Probing enhanced electron-phonon coupling in graphene by infrared resonance Raman spectroscopy

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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. 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. Here, we report on resonance Raman spectroscopy measurements with excitation photon energy down to 1.16 eV on graphene, to study how low-energy carriers interact with lattice vibrations. Thanks to the excitation energy close to the Dirac point at K, we unveil a giant increase of the intensity ratio between the double-resonant 2D and 2D' peaks with respect to that measured in graphite. Comparing with fully ab initio theoretical calculations, we conclude that the observation is explained by an enhanced, momentum-dependent electron-phonon coupling between electrons and zone-boundary optical phonons. Our findings pose the basis for a correct modeling of transport in graphene devices operating at room temperature and, moreover, could apply for any two-dimensional Dirac system.