Quantum interference in Raman scattering in few monolayer – MoTe₂

The resonant excitation of the Raman scattering results in rich spectra, which reflects the coupling of phonon modes to electronic states excited resonantly in a crystal. The Raman scattering in semiconductor transition metal dichalcogenides (TMDs) resonant with the A and B excitons related to the fundamental bandgap has been thoroughly studied. Much less is known on the effect of excitation deep within the bands, in resonance with higher-energy minima of the TMDs bandstructure. The Raman scattering excited under such conditions in thin MoTe₂ layers results in a complicated pattern of the spectra due to out-of-plane (A₁₁g/A₁1') vibrations. Davydov-split modes of the vibrations can be observed [1-2]. Their number and the energy splitting reflects van der Waals interactions between monolayers of MoTe₂. We report on the effect of temperature (5K to 300K) on the Raman scattering due to A₁₁g/A₁1' modes associated with the out-of-plane modes in 1L, 2L, 3L, and 4L MoTe₂. The temperature-evolution of the modes critically depends on the flake thickness (see Fig. 1). Most striking is the evolution of the A₁₁g mode intensity observed in 2L MoTe₂. The intensity decreases with decreasing temperature down to 200K and the A₁₁g mode vanishes from the Stokes scattering spectrum in the temperature range between 150K and 200K (see Fig. 2). The peak recovers at lower temperatures and at T=5K it becomes three times more intense that at room temperature. Similar non-monotonic intensity evolution is observed for the out-of-plane mode in 3L MoTe₂ which tellurium atoms in all three layers vibrate in-phase. On the contrary, the intensity of the other out-of-plane Raman-active mode in which vibrations of tellurium atoms in the central layer of 3L MoTe₂ are shifted by 180° with respect to the vibrations in outer layers, only weakly depends on temperature. Similar although weaker effect can be observed in 4L MoTe₂. Originally we related the observed effect to the quantum interference between the contributions to bond polarizability due to resonant (electronic excitations at the M point of the Brillouin zone) and non-resonant components [3]. Both the nonresonant and the resonant terms can cancel out, which results in the observed quenching of the Raman scattering due to out-of-plane modes in thin MoTe₂ layers. More recently another model has been proposed [4] to explain the behavior. It has been shown that the resonant contributions from the region between K and M point of the Brillouin zone destructively interfere with the contributions from the K−Γ and M−Γ regions. We discuss our results within both models. We argue that because of the substantial joint density of states the contribution to the bond polarizability from the transitions at K point of the Brillouin zone cannot be neglected. In our opinion the quantum interference of the contribution from M and K points of the Brillouin zone is responsible for the observed quenching of the Raman scattering due to out-of-plane vibrations observed in our experiment.

References

**Figures**

**Figure 1:** Temperature-dependent Raman spectra of MoTe$_2$

**Figure 2:** Temperature dependence of the relative intensities of the $A_{1g}/A_1'$ – related peaks in MoTe$_2$ in the Raman scattering spectra excited with 1.96 eV ($\lambda=632.8$ nm). Relative intensities of peaks due to in-phase $A_{1g}/A_1'$ modes in the Raman scattering spectra excited with 2.41 eV ($\lambda=514.5$ nm) are also shown with closed green circles.