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Supporting Information for

Anomalous Raman Modes in Tellurides

F. J. Manjón,[a,*] S. Gallego-Parra,[a] P. Rodríguez-Hernández,[b] A. Muñoz,[b]

C. Drasar,[c] V. Muñoz-Sanjosé,[d] and O. Oeckler[e]

[a] Instituto de Diseño para la Fabricación y Producción Automatizada, MALTA Consolider Team, Universitat Politècnica de València, 46022 Valencia, Spain

[b] Departamento de Física, Instituto de Materiales y Nanotecnología, MALTA Consolider Team, Universidad de La Laguna, 38205 Tenerife, Spain

[c] Faculty of Chemical Technology, University of Pardubice, Pardubice 532 10, Czech Republic

[d] Departamento de Física Aplicada y Electromagnetismo, Universitat de València, 46100 Burjassot, Spain

[e] Institut für Mineralogie, Kristallographie und Materialwissenschaft, Universitat Leipzig, Germany

* corresponding author: fjmanjon@fis.upv.es

Figure S1. Polarized and unpolarized Raman spectra of GaGeTe showing the anomalous Raman modes. Spectra have been normalized and vertically shifted for the sake of clarity. Note that horizontally and vertically polarized measurements refer to the use of analyzer in horizontal and vertical position, while unpolarized measurements refer to the fact that no analyzer has been used.

