Supplementary Information

Revealing Anisotropy and Thickness Dependence of Raman Spectra for SnS Flakes

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Fig. S1 AFM image of 13.2 nm SnS flake on mica substrate before transfer.
**Fig. S2** Polar plots of Raman intensities of $B_{3g}$ (164.0 cm$^{-1}$) of different thick SnS flakes with 632.8 nm laser excitation under parallel and perpendicular polarization configurations, respectively. The red curves are fitting results according to eqn (2) and eqn (4) in the main text.

**Fig. S3** The calculated polar plots of $A_g$ (//$)$ mode with different parameters of $|C|/|B|$ and $\phi_{CB}$ according to eqn (1) in the main text.
As shown in Fig. S2, $|C|^2/|B|^2$ is equal to the intensity ratio of the zigzag direction to the armchair direction. When $|C|<|B|$, the maximum values of the polar plots are along armchair direction (a,b). However, the maximum values of the polar plots become along zigzag direction if $|C|>|B|$ (c,d). $\phi_{CB}$ determines the concave degree of the intensity pattern along nearby 45° (or 225°) and 135° (or 315°) (b, d).

**Fig. S4** Polar plots of Raman intensities of $A_g$ (95.9 cm$^{-1}$, //) and $A_g$ (192.0 cm$^{-1}$, //) for different thick SnS flakes with 514.5 nm and 632.8 nm laser excitation, respectively. The red curves are fitting results.

As shown in Fig. S3, the maximum intensity values of $A_g$ (95.9 cm$^{-1}$, //) mode for SnS flakes with different thicknesses are always along armchair direction and their polar plots are more anisotropic than those corresponding to $A_g$ (192.0 cm$^{-1}$, //). So $A_g$ (95.9 cm$^{-1}$) is a more reliable way than $A_g$ (192.0 cm$^{-1}$) to identify the crystalline orientation of SnS.
Fig. S5 Variation of $\phi_{CB}$ with the thickness of SnS flake for $A_g$ (192.0 cm$^{-1}$) mode with 514.5 nm and 632.8 nm laser excitation, respectively.