MOCVD of Fully-Coalesced WS_2 Monolayers and W/WS_2 Heterostructures

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Abstract

MOCVD has evolved as mainstream technique for large-scale production of 2D TMDC (transition metal dichalcogenide) materials. Despite fruitful works reporting on the successful synthesis of various types of TMDC with excellent performance in prototype devices, details of growth processes are not fully unveiled yet. For instance, as the coalescence of a monolayer (ML) is approaching, the probability of premature bilayer (BL) nucleation and growth will inevitably be rising. This phenomenon can be attributed to the limited supply of adatoms to lateral growth due to their finite migration length on top of ML domains. In order to fine-tune ML growth of 2D WS_2 films and suppress BL nucleation, a novel two-step migration-enhanced (ME) MOCVD process is introduced, which involves: (i) a nucleation stage at 700 °C and (ii) a lateral-growth stage at 820 °C with the W precursor ramped down to 25% of its initial flux.
All experiments were carried out on (0001)-oriented sapphire substrates in an AIXTRON CCS reactor in 7×2" geometry equipped with LayTec in-situ monitoring and surface temperature control. Pure H_2 was employed as the carrier gas, and $W(CO)_6$ and DTBS (di-tert butyl sulphide) were selected as metalorganic precursors. Prior to growth start, sapphire was H_2-desorbed at 1050 °C. 20 hPa was chosen as the reactor pressure during deposition to avoid parasitic C incorporation. With W adatoms being the growth-limiting species, a high S/W molar ratio (>6,000) was implemented.
By initializing the nucleation at 700 °C, a nucleation density of WS_2 as high as ~140 μm^{-2} can be achieved after 15 min, with a typical size of ML triangles ≤50 nm and a total ML coverage of ~24%. To suppress further ML nucleation, the lateral-growth stage is initiated by a temperature ramp of +70 K with +10 K/min. Simultaneously, the $W(CO)_6$ flux is ramped down (-75% over 144 min), aiming at reducing the arrival rate of W adatoms and thus increasing their migration length on the surface of the already-formed ML domains. By analysing the morphology, an estimate about the migration length on the coalesced ML can be concluded to be ≤100 nm. A fully-coalesced ML (>99%) with small BL coverage (<20%) can be obtained within 3 hours.
In the second part of the study, we present an alternative approach to the direct fabrication of metallic electrodes and conductive/reflective interlayers for TMDC devices by MOCVD and demonstrate the growth of both W films and W/WS_2 heterostructures. The underlying W thin film is deposited on sapphire via the decomposition of $W(CO)_6$ at 700 °C for 1 h (deposition rate roughly 5-10 nm/h). In the first experiment, a W/WS_2 heterostructure is synthesized via introducing both $W(CO)_6$ and DTBS into the reactor at 580 °C after W deposition in the same growth run. As indicated by Raman measurements and XPS (X-ray photoemission spectroscopy), the formation of a W/WS_2 heterostructure can be confirmed although the top WS_2 film still exhibits a similar nanocrystalline morphology as W. Although further optimization and characterization are required and ongoing, this study demonstrates the applicability of MOCVD to a large-scale fabrication of TMDC films and heterostructures for novel 2D devices.