Measuring and then eliminating twin domains in SnSe thin films using a fast optical metrology and molecular beam epitaxy

W. Mortelmans,[1] **M. Hilse**,[2] **Q. Song**,[1] **S.S. Jo**,[1]
K. Ye,[1] **D. Liu**,[2] **N. Samarth**,[2] **R. Jaramillo**[1]

[1] *Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts, 02139*

[2] *Department of Materials Science and Engineering, The Pennsylvania State University, University Park, Pennsylvania 16802*

Van der Waals (vdW) layered chalcogenides have strongly direction-dependent properties that make them interesting for certain photonic and optoelectronic applications. Orthorhombic tin selenide (α-SnSe) is a triaxial vdW material with strong optical anisotropy within layer planes, which has motivated studies of optical phase and domain switching. As with every vdW material, controlling the orientation of crystal domains during growth is key to reliably making wafer-scale, high-quality thin films, free from twin boundaries. Here, we demonstrate a fast and easy optical method to quantify domain orientation in SnSe thin films made by molecular beam epitaxy (MBE). The in-plane optical anisotropy results in white-light being reflected into distinct colors for certain optical polarization angles and the color depends on domain orientation. We use our method to confirm a high density of twin boundaries in SnSe epitaxial films on MgO substrates, with square symmetry that results in degeneracy between SnSe 90° domain orientations. We then demonstrate that growing instead on a-plane sapphire, with rectangular lattice-matched symmetry that breaks the SnSe domain degeneracy, results in single-crystalline films with preferred orientation, with twin domains all-but-eliminated. Our SnSe bottom-up film synthesis by MBE is enabling for future applications of this vdW material that is particularly difficult to process by top-down methods. Our optical metrology is fast and easy and can apply to all triaxial vdW materials.

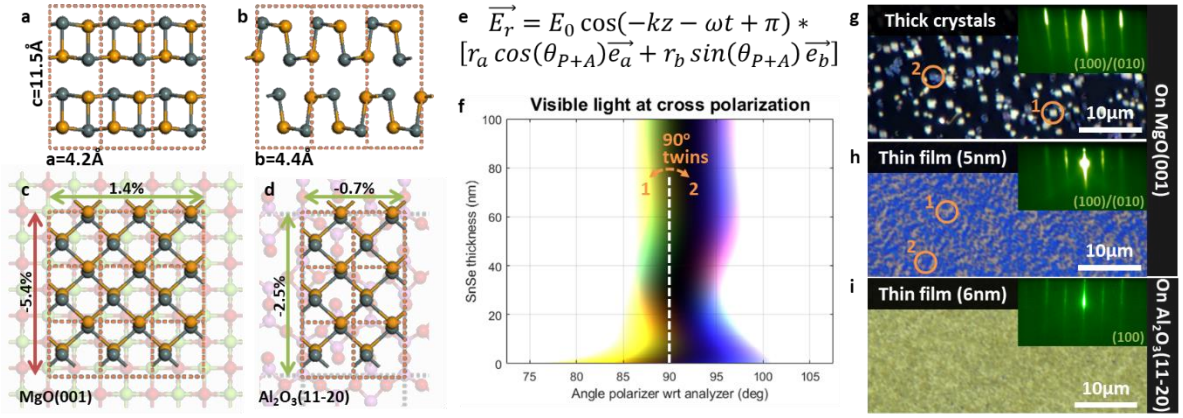

Figure 1: a-b) Cross-section of SnSe in (a) zigzag and (b) armchair direction. c-d) Top-view of SnSe on (c) MgO(001) and (d) Al_2O_3(11-20). The symmetry mismatch on MgO favors 90° twins, the symmetry/lattice match on a-plane sapphire favors single-crystalline epitaxy. e-f) Optical model for (e) reflection of white polarized light giving (f) yellow/blue contrast for 90° rotated domains. g-i) Self-heterodyned Kerr reflectometry on (g) thick crystals and (h) thin film on MgO and (i) thin film on Al_2O_3 confirming the optical model, the 90° twins on MgO and the single-crystallinity on sapphire.

+ Author for correspondence: wmortel@mit.edu

Supplementary Page

To further confirm the validity of the developed optical model and the single crystallinity of the SnSe epitaxy on a-plane sapphire, an in-plane reciprocal space map (RSM) is presented in Figure 2. This map highlights the various in-plane lattice parameters of 4.242Å in the zigzag direction and of 4.334Å in the armchair direction, and this in agreement with the anticipated unique epitaxial relation of [100] SnSe (001) // [1-100] Al_2O_3 (11-20).

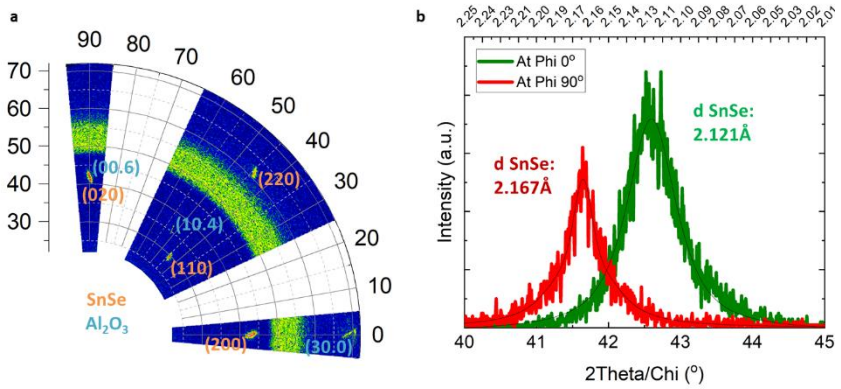

Figure 2: a) RSM of 6nm-thick orthorhombic SnSe epitaxially grown on Al_2O_3(11-20) highlighting the single-crystalline epitaxy and absence of 90° twins. b) 2Theta/Chi-Phi scans at Phi=0° (green) and Phi=90° (red) emphasizing the various in-plane lattice parameters.