Supporting Information

Revealing the Anisotropic Phonon Behaviors of Layered SnS by Angle/Temperature-Dependent Raman Spectroscopy

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1. Supporting Tables

Table S1 Selected bond lengths and angles for SnS.

Bond lengths (Å)		Bond angles (°)	
Sn-S	2.625(4)	S-Sn-S[1]	89.10(9)
Sn-S[1]	2.664(3)	S-Sn-S[2]	89.10(9)
Sn-S[2]	2.664(3)	S-Sn-S[2]	96.76(14)
S-Sn[3]	2.664(3)	Sn-S-Sn[3]	103.14(12)
S-Sn[4]	2.664(3)	Sn-S-Sn[4]	103.14(12)
		Sn[3]-S-Sn[4]	96.76(14)

[1] 1/2–X,2–Y,1/2+Z; [2] 1/2–X,1–Y,1/2+Z; [3] 1/2–X,1–Y,–1/2+Z; [4] 1/2–X,2–Y,–1/2+Z

Table S2 Fractional atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) for SnS.

Atom	*x*	*y*	*z*	*U(eq.)*[#]
Sn	3803.6(10)	7500	6197(3)	18.5(5)
S	1489(4)	7500	5213(10)	16.4(9)

[#] *U(eq.)* is defined as 1/3 of the trace of the orthogonalized U_{ij} tensor.

2. Supporting Figures

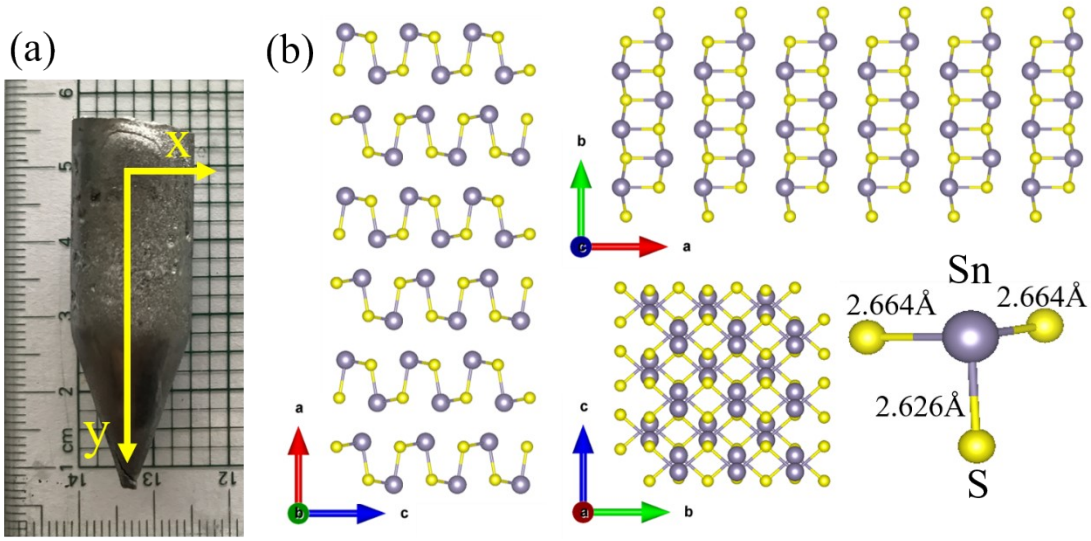

Figure S1. (a) Optical image of the SnS single crystal synthesized by the home-made Bridgman method, the single crystal ingots with a diameter of 14 mm and a height of 45 mm. (b) Schematic of SnS crystal structure along each axis, and the short Sn-S bond length.

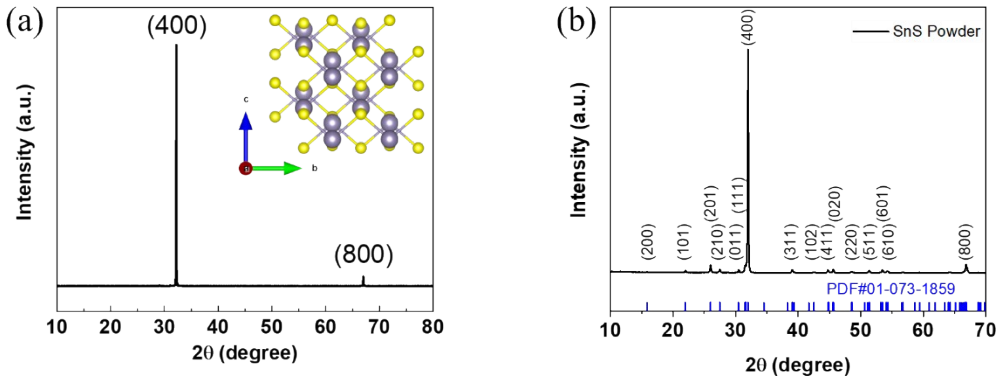

Figure S2. XRD diffraction pattern of SnS (a) single crystal and crystal structure diagram, (b) powder and corresponding PDF card # 01-073-1859.

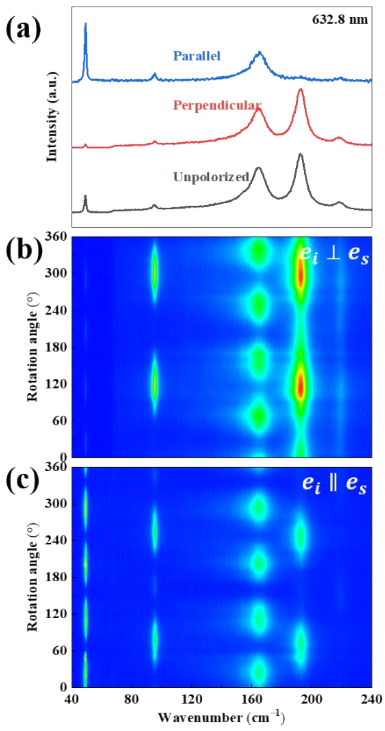

Figure S3. ARPRS of SnS single crystal excited by 632.8 nm. (a) Measuring Raman spectra at a 0° angle under unpolarized, parallel and perpendicular configurations. False-color plots of polarized Raman intensities under (b) perpendicular and (c) parallel polarization configurations, respectively.

