Supplementary Figures

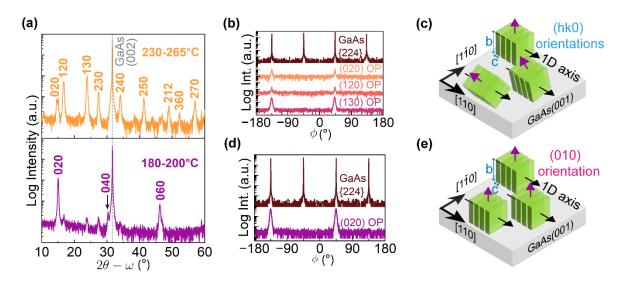


Figure S1. (a) Symmetric 2θ-ω out-of-plane (OP) scans of Sb₂Se₃ films on GaAs(001). Growth temperatures of 230–265 °C promote mixed (hk0)-type orientations such as (020), (120), and (130), which run parallel to the 1D axis or *c*-axis. 180–200 °C suppresses the growth of several (hk0) domains in favor of the (010) orientation. (b) Phi scans of a 230 °C film indicate Sb₂Se₃ (020)-, (120)-, and (130)-OP domains are aligned in-plane to GaAs [110]. For each OP orientation, only two 180°-separated film peaks arise in phi due to orthorhombic two-fold symmetry preserved in-plane. The asymmetric reflections probed are (120) for the (020)-OP orientation, and likewise (020) for the other dominant (120)- and (130)-OP orientations. (c) Schematic diagram of in-plane "rotated" texture with varying OP orientations and IP alignment of the Sb₂Se₃ 1D axis to GaAs [110]. (d) Phi scan of 200 °C epitaxial film shows in-plane alignment to GaAs [110]. Using the (120) asymmetric reflection, two 180°-separated film peaks arise from the orthorhombic Sb₂Se₃ two-fold symmetry. (e) Schematic diagram of epitaxial structure with a (010)-OP orientation and IP alignment of the Sb₂Se₃ 1D axis to GaAs [110].

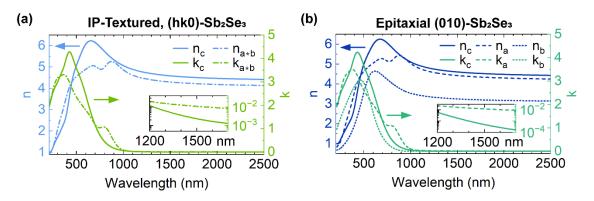


Figure S2. Large optical anisotropy is reflected in the near-infrared refractive indices and extinction coefficients (n, k) of (a) in-plane textured and (b) epitaxial (010)-orientation Sb₂Se₃ films. Insets show low extinction coefficients across 1200–1700 nm on a log scale. The in-plane indices for (a) in-plane textured Sb₂Se₃ are n_c , n_{a*b} . The in-plane indices for (b) epitaxial-Sb₂Se₃ are n_c , n_a and the out-of-plane index is n_b . The out-of-plane birefringence $(n_b - n_c)$ in epitaxial films exceeds 1.0 at telecom bands.

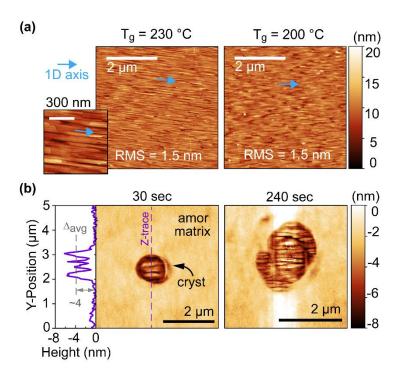


Figure S3. (a) Atomic force microscopy images of in-plane textured Sb_2Se_3 ($T_g = 230$ °C) and epitaxial Sb_2Se_3 ($T_g = 200$ °C) as-grown films. Both films exhibit ribbon-like structures extending along the 1D axis of Sb_2Se_3 (direction marked with blue arrow). The inset on the left (0.95 x 0.95 μ m² scan area) shows the ribbon morphology of the 230 °C film more closely. (b) Atomic force microscopy images of locally laser-crystallized regions of Sb_2Se_3 on GaAs using 633 nm, ~8.6 kW/cm² excitation. A ribbon-like morphology (similar to that of as-grown films) within the central crystalline region is tracked at 30 sec and 240 sec. The Z-trace (in purple) shows local height depression due to densification upon crystallization.