

Infrared resonance Raman of graphene: signatures of electron phonon coupling and of the band structure on the double resonant peaks

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Raman spectroscopy has been a key asset to study the electronic and vibrational properties of graphene and other two-dimensional materials[1, 2]. These systems display Raman spectra composed of first order modes together with narrow second-order double resonant modes arising from intervalley or intravalley scattering where two phonons or one phonon and one defect participate in the scattering of the excited electron/hole. Notably, by changing the excitation laser energy, different regions of the electron and phonon dispersions can be probed.

We report on resonance Raman spectroscopy measurements with excitation photon energy down to 1.17 eV on monolayer and bilayer graphene, to study how low-energy carriers interact with lattice vibrations. By lowering the excitation energy down to the Dirac point at \mathbf{K} , we unveil in the monolayer a giant increase of the intensity ratio between the double-resonant 2D and 2D' peaks with respect to that measured in graphite. Fully *ab initio* theoretical calculations, compared to the experimental results, allow us to conclude that the observation can be explained by an enhanced, momentum-dependent coupling between electrons and Brillouin zone-boundary optical phonons[3]. This finding applies to two dimensional Dirac systems and has important consequences for the modeling of transport in graphene devices operating at room temperature.

In bilayer graphene, the low excitation energy weakens some of the Raman processes and the sub-structures composing the resonance 2D peak in bi- and trilayer samples show a clearer spectral separation. We follow the excitation-energy dependence of the intensity of each sub-structure and, comparing experimental measurements on bilayer graphene with *ab initio* theoretical calculations, we trace back such modifications on the joint effects of probing the electronic dispersion close to the band splitting and enhancement of electron-phonon matrix elements[4].

References

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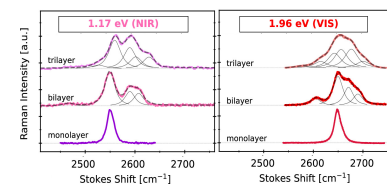


Fig. 1. Double resonant 2D peaks for monolayer, bilayer and trilayer graphene measured for red and infrared laser excitation energies.