Journal Name

RSCPublishing

ARTICLE

Supporting Information

Physical Vapor Deposition Synthesis of Two-dimensional Orthorhombic SnS Flakes with Strong Angle/Temperature-Dependent Raman Responses

Jing Xia,^a Xuan-Ze Li,^a Xing Huang,^a Nannan Mao,^b Dan-Dan Zhu,^a Lei Wang,^a Hua Xu^c and Xiang-Min Meng^{a*}

^aKey Laboratory of Photochemical Conversion and Optoelectronic Materials, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing, 100190, P. R. China.

^bCenter for Nanochemistry, Beijing National Laboratory for Molecular Sciences, College of Chemistry and Molecular Engineering, Peking University, Beijing, 100871, P. R. China

^cSchool of Materials Science and Engineering, Shaanxi Normal University, Xi'an, 710062, P.R. China



Fig. S1 (a, d) Optical images of the SnS flakes grown at 700 °C with pressure of 80 Torr and 800 °C with pressure of 300 Torr, respectively. (b, e) Statistics of the lateral size of the SnS flakes obtained from (a, d), respectively. (c, f) AFM images and the corresponding height information of the SnS flakes obtained from (a, d), respectively.



Fig. S2 (a) Optical image of a large SnS flake at 600 °C and 80 Torr, the directions of incident and scattered light polarization, the corner angle between (-101) and (-10-1) planes, and the sample rotation angle (θ) between the incident light polarization and the Y axis. (b, c) Raman spectra of the 2D SnS flake with different rotation angles under parallel or cross-polarization configurations, respectively. (d-g) Polar plots of the measured and fitted peak intensities of the A_g (190.7 cm⁻¹) and B_{3g} (162.9 cm⁻¹) modes as a function of the sample rotation angle under parallel or cross-polarization configurations, respectively.



Fig. S3 (a) Optical image of a large SnS flake at 700 °C and 80 Torr, the directions of incident and scattered light polarization, and the sample rotation angle (θ) between the incident light polarization and the Y axis. (b-e) Polar plots of the measured and fitted peak intensities of the A_g (190.7 cm⁻¹) and B_{3g} (162.9 cm⁻¹) modes as a function of the sample rotation angle under parallel or cross-polarization configurations, respectively.



Fig. S4 (a) Optical image of a large octagonal SnS flake at 800 °C and 300 Torr, the exposed planes, the intersection angles, the directions of incident and scattered light polarization, and the rotation angle (in this case, $\theta=0^{\circ}$) between the incident light polarization and the Y axis. (b-e) Polar plots of the measured and fitted peak intensities of the A_g (190.7 cm⁻¹) and B_{3g} (162.9 cm⁻¹) modes as a function of the sample rotation angle.

During the optical characterization, a few octagonal 2D SnS flakes were observed occasionally. Owning to the orthorhombic structure of the as-grown 2D SnS flakes, the exposed planes can be easily assigned to {101} and {200} planes based the angular relation between them, allow determining the armchair and zigzag directions, as shown in Figure S4a. The armchair and zigzag directions of this octagonal SnS flake were denoted along the Y axis and Z axis, respectively, and then we started to rotate this flake every 15° in clockwise direction. The final results are displayed in Fig. S4b-e. It can be clearly observed that the angles of the maximum peak intensities of the A_g (190.7 cm⁻¹) mode are always along the armchair direction of the octagonal SnS flake under parallel-polarization configuration, which further demonstrates that the A_g (190.7 cm⁻¹) mode can be used as a reliable fingerprint for identifying the crystalline direction of the 2D SnS flakes. It must be pointed out that the angles of the minimum peak intensities of the B_{3g} (162.9 cm⁻¹) mode are always parallel or perpendicular to the armchair direction of the 2D SnS flakes under parallel-polarization configuration, and the angles of the maximum peak intensities of the B_{3g} (162.9 cm⁻¹) mode are also parallel or perpendicular to the armchair direction of the 2D SnS flakes under perpendicular-polarization configuration. As a result, B_{3g} can help determining the zigzag or armchair direction.



Fig. S5 Polar plots of the calculated peak intensities of the A_g and B_{3g} modes as a function of the sample rotation angle under parallel-polarization configuration. For A_g , different values of B/C are used to optimize the fitted curves.