Mixed-Flake 2D Heterostructures via Langmuir Deposition for Tunable Optoelectronic Thin Films

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Heterostructures composed of two-dimensional (2D) materials offer unique opportunities to engineer optoelectronic properties by combining the distinct functionalities of different materials within a single thin-film system. While vertical stacking of mechanically exfoliated or CVD-grown layers has been widely explored, recent advances in solution processing now enable the fabrication of high-quality 2D materials in the form of dispersible flakes. This development paves the way for micrometer-scale lateral heterostructures, composed of interspersed flakes, which remain relatively underutilized despite their scalability and versatility.

Here, we present a fabrication strategy for compacted, laterally percolated 2D heterostructures based on electrochemically exfoliated graphene and MoS_2 flakes. Separate dispersions of each material were mixed and deposited using the Langmuir–Blodgett method to produce large-area flake films with controlled lateral compression. This technique enables edge-to-edge flake connectivity and a random but homogeneous in-plane distribution of semiconducting and conductive domains. The resulting films were characterized via scanning electron microscopy (SEM), Raman spectroscopy, and spatially resolved photoluminescence (PL) mapping. These analyses revealed lateral separation between the two materials, as evidenced by spatially anti-correlated Raman signatures of MoS_2 and graphene. Furthermore, PL mapping showed local variations in excitonic emission, with changes in the relative contributions of A^- trions, A excitons, and B excitons across the film. These results highlight the potential of this approach for engineering spatially heterogeneous optoelectronic properties in mixed-flake 2D material systems.

Figures

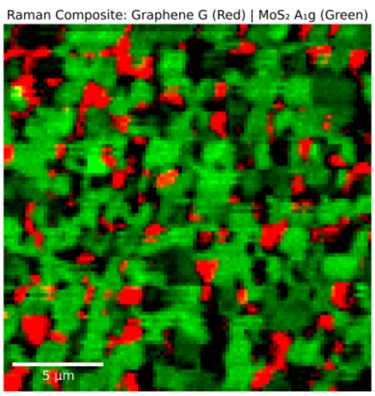

Figure 1: False-color composite map showing fitted Raman intensities of MoS_2 (A_1g mode, green) and graphene (G mode, red). Intensities were extracted via pixel-wise Lorentzian fits and filtered using plausibility-based thresholds to identify regions with significant material presence. The resulting overlay highlights spatial domains of MoS_2 and graphene.