELS system with energy resolution of 1.0 eV.

### References:

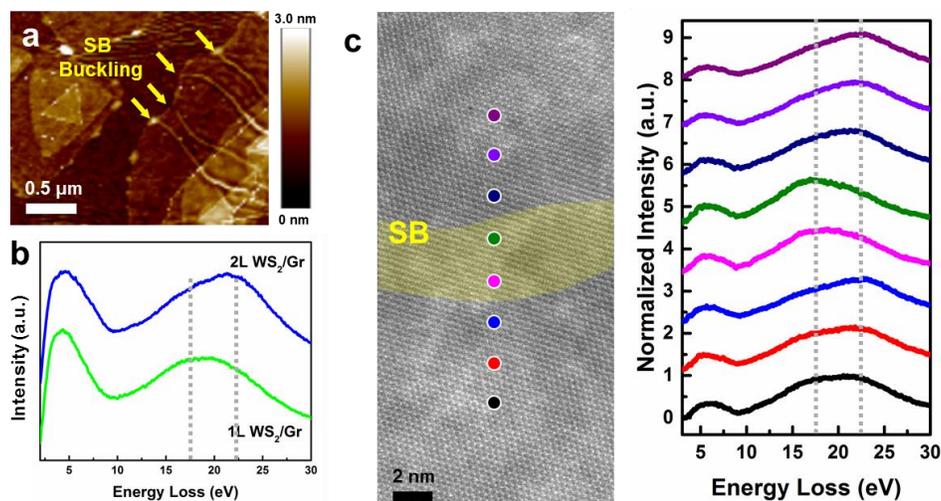
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**Figure 1.** (a-b) Dark-field TEM images obtained by (a)  $(0\bar{1}10)$  (1<sup>st</sup>) and (b)  $(\bar{2}110)$  (2<sup>nd</sup>) diffraction spots, respectively. (a) and (b) indicate stacking order and layer number, respectively. Orange and green arrows in burgers vector of partial dislocations using adjacent 2<sup>nd</sup> order dark-field images. (c) SEM image using backscattered electron mode, showing stacking order. SBs are formed near the graphene wrinkles.



**Figure 2.** (a) Height image using AFM, showing 1 nm of vertical height at SBs. (b-c) EELS line profile across the SB [5].