

# Understanding the Growth Mechanisms and the Origins of Heterogeneity in Optoelectronic Two-Dimensional Materials

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Like carbon nanotubes and graphene before them, the development of semiconducting two-dimensional (2D) layered material “building blocks” for novel electronic and optical devices (notably the transition metal dichalcogenides (TMDs) and their heterostructures) faces significant synthesis and processing challenges. These challenges limit their development, including wafer-scale, bottom-up synthesis of uniform layers of crystalline 2D materials that are comparable in quality to exfoliated flakes of bulk materials. For example, these materials currently display remarkable heterogeneity on both the atomistic level, including vacancies, dopants, and edge terminations, and on the mesoscopic length scale involving misoriented grains, layer orientations, and interactions with substrates and adsorbates. This heterogeneity can strongly influence the structure and electronic properties in 2D materials, offering at the same time a serious challenge to synthesis control for reliable properties and a tremendous opportunity to tailor functionality.

Here we will present recent developments in both vapor-transport and laser-based synthesis and processing approaches for the synthesis of a variety of atomically-thin 2D crystals (e.g., MoSe<sub>2</sub>, WS<sub>2</sub>, Mo[1-x]W<sub>x</sub>Se<sub>2</sub>, GaSe) and understanding of mechanisms for the introduction of heterogeneity during growth, including the incorporation of defects and dopants, the role of topology-induced strain, and the orientation of layers during heterostructure growth by van der Waals epitaxy (e.g., GaSe/MoSe<sub>2</sub>).

We will review time-resolved, *in situ* diagnostic techniques to “watch nanomaterials grow”. Often this involves utilizing the same remote, laser spectroscopic tools that are used *ex situ* to provide assessment of defects, stacking, and functional properties for optoelectronic applications. Such techniques will be briefly reviewed, and others such as low-frequency Raman spectroscopy, low-temperature photoluminescence, and ultrafast pump-probe spectroscopy techniques will be correlated with atomic-resolution electron microscopy to understand the exact nature, concentration, and density of defects as well as the evolution of preferred edges associated with changing crystal shapes during growth and etching. Associated theory and modeling is used to infer the responsible synthetic driving forces. Transport measurements with prototype devices are correlated to understand the effects on functionality.

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