Adam Babinski
K. Golasa, M. Grzeszczyk
Faculty of Physics, University of Warsaw, ul.Pasteura 5, 02-093 Warsaw, Poland
M.R.Molas, K.Nogajewski, M.Potemski
LNCMI CNRS-UGA-UPS-INSA-EMFL, 25 rue des Martyrs, 38042 Grenoble,France,

babinski@fuw.edu.pl

Quantum interference in Raman scattering in few monolayer – $MoTe_2$

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 $MoTe_2$ layers results in a complicated pattern of the spectra due to out-of-plane (A_{1g}/A_1') 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 $MoTe_2$. We report on the effect of temperature (5K to 300K) on the Raman scattering due to A_{1g}/A_1' modes associated with the out-of-plane modes in 1L, 2L, 3L, and 4L $MoTe_2$. The temperature-evolution of the modes critically depends on the flake thickness (see Fig. 1). Most striking is the evolution of the A_{1g} mode intensity observed in 2L $MoTe_2$. The intensity decreases with decreasing temperature down to 200K and the A_{1g} 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 $MoTe_2$ in 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 $MoTe_2$ 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 $MoTe_2$. 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 $MoTe_2$ 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 disscuss 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 Brilloin 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

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[2] G. Froehlicher, E. Lorchat, F. Fernique , et al. Nano Lett. 15 (2015) 6481.
[3] K. Golasa, M. Grzeszczyk, M.R. Molas, M. Zinkiewicz et al. Nanophotonics, (2017) DOI 10.1515/nanoph-2016-0150.
[4] H. P. C. Miranda, S. Reichardt, G. Froehlicher et al. Nano Lett. 17 (2017), 2381.

Figures

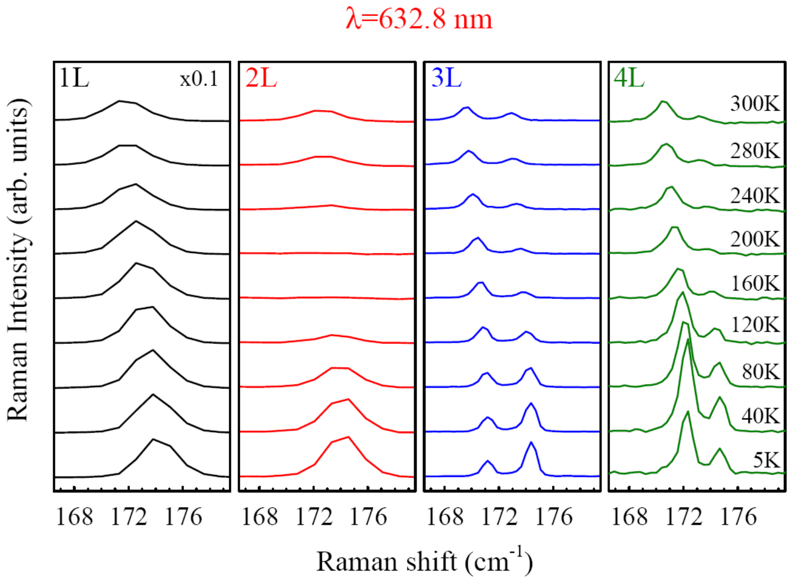

Figure 1: Temperature-dependent Raman spectra of $MoTe_2$

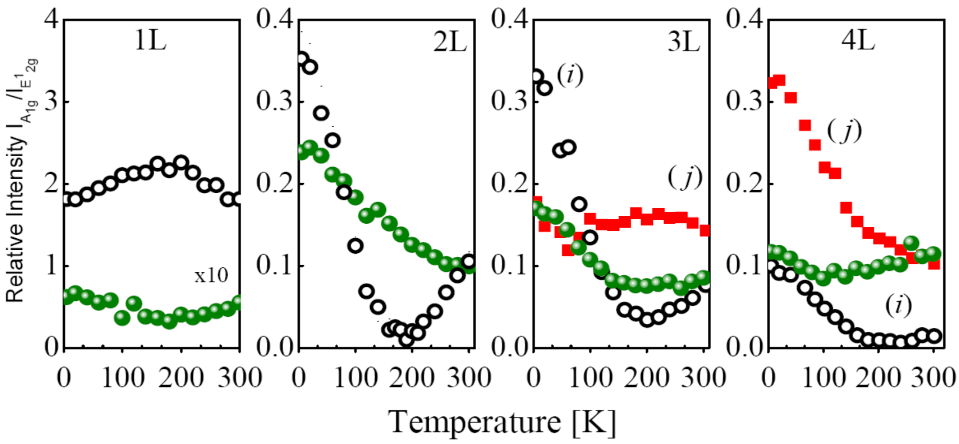

Figure 2: Temperature dependence of the relative intensities of the A_{1g}/A_1' – related peaks in $MoTe_2$ in the Raman scattering spectra excited with 1.96 eV (l=632.8 nm). Relative intensities of peaks due to in-phase A_{1g}/A_1' modes in the Raman scattering spectra excited with 2.41 eV (l=514.5 nm) are also shown with closed green circles.