Materials Science & Engineering Doctoral Defense

Layer Structured Gallium Chalcogenides: Controlled Synthesis and Emerging Properties

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abstract

Layer structured two dimensional (2D) semiconductors have gained much interest due to their intriguing optical and electronic properties induced by the unique van der Waals bonding between layers. The extraordinary success for graphene and transition metal dichalcogenides (TMDCs) has triggered a constant search for novel 2D semiconductors beyond them. Gallium chalcogenides, belonging to the group III-VI compounds, are a new class of 2D semiconductors that carry a variety of interesting properties including wide spectrum coverage of their bandgaps and thus are promising candidates for next generation electronic and optoelectronic devices. Pushing these materials toward applications requires more controllable synthesis methods and facile routes for engineering their properties on demand.In this dissertation, vapor phase transport is used to synthesize layer structured gallium chalcogenide nanomaterials with highly controlled structure, morphology and properties, with particular emphasis on GaSe, GaTe and GaSeTe alloys. Multiple routes are used to manipulate the physical properties of these materials including strain engineering, defect engineering and phase engineering. First, 2D GaSe with controlled morphologies is synthesized on Si(111) substrates and the bandgap is significantly reduced from 2 eV to 1.7 eV due to lateral tensile strain. By applying vertical compressive strain using a diamond anvil cell, the band gap can be further reduced to 1.4 eV. Next, pseudo-1D GaTe nanomaterials with a monoclinic structure are synthesized on various substrates. The product exhibits highly anisotropic atomic structure and properties characterized by high-resolution transmission electron microscopy and angle resolved Raman and photoluminescence (PL) spectroscopy. Multiple sharp PL emissions below the bandgap are found due to defects localized at the edges and grain boundaries. Finally, layer structured $GaSe_{1-x}Te_x$ alloys across the full composition range are synthesized on GaAs(111) substrates. Results show that GaAs(111) substrate plays an essential role in stabilizing the metastable single-phase alloys within the miscibility gaps. A hexagonal to monoclinic phase crossover is observed as the Te content increases. The phase crossover features coexistence of both phases and isotropic to anisotropic structural transition.Overall, this work provides insights into the controlled synthesis of gallium chalcogenides and opens up new opportunities towards optoelectronic applications that require tunable material properties.

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