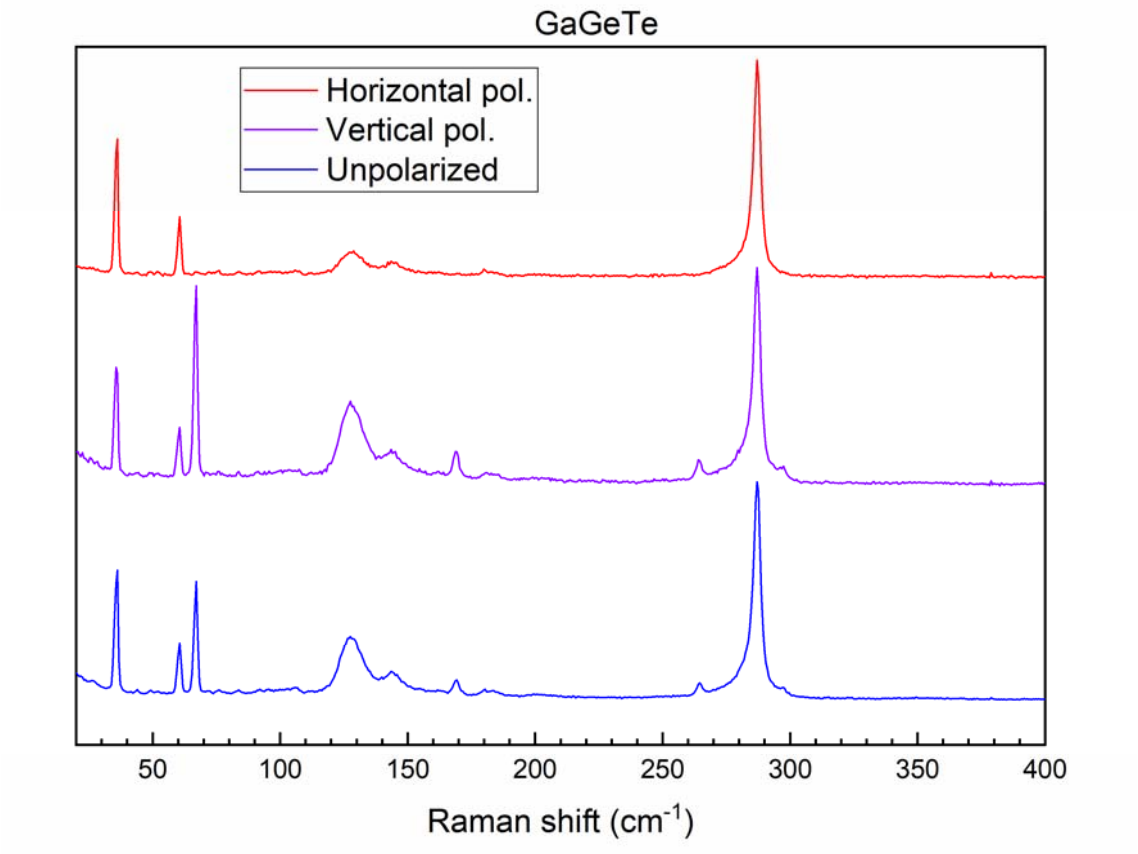
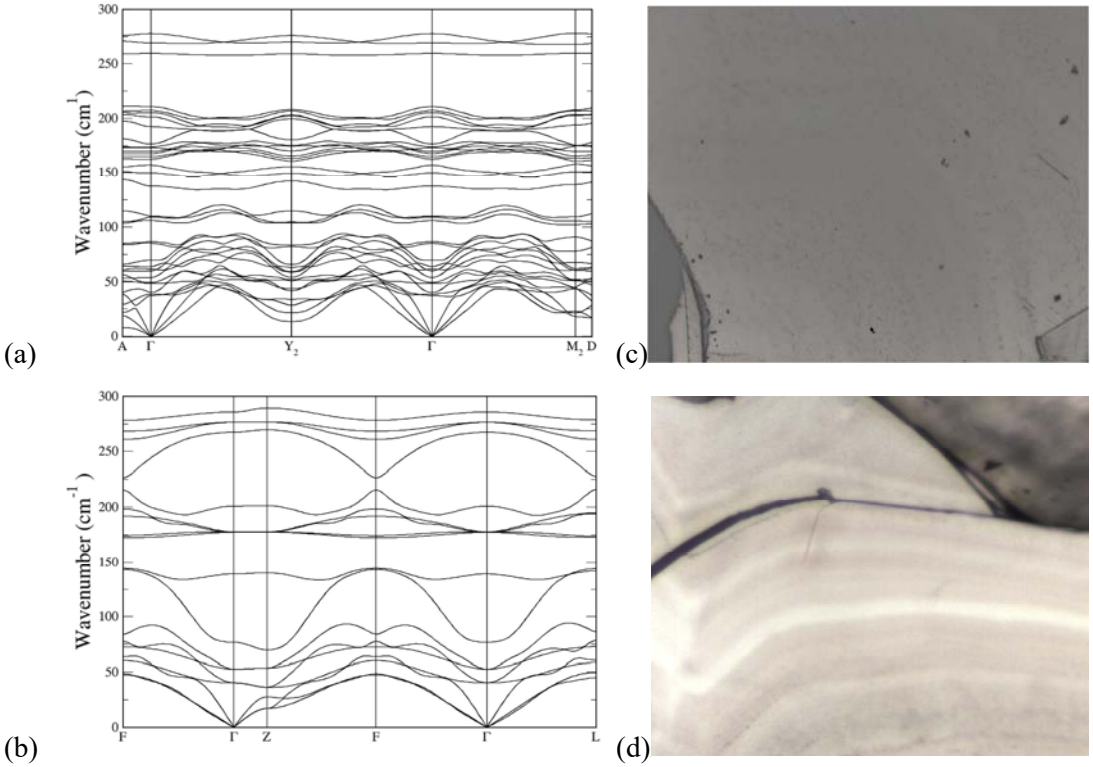