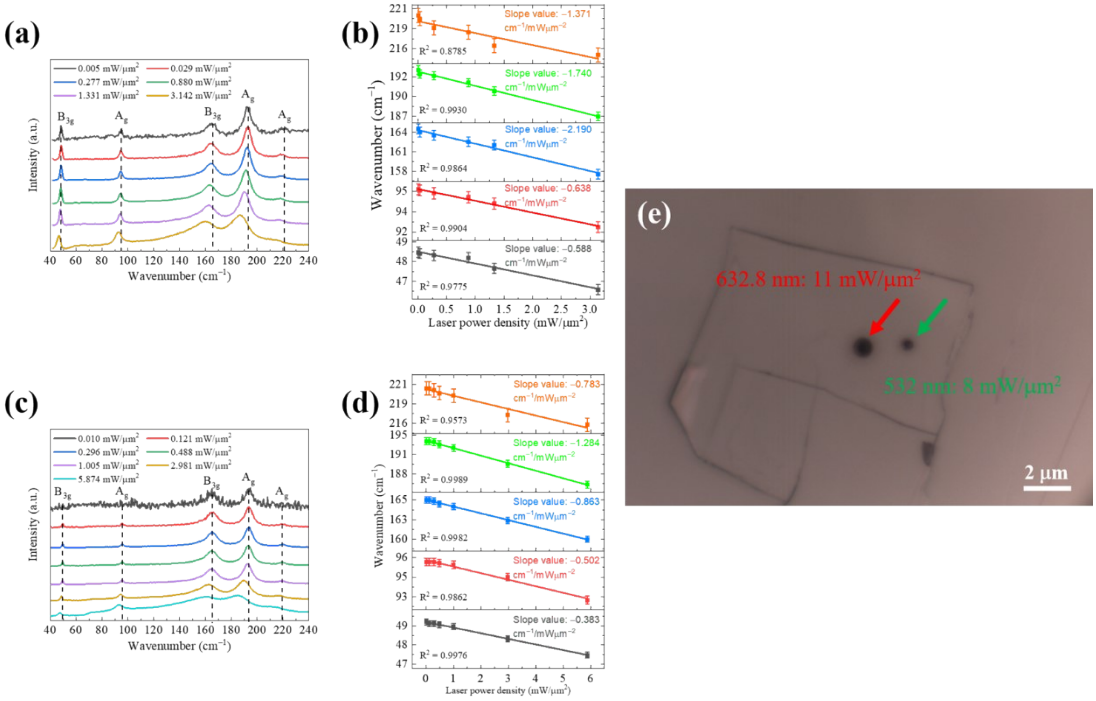

Figure S4. *In-situ* laser power-dependent Raman spectra of SnS single crystal. (a, c) Raman scattering spectrum of SnS at different laser powers excited by 532 nm and 632.8 nm lasers, respectively. (b, d) Laser power dependence of Raman peak positions fitted by linear equation for 532 nm and 632.8 nm, respectively. (e) Optical photograph of sample damaged by laser over threshold for 532 nm and 632.8 nm laser.

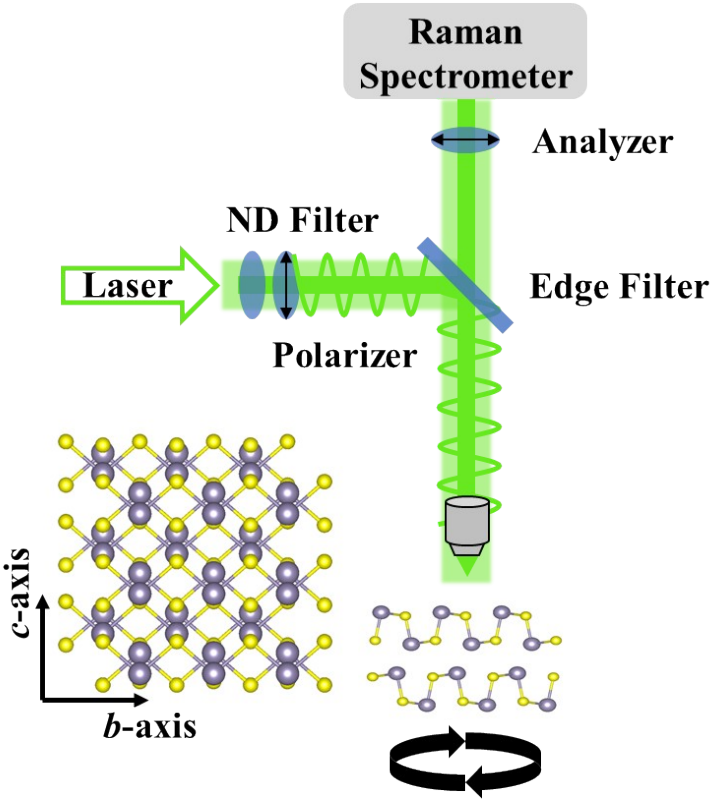

Figure S5. Schematic of polarized Raman system. The SnS single crystal is irradiated by the 532 nm and 632.8 nm lasers. Laser power and polarization of the incident beam are controlled by the neutral attenuation filter and polarizer, respectively.