2D Atomic and Molecular Lattices: Design, Synthesis, and New Properties

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Abstract:

The Kempa group develops rational chemical solutions to challenges in materials research. This talk will present our advances in the preparation and analysis of 2D atomic and molecular lattices supporting advanced and highly tunable properties. Atomic Lattices: 2D transition-metal dichalcogenide (TMD) crystals are a promising platform for optoelectronic, catalytic, and quantum device studies. However, explicit synthetic control of their morphology and dimensionality is a major challenge. We recently demonstrated a gas-phase synthesis method that significantly transforms the structure and dimensionality of TMD crystals. Synthesis of MoS$_2$ on Si(001) substrates pre-treated with phosphine (PH$_3$) yields high aspect ratio nanoribbons which are straight and have exceptionally uniform widths. Notably, we systematically control the width of these nanoribbons between approximately 50 nm and 500 nm by varying the total PH$_3$ dosage during the Si substrate treatment step. Moreover, these quasi-1D MoS$_2$ nanocrystals produce photoluminescence which is 50 meV higher in energy than that from 2D MoS$_2$ crystals and is progressively tunable through synthetic control of the crystal width. Finally, extensive synthetic studies and cluster expansion calculations suggest that the nanoribbons grow following heterogeneous nucleation at the edge of nanoscale seed crystals, which are uniquely stabilized on the pre-treated Si surface. Molecular Lattices: Metal-organic frameworks (MOFs) are versatile materials that have been used as tunable scaffolds for energy storage, catalysis, and separations. We are focused on developing new approaches for the synthesis and characterization of stimuli responsive 2D MOFs. We demonstrated the synthesis of 2D MOFs comprised of molecular complexes containing strongly-coupled di-Mo cores. These materials exhibit anomalous gas adsorption characteristics, redox activity, and photo-tunable charge transport. Notably, we demonstrate the versatility of chemical vapor deposition and the unique opportunities this method presents for the preparation of layered 2D MOFs. We show that single crystal device studies allow for not only detailed investigation of charge transport mechanisms within these materials, but also in situ identification of the unique response of these MOFs to optical, electronic, and chemical stimuli. Collectively, our studies underscore the importance of rational synthesis in elaborating materials with unique and prescribed properties.