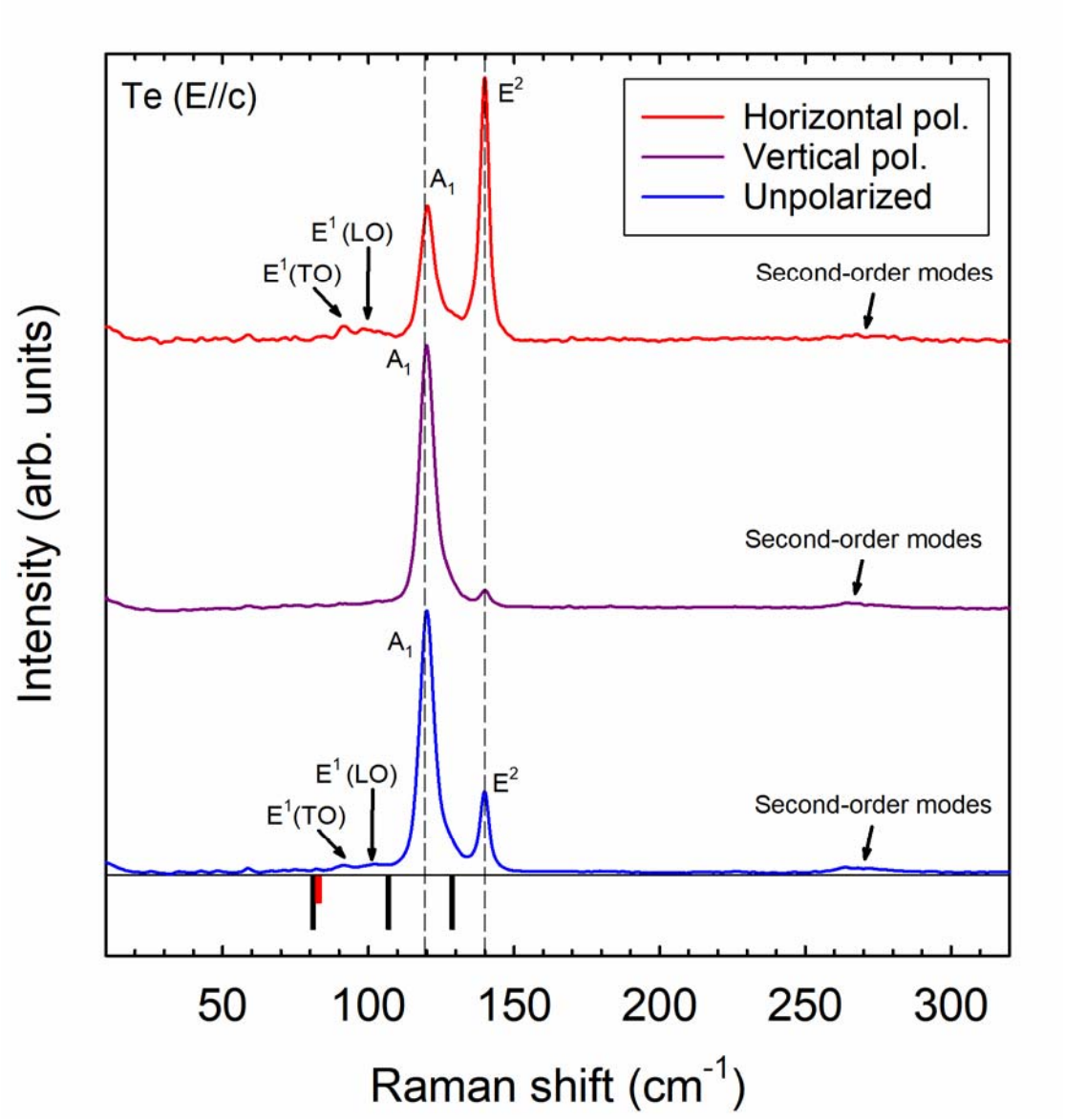


