Wafer-Scale Epitaxial 2D Transition Metal Dichalcogenide Films by Metalorganic Chemical Vapor Deposition

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Abstract:
The field of two-dimensional (2D) materials began with the advent of graphene but has expanded to a wide class of materials that occur naturally as layered crystals. Within this class of materials, semiconducting transition metal dichalcogenides (TMDs) such as MoS_2, WS_2, and WSe_2 have attracted significant interest in condensed matter physics and next generation electronics and optoelectronics due to their unique electronic and optical properties. Furthermore, TMD layers with different compositions and properties can be stacked to create unique heterostructures. Practical device applications, however, require large area single layer epitaxial TMD films which presents unique challenges. Metalorganic chemical vapor deposition (MOCVD) emerges as a key technique for epitaxial TMD growths, enabling the use of high substrate temperatures (900-1000°C) and precise control over precursor flow rates with chalcogen/metal ratios (10^3-10^5). Our work has focused on the development of MOCVD as a manufacturing-compatible approach for wafer-scale epitaxial semiconducting TMDs.

Wafer-scale TMD monolayers such as MoS_2, WSe_2, and WS_2 were epitaxially grown on 2" diameter c-plane sapphire substates using MOCVD systems in the 2D Crystal Consortium Materials Innovation Platform (2DCC-MIP) facility at Penn State University. Molybdenum hexacarbonyl ($Mo(CO)_6$) and Tungsten hexacarbonyl ($W(CO)_6$) were used as the metal precursor while hydrogen sulfide (H_2S) and hydrogen selenide (H_2Se) were the chalcogen source with H_2 as the carrier gas. The one step (only lateral growth) and multi-step (nucleation, ripening, lateral growth) growth process were utilized to achieve coalescence followed by post growth annealing in H_2S to 300 °C to inhibit the decomposition of the obtained TMD films. Surface morphology, epitaxial crystallinity, and optical properties of the grown TMDs were systematically characterized by AFM, FESEM, TEM/4D-STEM, Raman, Photoluminescence, UV-VIS-IR absorption spectrum, in-situ Ellipsometry, and In-plane XRD. Characterization results show interesting effects of critical growing parameters, substrate surface chemistry, orientation and miscut angle of the sapphire substrates on the film growth and their optical and electrical properties. These endeavors facilitate a profound comprehension of the fundamental mechanisms of wafer-scale epitaxial 2D TMD films growth, thereby paving the way for the advancement of next-generation electrical and optoelectronic devices.