

Atomic-scale Observation of Grains and Grain Boundaries in Monolayers of WS₂

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Transition metal dichalcogenides (TMDs), i.e. MoS₂ and WS₂, are fascinating materials due to their optical, electronic and catalytic properties. Monolayers of semiconducting TMDs possess a direct band gap and large optical absorption [1,2] ideal for constructing novel optical and electronic devices. In addition, MoS₂ and WS₂ have a high catalytic activity, and a wide range of physiochemical properties [3,4]. Such applications arise from the scalable synthesis of atomically thin TMD sheets via chemical vapor deposition (CVD). Grain boundaries (GB) and dislocations are inevitable defects in 2D layered crystals synthesized by the CVD [5]. Therefore, it is important to image and probe defects and their atomic and chemical structure before they can be nano-engineered.

Because of the unique three-atomic thickness in monolayers of TMDs, defects behave very differently from those observed in 2D monatomic films (i.e. graphene). In addition, this sandwich structure can lead to unique dislocations and different grain boundary structures and dynamics. We use ultra-high resolution aberration-corrected electron microscope (TEAMI) with sub-Ångstrom resolution in order to understand the atomic structure of GB and dislocations in WS₂ monolayers. Annular dark field (ADF) STEM images of GB in WS₂ show the atomic structure of the CVD-grown films [6]. The material contains monolayer, bilayer and trilayer regions of WS₂ with different stacking orders, and the layers display a slight rotational mismatch with respect to each other (Fig 1A). At 80 kV, we observe W atoms to be highly stable under the electron beam, while S atoms show significant movement around the edges and near the dislocation cores (see Fig 1). We investigated the structure of GB in monolayered WS₂ as well as their stability and dynamics under the electron beam. Figure 1D shows a tilt GB in monolayer WS₂ with a few edge dislocations along the GB. We identify that GB have a 6|8 dislocation component, which is consistent with that reported in predictions on TMDs under S-rich conditions, rather than the formation of the 5|7 dislocation [7]. This study further employs first principles calculations to elucidate the dislocation migration mechanisms in monolayers of WS₂. Our experimental observations coupled with theoretical simulations indicate that it is possible to achieve a high dislocation mobility in WS₂ which leads to the efficient reconstruction of GB.

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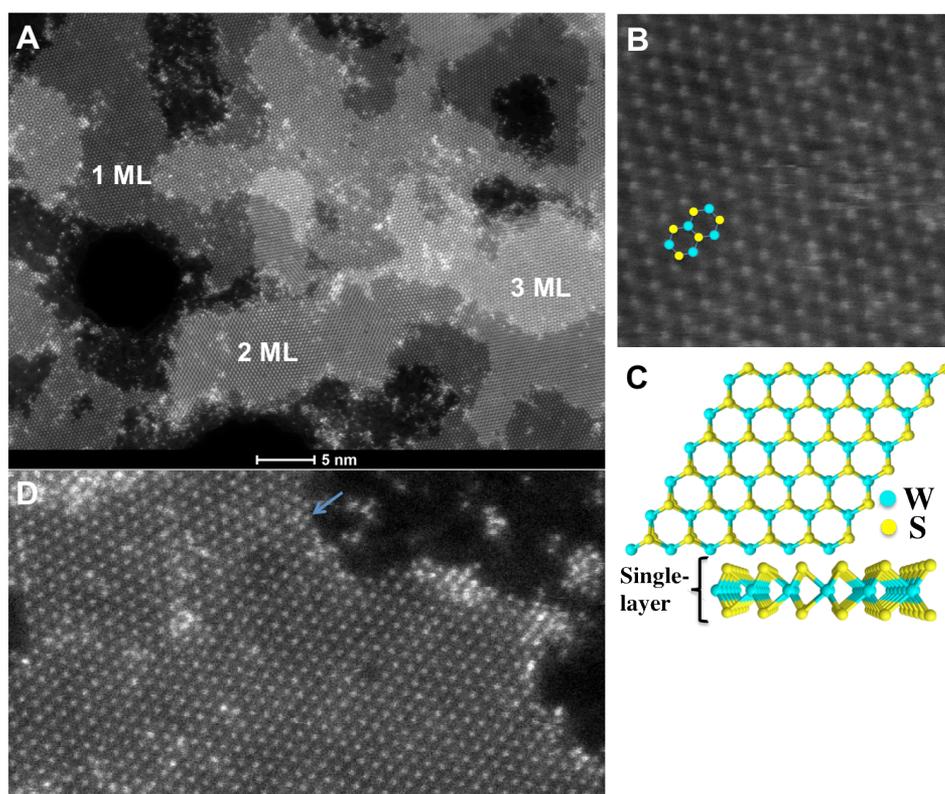


Figure 1. (A) The annular dark-field (ADF)-STEM image of a WS₂ film with monolayer, bilayer and trilayer regions, (B) Atomic-resolution ADF-STEM image of a single-layer WS₂, indicating W and S atoms, (C) the top and side view of a monolayer WS₂ film, and (D) ADF-STEM image of a tilt grain boundary in a single-layer WS₂