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 WS₂ and MoS₂ [1]. High temperature growth yields predominantly vertically stacked bilayer heterostructures, while low temperature growth creates mostly lateral heterostructures of WS₂ and MoS₂ 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 WS₂ epitaxially grown on top of the MoS₂ monolayer, following the preferred 2H stacking (Figure 1). A small amount (~ 3%) of W substitution in the MoS₂ layer and Mo substitution in the WS₂ layer was observed in the sample, which should only have minimum effect on the properties of the MoS₂ and WS₂ monolayers at such low concentration. Photoluminescence (PL) analysis shows that the MoS₂ and WS₂ layers in the bilayer heterostructure, on one hand, behave as individual monolayers, and, on the other hand, generate a new direct band gap of WS₂/MoS₂ 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 MoS₂ and WS₂ lattice within a single atomic row. Most of the abrupt lateral interfaces were achieved by lateral epitaxial growth of WS₂ on fresh MoS₂ 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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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].