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Hot photoluminescence and Raman processes in symmetric and asymmetric monolayer semiconductors

Energy relaxation of photo-excited charge carriers is of significant fundamental interest and crucial for the performance of layered semiconductors in optoelectronics. I will show how ultra-low frequency Raman spectroscopy can be used for the investigation of the primary stages of carrier relaxation in a layered semiconductor. A series of periodic maxima in the hot photoluminescence intensity with a period of ~15 meV, stemming from energy states higher than the exciton state is revealed for both below (Stokes) and above (anti-Stokes) the laser excitation energy (Figure1a,b).[1] These are assigned to phonon cascades, whereby carriers undergo phonon-induced transitions between real states above the free-carrier gap with a probability of radiative recombination at each step and provide information on the relaxation pathway in the Brillouin zone.
Phonon cascades are also observed in novel, high-quality Janus SeMoS monolayers with a broken out-of-plane mirror symmetry and an in-built electric dipole. I will also discuss how the structure of these asymmetric monolayers can be assessed by Raman spectroscopy, showing a striking difference in the vibrational properties between Janus SeMoS and random MoSSe alloys (Figure1c). [2,3]

References

[1] I. Paradisanos, G. Wang et al., Nat Commun, 12 (2021) 538
[2] M. M. Petric, M Kremser M. Barbone et al., Phys Rev B, 103 (2021) 035414
[3] Z. Gan, I. Paradisanos et al., arXiv:2205.04751 (2022)

Figures

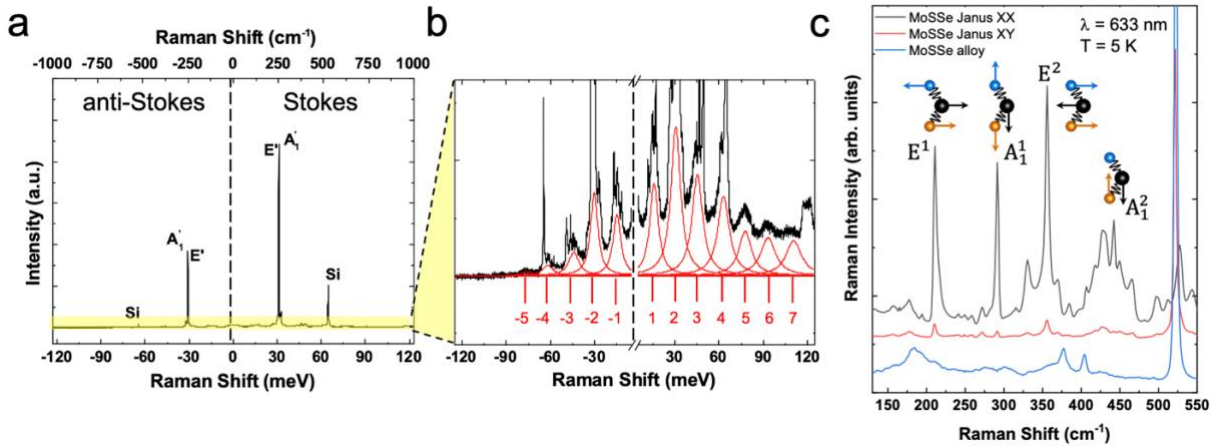

Figure 1: a) Emission and scattering spectrum of 1L-WSe_2 as a function of energy shift with respect to the excitation laser (here set to zero). **b)** Magnified portion of the spectrum in yellow in **a**. This reveals 7 periodic Stokes peaks and 5 anti-Stokes, assigned to phonon cascades. c) Comparison of Raman scattering data between SeMoS Janus monolayer (black and red graphs correspond to co- and cross- polarized detection) and MoSSe alloy monolayer (blue graph) which shows MoS_2-like doublet around 400 cm^{-1}. The main identified modes for the Janus sample are marked.