Raman Scattering at Infrared Energies of Extremely Thin Semiconductors

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Since the first mechanical exfoliation of a single layer of graphite, two dimensional materials have attracted a lot of interest. In particular, transition metal dichalcogenides (TMD), one of the most promising class of layered materials, are currently intensively studied due to their very interesting electronic and optical properties. Raman spectroscopy is considered the golden technique to probe the fundamental properties of this kind of materials, therefore not only the vibrational properties can be investigated, but also the role of the electrons and electronphonon coupling can be extrapolated from Raman spectra. In particular, these latter information can be understood through the so called resonant Raman scattering, which involves a real electronic level instead of a virtual one. Raman spectroscopy studies of this material are widely present in literature, but a systematic study down to1.16 eV of excitation energy is still lacking. In fact, the Raman cross section scales with the inverse of the fourth power of the wavelength, hence the signal-to-noise ratio (SNR) at infrared wavelengths is very low and the detection of Raman scattering for few layers samples very challenging. We study the optoelectronic properties of molybdenum diselenide (MoSe2) by means of Raman spectroscopy using three different excitation energies (1.16 eV, 1.96 eV, 2.33 eV). We also vary the number of layers from bulk crystals to the monolayer (ML) limit. The very low SNR at infrared wavelength has been partially overcome by placing the flakes on top of hexagonal Boron Nitride (hBN) on a gold substrate: the constructive interference increases the signal of more than 30 times. By analyzing the second order Raman scattering, we attribute the high Raman shift peaks to a two-phonon scattering process partially resonant with the indirect band gap of MoSe2.