Supporting Information

Growth Mechanisms of Anisotropic Layered Group IV Chalcogenides on van der Waals Substrates for Energy Conversion Applications

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Supporting Figures

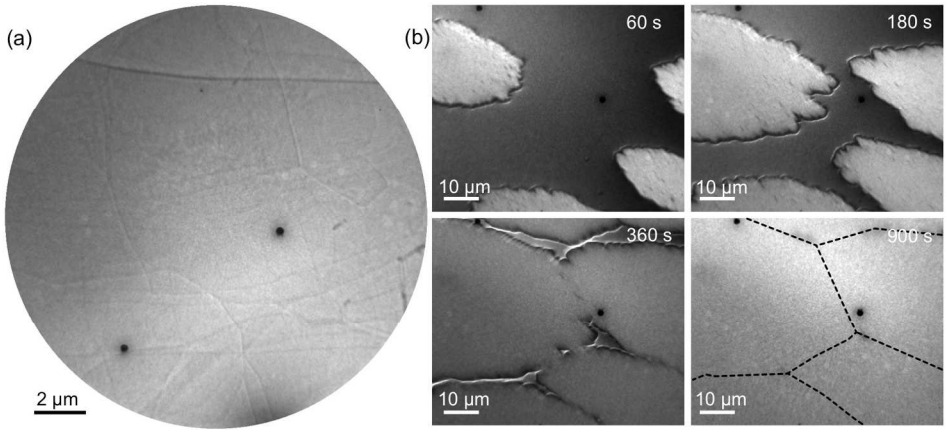

Figure S1: Van der Waals substrates used in this work. (a) Low-energy electron microscopy (LEEM) image of the starting graphite (HOPG) surface prepared by mechanical exfoliation followed by degassing in ultrahigh-vacuum. **(b)** Growth of monolayer graphene on Ru(0001) by exposure to ethylene at high temperature. The LEEM image series shows sparse nucleation of graphene, which translates into large domain size and correspondingly low density of domain boundaries, which are the primary defects in high-quality graphene/Ru(0001) films. Temperature: 840°C.

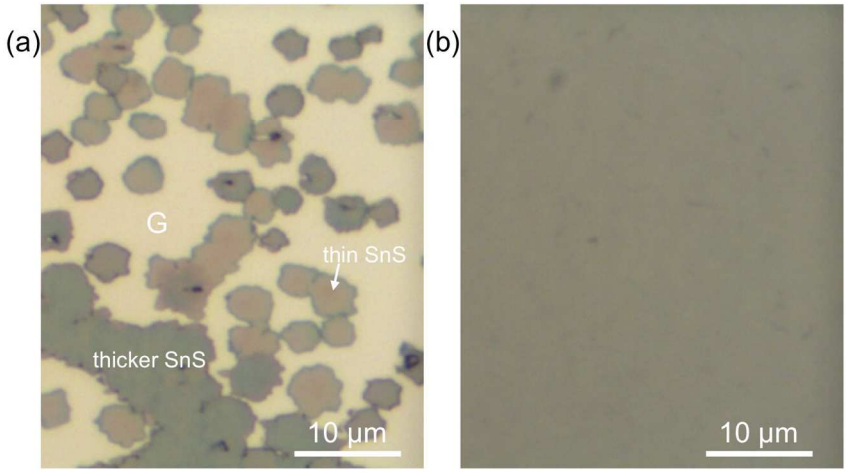

Figure S2: Partially and fully coalesced basal-plane oriented SnS film on monolayer graphene/Ru(0001). (a) Optical micrograph showing the morphology and size of single-crystalline SnS domains at partial surface coverage. **(b)** Optical micrograph at full surface coverage of coalesced SnS, which forms a dense layer with uniform thickness and basal-plane orientation.

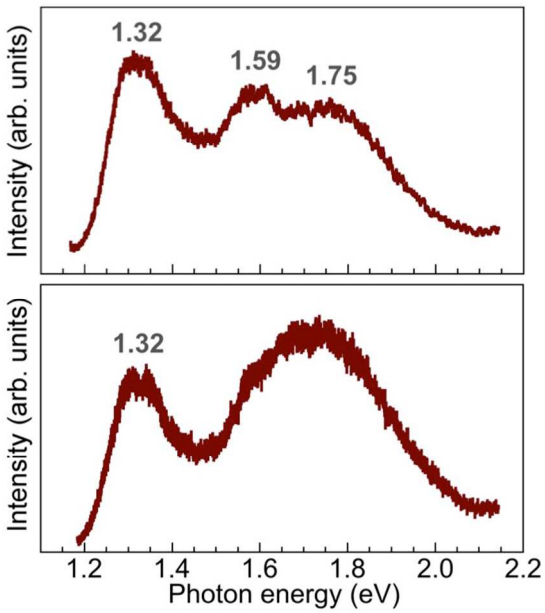

Figure S3: Photoluminescence (PL) spectroscopy of high-quality basal-plane oriented SnS grown on a graphene/Ru vdW substrate. Typical room temperature PL spectra obtained on different samples, showing a characteristic emission at a photon energy hv = 1.32 eV, along with several higher energy transitions.

