Domain orientation-controlled epitaxial growth of tungsten diselenide monolayers

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Semiconducting transition metal dichalcogenide (TMDs) monolayers have been of considerable interest for device applications. However, commonly used exfoliated flakes have size limitations and difficulties controlling the number of layers attained which limits their utility. Vapor phase techniques such as chemical vapor deposition (CVD) and metalorganic CVD (MOCVD) have been demonstrated as auspicious methods for the epitaxial growth of TMDs over wafer-scale areas. TMDs, however, exhibit a three-fold symmetry which typically results in two energetically equal domain orientation alignments at 0° and 60° when grown on common substrates such as sapphire, hBN and graphene. When these oppositely oriented domains coalesce during growth it produces inversion domain boundaries (IDBs) that exhibit metallic behavior which is undesired. Recent work has demonstrated that steps on the sapphire surface due to miscut can alter the energy landscape for nucleation and give rise to a preferred domain orientation for WS_2 [1] and MoS_2 [2] which enables a significant reduction in IDBs in wafer-scale TMD monolayers. In this work, we demonstrate that for WSe_2 grown by MOCVD on c-plane sapphire, growth conditions also play a significant role in domain orientation and can be used to control the crystallographic direction of domains on the sapphire substrate.

MOCVD growth was carried out in a stainless-steel cold-wall reactor using $W(CO)_6$ and H_2Se as precursors in a H_2 carrier gas. The substrates used for growth were 2" diameter c-plane (0001) sapphire with nominal miscut of 0.2+/- 0.1° toward the m-plane. A three-step nucleation-ripening-lateral growth process was used to control the nucleation density and lateral growth rate to achieve epitaxial films [3]. The effects of growth temperature (850°C to 1050°C) and reactor pressure (50 to 700 Torr) on surface coverage and domain alignment were investigated. In all cases, the WSe_2 grows epitaxially on the (0001) sapphire surface with an epitaxial relationship of $[11\bar{2}0]$// $[11\bar{2}0]$, as determined by in-plane x-ray diffraction. However, the preferred direction of the WSe_2 domains on sapphire was observed to change from 0° to 60° as the reactor pressure was raised from 200 Torr to 700 Torr holding all other parameters constant. The reactor pressure is believed to control the concentration of Se adsorbed on the sapphire which alters the surface chemistry and preferred nucleation site for the WSe_2. Further studies are underway to investigate this mechanism and determine the impact of preferred alignment on film properties.

1. M. Chubarov, et al., ACS Nano, 2021, 15, 2532-2541.
2. T. Li, et al. Nature Nanotechnol., 2021, https://doi.org/10.1038/s41565-021-00963-8.
3. X. Zhang, et al. Nano Lett, 2018, 18, 1049-1056.