Strain induced frequency shifts of the second order Raman modes of monolayer WS_2

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Atomically thin transition metal dichalcogenides (2D - TMDCs) are an emerging class of materials with great prospect in fundamental as well as applied science. Importantly, their optical and vibrational properties can be significantly tuned by external stimuli, such as mechanical strain [1]. Despite the fact that the strain dependence of the first order Raman modes have been studied extensively, the impact of strain on the higher order Raman scattering remains rather unexplored. Important insight on the lattice dynamics can be obtained from higher order Raman modes, since phonons with non-zero momenta can be probed [2].

In this work, WS_2 monolayer single crystals, fabricated by an atmospheric pressure chemical vapor deposition method, are subjected to controlled biaxial deformations. *In-situ* Raman microscopy is employed to study the evolution of the second order Raman modes under various levels of mechanical strain. Under resonant excitation (2.41 eV) numerous higher order peaks emerge in the Raman spectrum of WS_2 attributed to many-phonon combination processes. In total, more than 15 higher order Raman modes are investigated, and their corresponding strain induced shift rates and mode Grüneisen parameters are determined. Using Density Functional Perturbation Theory, the two-phonon density of states was calculated and compared with the experiment.

References

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