Figure S2. Phonon dispersion curves of monoclinic GaTe (a) and rhombohedral GaGeTe (b) and optical microscopic images of both samples (c) and (d), respectively. The c-a axis of the monoclinic unit cell of GaTe is perpendicular to the image, while the c axis of the hexagonal unit cell of GaGeTe is perpendicular to the image.

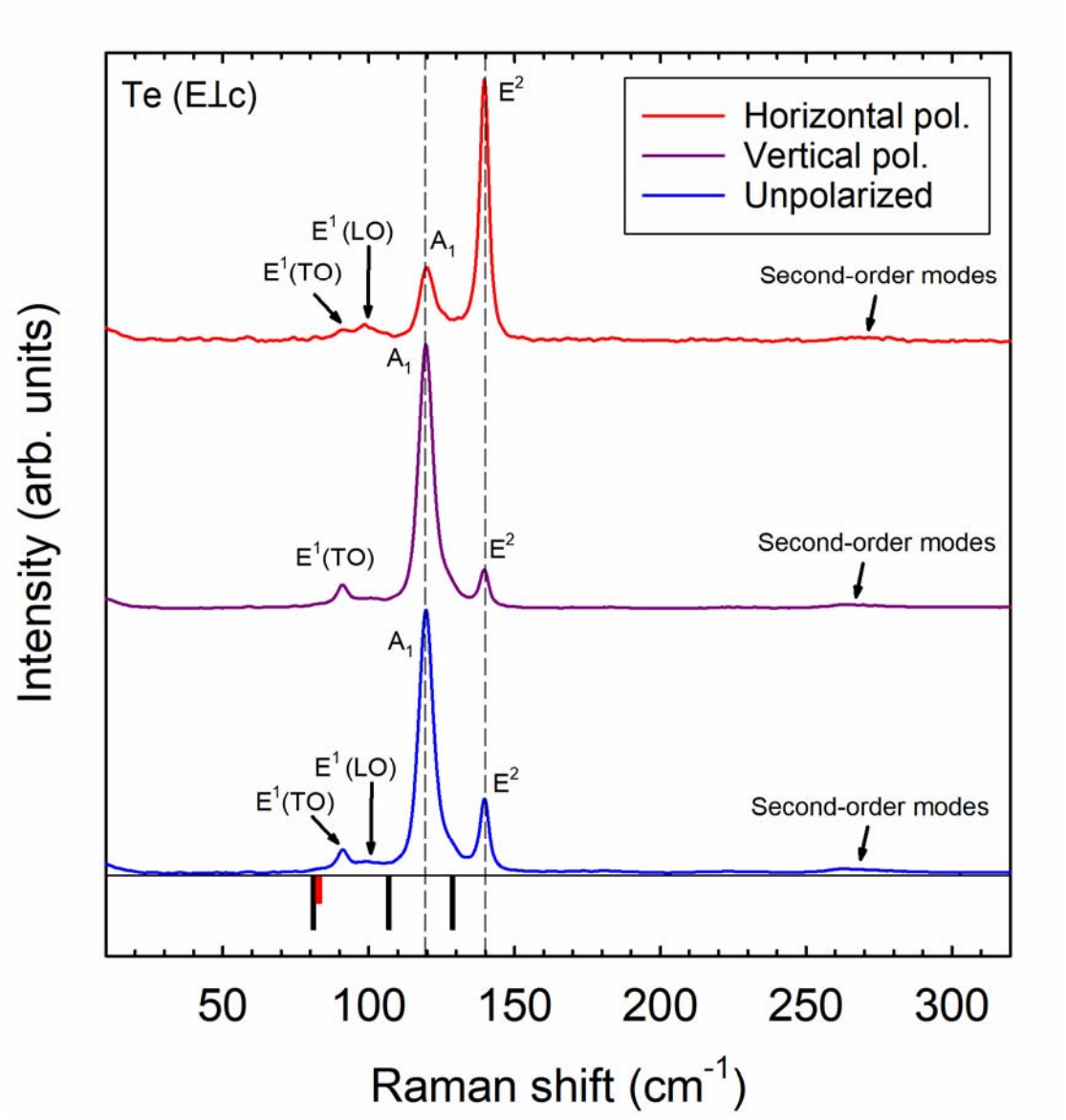


In relation to Fig. S2, GaTe shows rather flat dispersion curves in the frequency range 100-150 cm^{-1}, while GaGeTe shows flat dispersion curves near 140 cm^{-1}. These flat dispersion curves come from the layered nature of GaTe and GaGeTe so one cannot consider that this is a sufficient reason for the observation of ARMs coming by folding of the Brillouin zone in tellurides or due to defects or disorder. Note that flat phonon bands between 100 and 150 cm^{-1}, are not observed in the phonon dispersion curves of many tellurides, like bulk hexagonal CdTe (see Materials Database 10.17188/1189186) and ARMs are also found in CdTe as explained in the main text.

