

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

W. Mortelmans^{1,*}, M. Hilse², Q. Song¹, S.S. Jo¹, K. Ye¹, D. Liu², N. Samarth², 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

Corresponding authors: wmortel@mit.edu, rjaramil@mit.edu.

Van der Waals (vdW) layered chalcogenides have strongly direction-dependent (i.e., anisotropic) 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.