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## Supplementary infromation:

## Micrometer-scale monolayer SnS growth by physical vapor deposition

H. Kawamoto<sup>a</sup>, N. Higashitarumizu<sup>a</sup>, N. Nagamura<sup>b,c</sup>, M. Nakamura<sup>b</sup>, K. Shimamura<sup>b</sup>, N. Ohashi<sup>b</sup>, and K. Nagashio<sup>\*a</sup>

<sup>a</sup>Department of Materials Engineering, The University of Tokyo, Tokyo 113-8656, Japan <sup>b</sup>National Institute for Materials Science, Ibaraki 305-0044, Japan <sup>c</sup>PRESTO, Japan Science and Technology Agency, Saitama, 332-0012, Japan <sup>\*</sup>nagashio@material.t.u-tokyo.ac.jp



**Fig. S1.** (a) Schematic illustration of three zone furnace for PVD growth. (b) Typical temperature setup for monolayer SnS growth. The right, center, and left heater temperatures are 410°C, 440°C, and 470°C, respectively. The distance from the SnS feed powder to the mica substrate is approximately 30 cm. The pressure during the growth was kept at 10 Pa.



**Fig. S2.** Equilibrium vapor pressure as a function of temperature for different reactions calculated from the standard Gibbs free energy change ( $\Delta G^{\circ} = RT \ln K$ ). R is the gas constant, and K is the equilibrium constant and can be approximately expressed as the partial pressure of the gas phase ( $P_{gas}$ ). Although the product of two partial pressures is only available in reactions (iii), (iv), and (vi), the vapor pressure for each gas phase can be considered to be higher than the product pressures. Therefore, these data clearly indicate that the vapor pressures of SnO<sub>2</sub>(g) and SnO(g) at the temperature for the SnS feed powder are much smaller than those of SnS(g) and S(g). This is consistent with the fact that SnO<sub>2</sub> remained in the purchased powder even after the growth run.



**Fig. S3.** (a) AFM images for SnS films with small grains. (b) Optical images of SnS film on a mica substrate before and after the reheating experiment. First, an AFM image of the SnS film was taken. A graphite flake was placed at the predetermined position near the desired SnS film as a position mark using a transfer system (following refs). Then, the sample was reheated in  $N_2$  atmosphere at 400°C and a furnace pressure of 10 Pa for 20 min. Finally, an AFM image was taken at the same position as before.

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T. Uwanno, Y. Hattori, T. Taniguchi, K. Watanabe, and K. Nagashio, *2D mater.*, 2015, 2, 041002.



Fig. S4. EDS spectra for a submicron grain as highlighted in Fig. 4c.



**Fig. S5.** 3D-nano ESCA installed at The University of Tokyo outstation beamline BL07LSU in SPring-8 was used for the chemical analysis. (a) Sample holder for 3D-nano ESCA, where the Ni/Au electrodes are grounded to the sample holder using Cu wire and Ag paste. (b) Low magnification optical image of the device. (c) Magnified optical image of the SnS film with Ni/Au electrodes.