

Synthesis of two-dimensional transition metal dichalcogenides and heterostructures by physical vapor deposition

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Abstract: Two-dimensional (2D) transition metal dichalcogenides (TMDs) such as MoS₂ and WS₂ have attracted significant attention as semiconductor thin-film materials for optoelectronics, memristive devices, and energy applications. Their single- or few-layer forms exhibit direct bandgaps, strong light-matter coupling, and bandgap tunability through composition, layer number, and heterostructure engineering, properties inaccessible in their bulk counterparts. A key challenge in translating these materials to applications is achieving scalable, reproducible synthesis of continuous, high-quality films. In the space-confined microreactor method, two substrates are placed face-to-face in a horizontal configuration: the source substrate carries the precursor material, while the blank growth substrate is placed on top. Using a colloidal dispersion of MoS₂ nanoparticles in saline solution as the source material, this horizontal microreactor geometry produces few-layer MoS₂ nanosheets with lateral dimensions of 5-60 μm [1]. Growth proceeds by a Stranski-Krastanov mechanism, generating abundant undercoordinated edge sites well-suited for electrocatalytic applications such as solar water splitting, but yielding discontinuous isolated islands rather than continuous films. Obtaining continuous monolayer TMD films requires a solid thin-film precursor. We show that replacing the colloidal dispersion with a magnetron-sputtered monolayer W thin film as the source material in the same horizontal microreactor geometry enables large-scale synthesis of continuous monolayer WS₂ by low-temperature sulfurization [2]. While the sputtered-precursor approach yields continuous monolayer films, the horizontal reactor geometry is limiting film uniformity over large substrate areas. By replacing the microreactor with confined-space, we demonstrate the fabrication of high-quality MoS₂/graphene lateral heterostructure memristors [3]. An amorphous MoS₂ precursor is RF-sputtered directly onto CVD few-layer graphene and crystallized by confined-space sulfurization at 800°C, yielding residue-free 3-4 layer MoS₂. The MoS₂/graphene memristors exhibit reproducible non-volatile analogue resistive switching with a SET transition near +6 V and an ON/OFF ratio of ≈2.1, attributed to vacancy-induced Schottky-barrier modulation at the MoS₂/graphene interface. However, in vertical MoS₂/W precursor stacks, we find that sulfurization at 800°C does not produce discrete heterostructures but instead results in vertically graded W_xMo_{1-x}S₂ alloy films due to Mo-W interdiffusion at the high processing temperature. To overcome the limitations of physical deposition routes and enable reproducible, 2D material synthesis, we present a custom-designed automated vertical CVD system.

Keywords: MoS₂, WS₂, transition metal dichalcogenides, heterostructures, chemical vapor deposition, microreactor, magnetron sputtering, sulfurization, memristors

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