Efficient phonon cascades in an atomically thin semiconductor

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Energy relaxation of photo-excited charge carriers is of significant fundamental interest and crucial for the performance of layered semiconductors in optoelectronics. The primary stages of carrier relaxation affect a plethora of subsequent physical mechanisms. Here we measure light scattering and emission in tungsten diselenide (WSe_2) monolayers close to the laser excitation energy (down to ~0.6 meV). We reveal a series of periodic maxima in the hot photoluminescence intensity, stemming from energy states higher than the A-exciton state. We find a period ~15 meV for 7 peaks below (Stokes) and 5 peaks above (anti-Stokes) the laser excitation energy, with a strong temperature dependence. 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. We infer that intermediate states in the conduction band at the Λ-valley of the Brillouin zone participate in the cascade process of WSe_2 monolayers. This provides a fundamental understanding of the first stages of carrier–phonon interaction, useful for optoelectronic applications of layered semiconductors. [1]

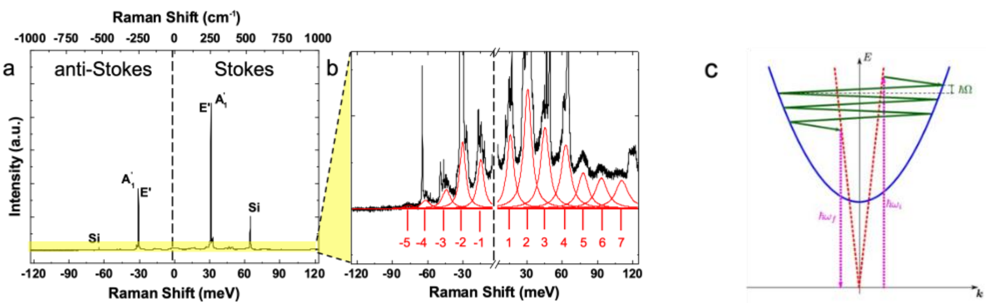

Figure 1: **(a)** Scattering spectrum of 1L-WSe_2 at 295K. The degenerate in-plane (E′) and out-of-plane (A'_1) Raman mode ~250 cm^{-1}, as well as the Si Raman peak ~521 cm^{-1}, are prominent in both Stokes (S) and anti-Stokes (AS). **(b)** Magnified portion of the spectrum in yellow in **a**. This reveals 7 periodic S peaks and 5 AS. **(c)** Scheme of phonon-assisted hot PL. Incident and outgoing photons are shown by dotted magenta vertical arrows. The phonons participating in the cascade are indicated by the green arrows. The exciton dispersion curve is the blue parabola.

References

[1] I. Paradisanos, G. Wang., E. M. Alexeev *et al.*, Nat Commun **12**, 538 (2021)

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