Resonant Raman scattering in MoS_2– from bulk to monolayer

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Studies of low dimensional structures become one of the most intensely investigated areas of the material research in recent years [1]. With a seminal example of graphene, layered transition metal chalcogenides draw attention of researchers. Molybdenum disulfide (MoS_2) is a member of the family. Raman scattering spectroscopy appears to be a technique of choice to study its properties. The non-resonant Raman spectrum of MoS_2 is dominated by two basic vibrational modes: E^1_{2g} and A_{1g}. The sensitivity of those peaks to the number of layers has been employed in a standard method of a thin MoS_2 [1] layer characterization.

More complex is the optical spectrum due to the resonant Raman scattering in which second-order processes, enhanced by the electron-phonon interaction are more effective than the modes related to the first-order Raman scattering processes. While the resonant Raman scattering in bulk MoS_2 has been widely investigated, there are only a few reports on the effect of the MoS_2 layer thickness on the resonant Raman scattering [2].

We analyze experimental spectra obtained on 1ML, 2ML, 3ML and bulk MoS_2 at room temperature (see Figure). In agreement with previous studies [2] we observe substantial changes of the spectrum with decreasing flake thickness. In particular higher order processes become less effective with decreasing number of layers. Moreover similar characteristic change in the line shape of Raman peaks at ~460 cm^{-1} and ~640 cm^{-1}can be seen. The bimodal structure of those peaks becomes evident on flakes of a monolayer thickness. In our opinion the behavior can be understood in terms of recently proposed attribution of Raman peaks to multiphonon replica involving transverse acoustic phonons in MoS_2 [3]. According the attribution the high-energy components of the ~460 cm^{-1} and ~640 cm^{-1} peaks are due to combined E_{1g} (M) +XA(M) and E_{1g} (M) +2 XA(M) processes respectively with XA – the transverse acoustic phonon from M point of the Brilloiun zone. Their intensity decreasing with the decreasing number of layers suggests that the processes involving transverse phonons become less efficient in thin flakes. This is in contrary to combined processes involving longitudinal acoustic phonons, with Raman peaks well resolved also in a 1 ML sample. In our communication we also report on low-temperature thickness-dependent measurements of resonant Raman scattering in MoS_2, which further support our model.

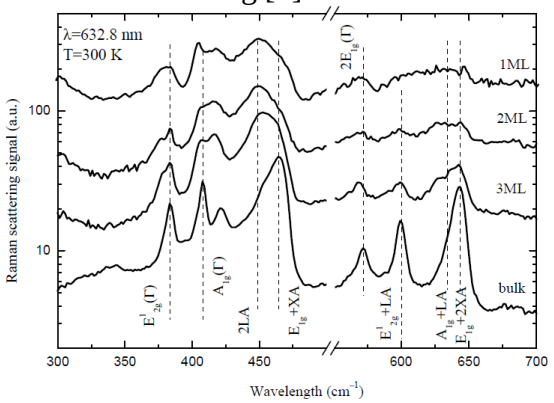

Fig. Resonant Raman spectra of MoS_2 flakes of different thickness measured at room temperature. The attributed phonons originate from M point of the Brillouin zone unless stated otherwise.

[1] C. Lee et al., *ACS Nano* **4**, 2695 (2010).
[2] A. B. Chakraborty et al., *J. Raman Spectr*. **44**, 92 (2013); H. Li et al., *Adv. Funct. Mater*. **22**, 1385 (2012).
[3] K. Gołasa et al., *Appl. Phys. Lett.* **104**, 092106 (2014).