Materials Science and Engineering Dissertation Defense

Highly Kinetic Deposition Technique and Phase Engineering of 2D Transition Metal Dichalcogendies, Janus Structures, and Next-Generation High-Mobility Semiconductor

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Abstract

Two-dimensional (2D) layered materials have emerged as promising candidates to overcome the challenges faced by scaling silicon-based field-effect transistors (FETs) below the sub-10 nanometer technology node. Among them, van der Waals (vdW) transition metal dichalcogenides (TMDs) have attracted considerable attention for their ability to be thinned down to a single monolayer while maintaining a robust bandgap. Alternatively, bismuth oxyselenide (Bi_2O_2Se), a quasi-2D layered semiconductor, has shown exceptional electronic carrier mobility, environmental stability, and importantly, the formation of a high-κ native oxide (Bi_2SeO_5) with atomically sharp interface.

Synthesis and processing techniques for these layered materials were developed using chemical and physical vapor deposition approaches. Synthesis of semiconducting and metallic TMD monolayers, namely WS_2, MoS_2, NbS_2, was achieved using atmospheric-pressure chemical vapor deposition (APCVD) and mechanical exfoliation. The as-synthesized monolayers are converted into Janus counterparts using non-thermal plasma technique known as selective epitaxy atomic replacement (SEAR) and kinetically driven pulsed laser deposition (PLD). The conversion replaces the top chalcogen layer with a different chalcogen species, thereby breaking the mirror symmetry along the c-axis and introducing new phononic and excitonic characteristics.

A comprehensive growth process for high-quality, continuous thin film Bi_2O_2Se was developed using reactive pulsed laser deposition (RPLD). By ablating a Bi_2Se_3 target in a controlled O_2 atmosphere, Bi_2O_2Se grew between 350 and 400 °C, temperatures compatible with back-end-of-line (BEOL) CMOS processing. Fine-tuning substrate temperature, background pressure, and laser fluence further influences the formation of distinct structural and oxide variants of Bi_2O_2Se not previously reported. The deposition parameter is optimized across different substrates (a-SiO_2, c-Al_2O_3, and $SrTiO_3$), revealing the effect of substrate crystal structure on the quality of Bi_2O_2Se. FET devices fabricated using epitaxial Bi_2O_2Se grown on $SrTiO_3$ demonstrated efficient gate control and excellent current driving capability. The RPLD-grown Bi_2O_2Se were further oxidized into Bi_2SeO_5 and chalcogenized into Bi_2X_3 (X = S, Se, Te) using CVD-based thermal treatments, demonstrating the versatility of Bi_2O_2Se for phase engineering into adjacent family groups. Together, these results establish RPLD as a robust and scalable pathway for synthesizing high-quality Bi_2O_2Se thin films and provide a baseline for development of doping, alloying, and device integration in next-generation nanoelectronics.

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