

Lattice dynamics of Sb_2Te_3 at high pressures

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Supplementary materials

Supplementary Table I. Theoretical (th.) *ab initio* IR-mode frequencies and pressure coefficients observed in α -Sb₂Te₃ (R-3m phase) at room temperature and $P_0=1$ atm, as obtained from fits to the data using $\omega(P) = \omega(P_0) + a_1 \cdot (P - P_0)$. Experimental IR-mode frequencies at room pressure are given for comparison after Ref. 41.

Mode	ω_0 (cm ⁻¹)	a_1 (cm ⁻¹ /GPa)	ω_0 (th.) (cm ⁻¹)	a_1 (th.) (cm ⁻¹ /GPa)
E _u ¹	67	-	78.0	2.95
E _u ²	-	-	100.4	1.21
A _{2u} ¹	-	-	109.9	1.91
A _{2u} ²	-	-	138.7	3.54

Supplementary Table II. Theoretical (th.) *ab initio* IR-mode frequencies and pressure coefficients observed in β -Sb₂Te₃ (C2/m phase) at room temperature at P₀= 8.6 GPa as obtained from fits using $\omega(P) = \omega(P_0) + a_1 \cdot (P - P_0)$.

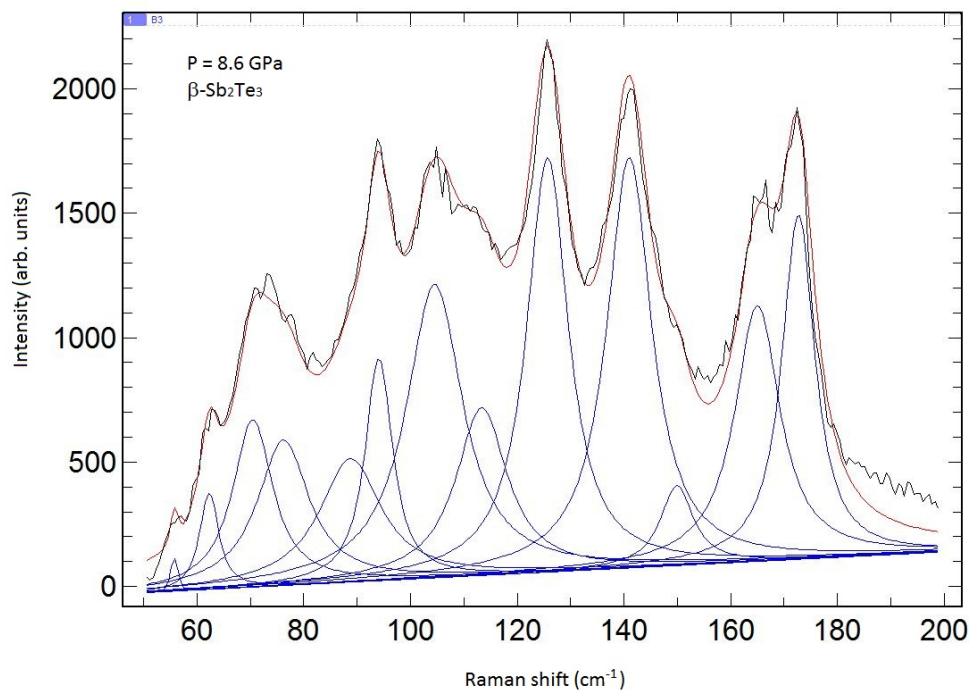
Mode	$\omega (P_0)$ (th.) (cm ⁻¹)	a_1 (th.) (cm ⁻¹ /GPa)
A _u ¹	66.9	1.92
B _u ¹	66.9	0.95
B _u ²	79.2	1.58
A _u ²	93.8	2.16
B _u ³	96.7	2.23
A _u ³	98.9	2.04
B _u ⁴	109.3	2.81
A _u ⁴	128.5	1.93
B _u ⁵	132.3	1.48
B _u ⁶	138.5	2.64
B _u ⁷	155.6	1.52
B _u ⁸	163.3	2.36

Supplementary Table III. Theoretical (th.) *ab initio* IR-mode frequencies and pressure coefficients observed in γ -Sb₂Te₃ (C2/c phase) at room temperature at P₀= 15.2 GPa as obtained from fits using $\omega(P) = \omega(P_0) + a_1 \cdot (P - P_0)$.

Mode	$\omega(P_0)$ (th.) (cm ⁻¹)	a_1 (th.) (cm ⁻¹ /GPa)
B _u ¹	53.2	0.19
A _u ¹	53.9	1.18
A _u ²	78.5	0.20
B _u ²	79.4	0.55
A _u ³	99.8	1.19
B _u ³	121.1	1.43
B _u ⁴	121.7	2.19
A _u ⁴	136.7	0.98
B _u ⁵	140.7	2.15
A _u ⁵	159.2	1.80
A _u ⁶	162.2	2.34
B _u ⁶	168.6	2.20

Supplementary Figure 1.

Raman spectrum of β -Sb₂Te₃ at 8.6 GPa decomposed into Voigt profiles. The five high-frequency peaks above 120 cm⁻¹ can be clearly attributed to Raman-active modes of the C2/m structure (see correspondence with bottom marks in Fig. 4(b) in the main text). This gives us confidence in our assignment of this phase to the C2/m structure recently found in Bi₂Te₃. The assignment of the low-frequency modes below 120 cm⁻¹ to Raman-active modes of the C2/m structure is more difficult because there are 8 Raman-active modes between 50 and 120 cm⁻¹. The two lowest-frequency Voigt lines fitted around 56 and 62 cm⁻¹ have not been attributed to Raman-active modes because they are in the region where the edge filter cuts the Raman spectrum and its pressure evolution does not seem to match to the calculated pressure dependence of the modes expected in that region. Additionally, the mode A_g² does not appear at 8.6 GPa and is detected above 10 GPa as can be seen in Figs. 4(a) and 4(b) in the main text. The rather broad band fitted at 88 cm⁻¹ cannot be assigned to β -Sb₂Te₃ and could be a second-order Raman peak. Finally, the small shoulder at about 118.4 cm⁻¹ (not fitted due to its low intensity) can be attributed to the pair A_g⁵, B_g⁵ after studying its evolution with pressure.



Supplementary Figure 2.

Raman spectrum of γ -Sb₂Te₃ at 18.6 GPa decomposed into Voigt profiles. We have attributed Raman-active modes of the C2/c structure to all Voigt peaks above 120 cm⁻¹ and below 80 cm⁻¹ (see correspondence with Fig. 5(b) in the main text). However, we have not assigned the broad band around 91 cm⁻¹ to a first-order mode because the A_g³ Raman-active mode is theoretically predicted to be around 106 cm⁻¹ at 18.6 GPa and additionally the mode at 91 cm⁻¹ is a broad band whose frequency is difficult to follow at higher pressures. Note that at least 9 bands can be clearly observed without fitting the Raman spectrum; however, the full Raman spectrum cannot be fitted with only 9 Voigt profiles.

