Multilayer Van der Waals Heterostructures: Opportunities Beyond the 2D Limit

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Heterostructures of two-dimensional (2D) crystals have attracted broad interest due to novel properties that arise as atomically thin materials are stitched laterally or stacked vertically. Potentially as interesting scientifically and technologically, but much less explored are synthetic heterostructures between few-layer or multilayer van der Waals crystals. Major challenges exist in growth, doping, controlled interface formation, and the probing of emerging properties of such layered heterostructures at the relevant (nanometer) scale.Here, we discuss recent research that addresses these challenges by combining microscopy of growth and processing with advanced materials characterization and spectroscopy of optoelectronics at the ultimate resolution limit. We focus on group IVA monochalcogenides, MX (where M = Ge, Sn; X = S, Se), an emerging class of isostructural layered semiconductors with bandgaps between 0.9 – 1.6 eV. Lateral heterostructures combining multilayer SnS/GeS or SnS/GeSe show stunning interface control with atomically sharp layer-by-layer connectivity across hundreds of individual van der Waals layers [1–3]. Combined cathodoluminescence spectroscopy and ab-initio calculations shed light on carrier transfers and charge separation at such lateral interfaces. Doping of MX semiconductors has been a long-standing issue due to ubiquitous self-doping by native point defects. Overcoming this problem through controlled substitutional doping has paved the way for the realization of electrically active pn-junctions in lateral heterostructures [4]. In vertical heterostructures, new opportunities arise for valleytronics via valley-selective carrier transfers across planar van der Waals interfaces [5], and for templating ferroelectric order between different MX crystals [6]. Our results highlight the rich functionalities that can be realized in both lateral and vertical van der Waals heterostructures beyond the 2D limit.

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