Controlled Layer-by-Layer Growth of WSe_2 by Water-Promoted Metal-Organic Chemical Vapor Deposition

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Abstract

With the growing exploration of TMDC (transition metal dichalcogenides), a wide range of applications have been proposed. The direct deposition of multilayer TMDC structures incl. TMDC/TMDC heterostructures via MOCVD promises excellent scalability, a virtually contamination-free interface, and superior reproducibility.[1] However, commonly, TMDC growth strongly deviates from the ideal layer-by-layer mode. Parasitic bilayer (BL) nuclei form on the first TMDC monolayer (ML) before its coalescence. These nuclei serve as preferred growth sites during the subsequent deposition, resulting in increasing corrugation and deviations from planarity with each deposited layer. In this work, an optimized multi-step WSe_2 MOCVD process based on our migration enhancement approach[2] is chosen as the starting point. As an additive, ultra-pure water is introduced (molar ratio H_2O:WCO (initial value in WCO ramp-down stages) = 1:1). A WSe_2 ML sample with only ~ 2% BL coverage was prepared in 37 min growth time using a water-promoted multi-step MOCVD recipe (WP-MOCVD), significantly lower than ~15% BL w/o water introduction.[3] SEM (scanning electron microscopy) and AFM (atomic force microscopy) analyses were performed to validate the reduced BL formation. The effective migration length of W adatoms is defined as the radius of the largest circle fitting in regions free from BL nuclei on the already merged ML in SEM images. The effective migration length increased substantially from ~0.47 µm to ~1.27 µm with WP-MOCVD. In line with Ref[4], we attribute this to the formation of volatile species from a reversible reaction between metal (precursors) and H_2O.[4] To grow multilayer films, following ML coalescence, the temperature was reduced from 820 °C (lateral growth) to 720 °C (2nd nucleation) with H_2+DiPSe stabilization. Despite identical nucleation conditions, the density of BL nuclei on the ML reached only ~29 μm^{-2}, significantly lower than ~86 μm^{-2} for nucleation on sapphire. Obviously, uniform nucleation on a van der Waals (vdW) surface is more challenging than on the dangling-bond-rich sapphire. Extending lateral growth time without H_2O injection resulted in 87% BL coverage but was accompanied by 53% parasitic TL formation. Introducing H_2O improved BL coverage to 92% while suppressing TL formation to 34%. By further implementing a ramping-down growth stage, we achieved fully coalesced BL WSe_2 films, and the final TL coverage was restricted to ~40%. Further characterization (X-ray photoelectron, Raman and photoluminescence spectroscopy) confirmed that water does not significantly alter the WSe_2 properties. Back-gated FETs (field-effect transistors) are successfully fabricated using ML WSe_2. Based on the TLM method, the H_2O-promoted samples exhibited a p-type field-effect conductivity mobility of 0.35 cm^2/Vs. In comparison, WSe_2 FETs grown without water (with a different morphology) showed a slightly higher mobility of 0.43 cm^2/Vs, which is however within the typical variability of device processing. These data indicate that water injection does not seem to significantly alter the electrical properties of WSe_2, focusing its role on the suppression of parasitic nucleation.

References

[1] S.Cwik, et al., Advanced Materials Interfaces, 1800140 (2018), 5(16).
[2] S. Tang, et al., Crystal Growth & Design, 1547-1558 (2023), 23(3).
[3] Y. Wang, et al., MRS Bulletin, 50(11), 1283-1295.
[4] V. Voronenkov, et al., physica status solidi (a), 2300943 (2024), 221(9).