**Epitaxy of transition metal dichalcogenides: The route to wafer-scale single crystal monolayers and heterostructures**

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**Introduction:** Transition metal dichalcogenides (TMDs) form a compelling class of 2D materials with potential applications in optoelectronics, flexible electronics, chemical sensing and quantum technologies. At the monolayer limit, the semiconducting TMDs (*e.g.*, MX2 where M = Mo/W and X = S/Se) exhibit direct band gaps within the visible range, large exciton binding energies and valley polarization. The lack of out-of-plane bonding on the van der Waals surface of these materials enables heterostructure formation without the constraints of lattice matching. Current research on TMD monolayers is often carried out using flakes exfoliated from bulk crystals which is a limiting factor for device fabrication.

**Aims:** Our research is aimed at the development of an epitaxial growth technology for TMDs based on metalorganic chemical vapor deposition (MOCVD). The high growth temperatures (>700oC) and chalcogen/metal precursor ratios (>10,000) attainable in an MOCVD environment are advantageous for promoting metal surface diffusion needed for epitaxy while maintaining growth stoichiometry.

**Methods:** Our studies employ metal hexacarbonyls and hydrides (H2S, H2Se) as precursors in an H2 carrier gas. Substrates include c-plane sapphire and hBN flakes. A multi-step precursor modulation growth method was developed to independently control nucleation density and the lateral growth rate of monolayer domains on the substrate for growth on 2” diameter sapphire substrates [Zhang 2018].

**Results and Discussion:**  Using this approach, uniform, coalesced monolayer and few-layer films of WS2, MoS2, WSe2 and MoSe2 were obtained on 2” sapphire at growth rates on the order of 0.02-0.1 monolayer/min. The films are epitaxially oriented with respect to the sapphire as determined by in-plane x-ray diffraction [Chubarov 2018]. Post-growth dark-field transmission electron microscopy carried out on WS2 monolayers removed from the sapphire demonstrate that the films are nominally single orientation with translation boundaries that arise from coalescence of WS2 domains with lattices that are offset by sub-unit cell lengths. WS2 domains nucleate at step edges on the c-plane sapphire substrate which imparts a preferential orientation to the domains. The WS2 monolayers exhibit intense room temperature photoluminescence at ~2.0 eV and variable-temperature measurements reveal PL associated with excitons and trions with negligible defect-related emission. The field effect mobility in the WS2 monolayer, as measured at room temperature in back-gated devices that employ a 50 nm Al2O3 dielectric, is in the range of 20-30 cm2/Vs.

Van der Waals epitaxy of TMDs on hBN and TMD monolayers has also been investigated. In the case of WSe2 epitaxy on hBN, single atom B and N vacancies in the hBN can trap metal atoms and thereby serve as nucleation sites for the WSe2 domains [Zhang 2019]. The vacancy-assisted nucleation also imparts directionality to the WSe2 domains which results in a significant decrease in inversion domain in the films. Epitaxial growth of vertical TMD heterostructures will also be discussed.

**References**

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