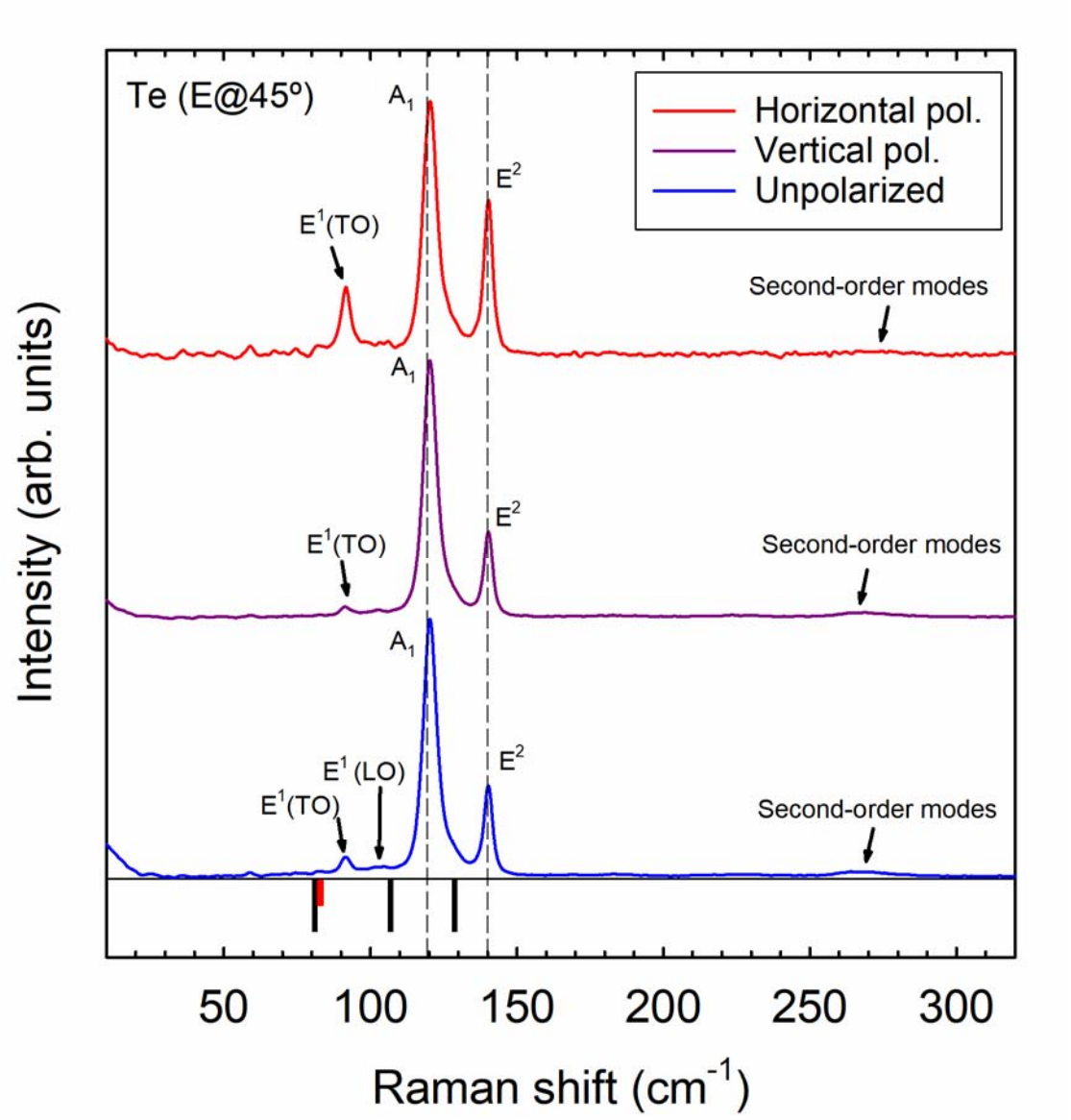
Figure S3. Raman spectra of trigonal Te in backscattering geometry. (a) With laser polarization parallel to the rods; i.e. the c axis (E ∥ c). (b) With laser polarization perpendicular to the rods (E ⊥ c). (c) With laser polarization at 45° of the rods (E@45°). Unpolarized as well as parallel (vertical pol.) and cross (horizontal pol.) polarized measurements were performed. Bottom black (red) marks show the calculated Raman-active (IR-active) TO modes of Te. Note that the two E modes of Te are also IR-active. Spectra have been normalized and vertically shifted for the sake of clarity. Note that horizontally and vertically polarized measurements refer to the use of analyzer in horizontal and vertical position, while unpolarized measurements refer to the fact that no analyzer has been used.



(a)


Te (E⊥c)
Horizontal pol.
Vertical pol.
Unpolarized
E²
E¹ (LO)
E¹(TO)
A₁
Second-order modes
Intensity (arb. units)
50
100
150
200
250
300
Raman shift (cm⁻¹)

(b)


Te (E@45º)
A1
E2
E1(TO)
Horizontal pol.
Vertical pol.
Unpolarized
Second-order modes
A1
E2
E1(TO)
Second-order modes
A1
E1 (LO)
E2
E1(TO)
Second-order modes
Intensity (arb. units)
50
100
150
200
250
300
Raman shift (cm-1)

(c)

Figure S4. Unpolarized Raman spectra of GaGeTe for different laser powers. Spectra have been normalized and vertically shifted for the sake of clarity. The notation 1mW[a] indicates that this spectrum has been excited with 1 mW power at the same spot previously excited with 20 mW once the sample has thermalized.

