

Kinetically-driven assembly of TaS₂-SnS heterostructures with flexible stacking architectures

D. M. Roberts,¹ **D. Bardgett**,^{1,2} **B. Gorman**,³ **J.D. Perkins**,¹ **A. Zakutayev**,¹ **S. R. Bauers**¹

¹ National Renewable Energy Laboratory, 15013 Denver West Pkwy, Golden, CO 80401

² University of Oregon, 1253 University of Oregon, Eugene, OR 97403

³ Colorado School of Mines, 1500 Illinois St, Golden, CO 80401

Chalcogenide heterostructures enable access to a remarkable range of materials properties by virtue of structural flexibility at the nanoscale. An interesting approach to the preparation of such heterostructures utilizes layered amorphous precursor films designed to mimic a desired superlattice heterostructure product, and has been successful in creating a wide range of metastable selenide heterostructures. An example of such tunable properties is seen in the SnSe-VSe₂ system, where changing layer sequencing can both enhance and quench charge density wave (CDW) transitions, as well as modify their onset temperatures.[1] Such CDWs in TaS₂ are hysteretic and have been proposed for phase change memory applications,[2] and so incorporating TaS₂ into superlattice structures known to enhance CDW transitions might further enhance this material towards these applications.

Previous work from our group developed a framework for preparing amorphous sulfide precursor films by RF sputtering.[3] Here we present the first crystalline sulfide heterostructures prepared using this precursor approach, composed of tunable numbers of SnS sandwiched between monolayers of TaS₂ in a superlattice. We also demonstrate the utility of combinatorial synthesis for creating precursors of several different stacking sequences in a single deposition. Structural measurements confirm smooth films with superlattice ordering as well as the presence of constituent TaS₂ and SnS layers. Superlattice architecture is confirmed by high resolution transmission electron microscopy (TEM) and EDS, resolving atomically precise sequencing of SnS and TaS₂ monolayers in the arrangement experimentally instilled into the precursors. Electronic behavior as a function of number of SnS layers is measured.

[1] R. Atkins, Chem. Mater. **26** (2014) [2] M. Yoshida, Sci. Rep. **4** (2014) [3] D. M. Roberts, JVST B **37** (2019)

⁺ Author for correspondence: dennice.roberts@nrel.gov

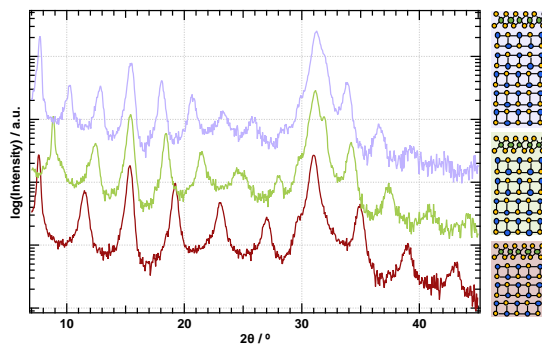


Figure 1 X-ray diffraction pattern of films with varied number of SnS layers between TaS₂ layers. Structure schematic shown at right.

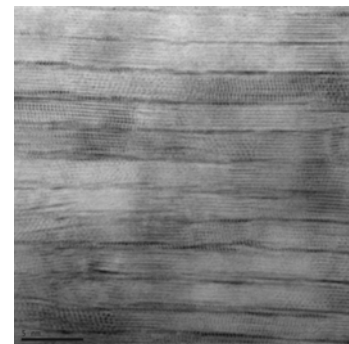


Figure 2 Hall coefficient and resistivity of heterostructures as a function of number of SnS layers.