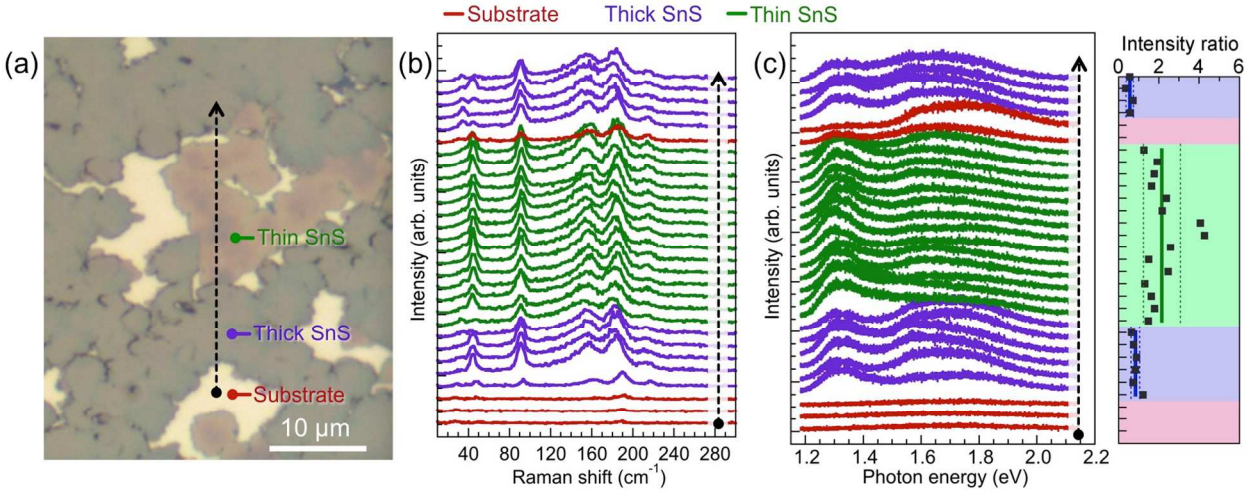

Figure S4: Raman and photoluminescence (PL) line scans across thick and thin SnS. (a) Optical micrograph of partially coalesced SnS grown on graphene/Ru. **(b)** Raman spectra measured along the line marked in (a). **(c)** PL spectra measured along the line marked in (a). The right-hand panel summarizes the analysis of the ratio of PL intensity between the band-edge peak (1.32 eV) and the broader peak at higher energy (1.6 – 1.8 eV), showing ~2 times higher intensity of the band-edge peak for thin SnS and intensity ratios below 1 for thicker SnS. Black symbols represent the measured data. Solid lines indicate the mean of all measured points in the same shaded region. Dashed lines represent $\pm 1\sigma$ (standard deviation). All measurements were performed at room temperature.

Supporting Movies (captions)

Movie S1: MBE growth of SnS on graphite at 260°C. Corresponding to the image sequence shown in Fig. 2 a. Real-time low-energy electron microscopy (LEEM) movie; original capture rate: 1 frame per second (every second frame is shown). Elapsed time is given in seconds.

Movie S2: SnS growth on monolayer graphene/Ru(0001) at 280°C. Corresponding to the image sequence shown in Fig. 3 a. Real-time LEEM movie, capture rate: 1 frame per second. Elapsed time is given in seconds. Electron energy ε = 5.8 eV.

Movie S3: Edge-flow growth of few-layer SnS on monolayer graphene/Ru(0001) at 280°C. Dark lines are edges of individual SnS atomic sheets. Real-time LEEM movie, capture rate: 1 frame per second. Elapsed time is given in min:sec. Electron energy ε = 2.6 eV.

Movie S4: Onset of spiral growth of few-layer SnS on monolayer graphene/Ru(0001) at 280°C. Dark lines are edges of individual SnS atomic sheets. Several growth spirals develop during the period captured by the movie. Real-time LEEM movie, capture rate: 1 frame per second. Elapsed time is given in seconds. Electron energy ε = 2.6 eV. Scale bar: 0.5 µm.

Movie S5: Spiral decomposition during sublimation of few-layer SnS on monolayer graphene/Ru(0001) at 330°C. Dark lines are edges of individual SnS atomic sheets, which retract in inverse edge-flow during sublimation from the surface. Growth spirals are eliminated due to materials removal by sublimation during the period captured by the movie. Real-time LEEM movie, capture rate: 1 frame per second. Elapsed time is given in seconds. Electron energy ε = 2.6 eV. Scale bar: 0.5 µm.