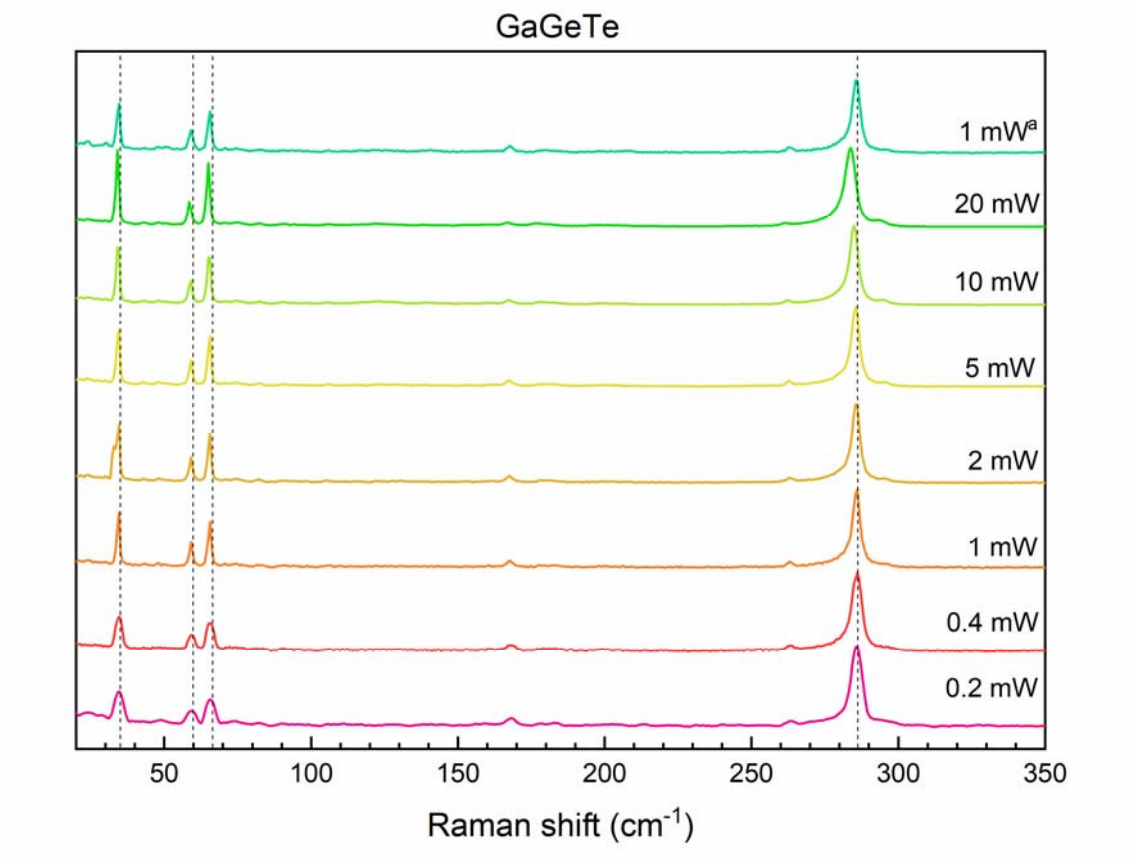


Figure S5. Unpolarized Raman spectra of Bi_2Se_3 (a) and Sb_2Se_3 (b) with different laser powers. Spectra have been normalized and vertically shifted for the sake of clarity. Dashed lines show some of the normal Raman peaks of the compounds. Black (blue) asterisks correspond to Sb_2O_3 (Se) modes. The notation 1mW[a] in (a) indicates that the spectrum has been excited with 1 mW power at one spot previously excited with 20 mW (different to that plotted in the figure) once the sample has thermalized. The notation 1mW[a] in (b) indicates that the spectrum has been excited with 1 mW power at the same spot previously excited with 20 mW (the one shown in the figure) once the sample has thermalized. The notation 1mW[b] in (b) indicates that the spectrum has been excited with 1 mW power in a region close to that of the spot previously excited with 20 mW (the one shown in the figure) once the sample has thermalized.