Microsc. Microanal. 21 (Suppl 3), 2015
doi:10.1017/S1431927615001324 Paper No. 0053

Interfaces in Two-Dimensional Heterostructures of Transition Metal Dichalcogenides

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Two-dimensional (2D) transition-metal dichalcogenides (TMDs) are promising candidates for flexible nanoelectronics, with exceptional optical and electrical properties at monolayer thickness. Monolayers of different TMDs can be further combined to create van der Waals heterostructures, where multiple 2D layers are stacked vertically layer-by-layer, or stitched seamlessly in plane to form lateral heterojunctions. The coupling between the different 2D components provides unique opportunities for bandgap engineering and can create very unusual properties at the interface [1-4]. Revealing the atomic structure, including the stacking orientation, stacking order, and chemical inter-diffusion, is therefore important for understanding the novel properties generated by the heterostructure interfaces.

Recently, we have demonstrated a simple one-step vapor phase growth of high quality heterostructures of WS_2 and MoS_2 [1]. High temperature growth yields predominantly vertically stacked bilayer heterostructures, while low temperature growth creates mostly lateral heterostructures of WS_2 and MoS_2 within the same monolayer. The atomic structure and electronic properties of the heterostructure interfaces are studied by aberration-corrected scanning transmission electron microscopy (STEM) annular dark field (ADF) imaging, electron energy-loss spectroscopy (EELS) at low voltage, and density functional calculations,

STEM-ADF imaging reveals that the vertical heterostructures were obtained with WS_2 epitaxially grown on top of the MoS_2 monolayer, following the preferred 2H stacking (Figure 1). A small amount (~ 3%) of W substitution in the MoS_2 layer and Mo substitution in the WS_2 layer was observed in the sample, which should only have minimum effect on the properties of the MoS_2 and WS_2 monolayers at such low concentration. Photoluminescence (PL) analysis shows that the MoS_2 and WS_2 layers in the bilayer heterostructure, on one hand, behave as individual monolayers, and, on the other hand, generate a new direct band gap of WS_2/MoS_2 heterostructure via interlayer coupling owing to the clean interface.

Atomically sharp interfaces were frequently observed in the lateral heterojunctions, with seamless connection and abrupt transition between the MoS_2 and WS_2 lattice within a single atomic row. Most of the abrupt lateral interfaces were achieved by lateral epitaxial growth of WS_2 on fresh MoS_2 edges along the zigzag direction, and sharp armchair interfaces were only occasionally observed. Lateral interfaces with large chemical inter-diffusion over a width of a

few hundred nanometers were also observed, presumably due to local fluctuations in the growth conditions. The different degrees of chemical inter-diffusion are most likely responsible for the observed inhomogeneous PL enhancement along the lateral interfaces.

Besides the WS_2-MoS_2 system, results from WSe_2/$MoSe_2$ heterostructures will also be discussed, which provides insights into the growth mechanism and guidance for the growth of superlattice structures [5].

References:

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[5] This research was supported in part a Wigner Fellowship of Oak Ridge National Laboratory (WZ), by the Office of Science, Basic Energy Science, Materials Sciences and Engineering Division, U.S. DOE (WZ), by U.S. DOE grant DE-FG02-09ER46554 (JL, STP), and through a user project supported by ORNL's Center for Nanophase Materials Sciences (CNMS), which is sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, U.S. DOE. YG and PMA were supported by Army Research Office MURI grant W911NF-11-1-0362.

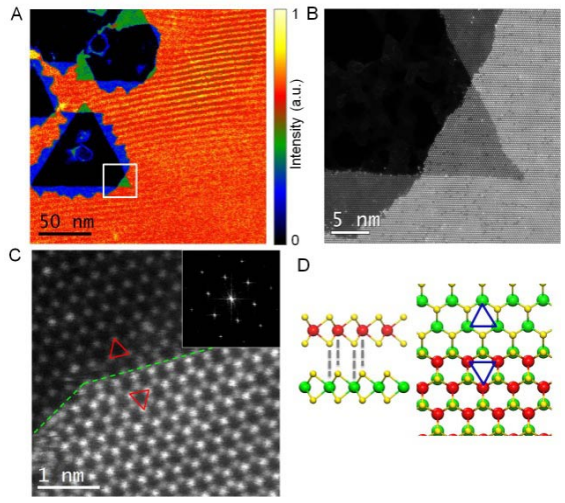

Figure 1. STEM-ADF imaging of vertical heterostructure of WS_2/MoS_2 at different magnifications. Figure A is shown in color scale where monolayer MoS_2 is in blue, monolayer WS_2 in green and WS_2/MoS_2 bilayer in orange. (D) is the structure model illustrating the 2H stacking [1].

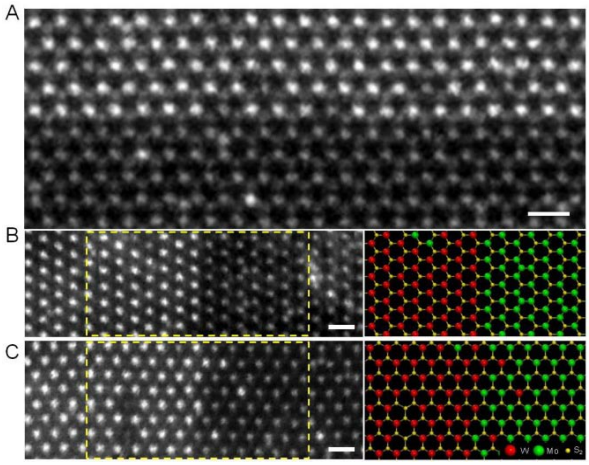

Figure 2. STEM-ADF imaging of atomically sharp lateral interfaces between WS_2 and MoS_2 along the zigzag (A, B) and armchair (C) directions. Scale bars: 0.5 nm [1].