Charge Density Waves phase transition in bulk 1T-$TaSe_2$ using angle-resolved temperature dependence Raman spectroscopy

Mariangela Ruggeri
D. Wolverson, V. Romano, G. Cerullo, C. J. Sayers, G. D'Angelo
Università degli Studi di Messina, Viale F. Stagno d'Alcontres 31, S. Agata, 98166 Messina, Italy
Mariangela.ruggeri@studenti.unime.it

Charge density wave (CDW) refers to the periodic modulation of electronic distributions accompanied by lattice distortion. Controlling collective electron states is crucial in materials research for developing novel devices. Transition metal dichalcogenides (TMDs) exhibit a rich collection of CDW. 1T-$TaSe_2$ is a prototypical CDW material for which electron-phonon coupling and associated lattice reconstruction play an important role in driving and stabilizing the CDW phase. Here, we investigate the lattice dynamics of bulk 1T-$TaSe_2$ using angle-resolved ultralow wavenumber Raman spectroscopy down to 10 cm^{-1}. Our high-resolution Raman spectra allow us to identify at least 27 peaks in the commensurate (CCDW) phase in the region 50 – 300 cm^{-1} [**Fig 1a**]. Contrary to other layered materials, we do not find evidence of interlayer breathing or shear modes, suggestive of AA stacking in the bulk. Polarization dependence of the mode intensities combined with DFT calculations allow the assignment of their symmetry [**Fig 1b**]. A detailed temperature dependence in the range T= 80 – 500 K allows us to clearly identify the soft modes associated with the CDW superlattice. Above the CCDW at 473 K, we observe a dramatic loss of resolution of all modes, and significant linewidth broadening associated with a reduced phonon lifetime as the charge-order becomes incommensurate with the lattice [**Fig 1c**]. This work is published in Physical Review B.

References

Figures

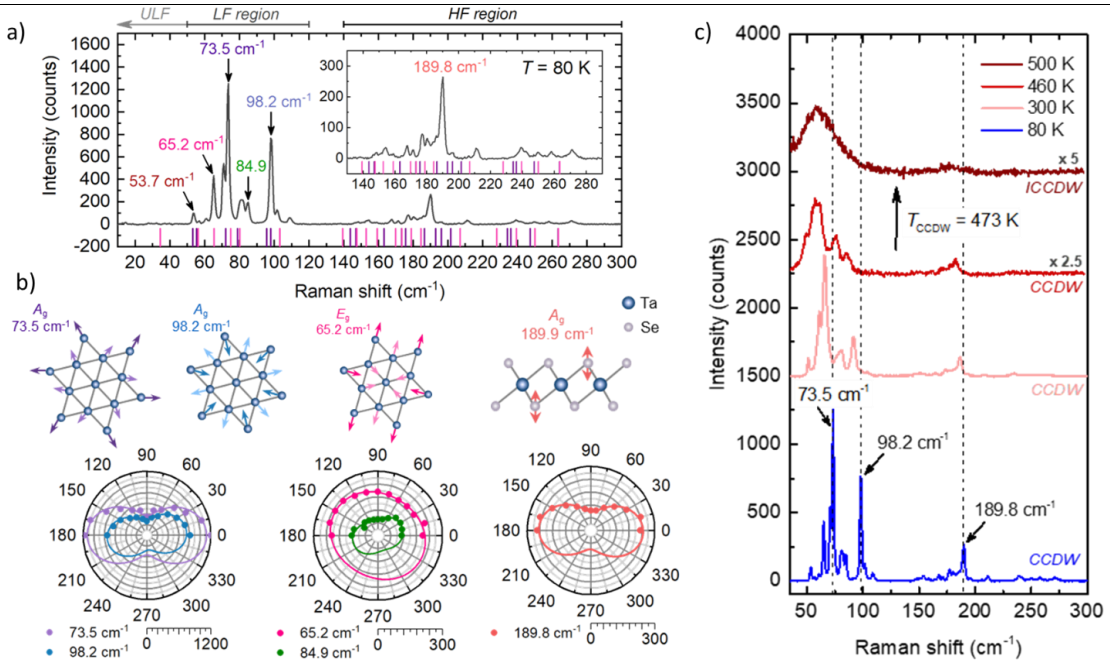

Figure 1 a) High resolution Raman spectrum at 80 K (θ = 0°) where 27 well-resolved peaks can be identified. Modes of interest are labelled. Pink and purple lines at the bottom represent the calculated theoretical frequencies of vibrations. b) Identification of selected LF and HF modes with A_g and E_g symmetry based on polar plots of the mode intensities as a function of polarisation angle. Sketches of the corresponding atomic displacements of Ta atoms in the star cluster and Se atoms obtained by DFT are shown. c) Raman spectra at selected temperatures. Data is offset for clarity. The vertical dashed lines indicate the position of the 73.5 cm^{-1}, 98.2 cm^{-1}, and 189.8 cm^{-1} peaks corresponding to their frequency at 80 K.