Materials Science & Engineering Dissertation Defense

Deciphering the Intrinsic Structure-Property Correlations in2D Janus Transition Metal Dichalcogenides

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Abstract

Janus Transition Metal Dichalcogenides (TMDs) are emerging 2D quantum materials with an asymmetric chalcogen configuration that induces an out-of-plane dipole moment. Their synthesis has been a limiting factor in exploring these systems' many-body physics and interactions. This dissertation examines the challenges associated with synthesis and charts the excitonic landscape of Janus crystals by proposing the development of the Selective Epitaxy and Atomic Replacement (SEAR) technique.

SEAR utilizes ionized radical precursors to modify TMD monolayers into their Janus counterparts selectively. The synthesis is coupled with optical spectroscopy and monitored in real-time, enabling precise control of reaction kinetics and the structural evolution of Janus TMDs. The results demonstrate the synthesis of Janus TMDs at ambient temperatures, reducing defects and preserving the structural integrity with the hitherto best-reported exciton linewidth emission value, indicating ultra-high optical quality.

Cryogenic optical spectroscopy (4K) coupled with a magnetic field on Janus monolayers has allowed the isolation of excitonic transitions and the identification of charged exciton complexes. Further study into macroscopic and microscopic defects reveals that structural asymmetry results in the spontaneous formation of 2D Janus Nanoscrolls from an in-plane strain. The chalcogen arrangement in these structures dictates two types of scrolling dynamics that form Archimedean or inverted C scrolls. High-resolution scanning transmission electron microscopy (HR-STEM)of these superlattices shows a preferential orientation of scrolling and formation of Moiré patterns. These materials' thermodynamically favorable defect states are identified and shown to be optically active. The encapsulation of Janus TMDs with hexagonal Boron Nitride (h-BN) has allowed isolation defect transitions. DFT coupled with power-dependent PL spectroscopy at 4K shows the broad defect band to be a convolution of individual defect states with extremely narrow linewidth (2 meV) indicative of a two-state quantum system.

The research presents a comprehensive synthesis approach with insights into the structural and morphological stability of 2D Janus layers, establishing a complete structure-property correlation of optical transitions and defect states, broadening the scope for practical applications in quantum information technologies.

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