The Raman spectrum of Graphene in presence of highly excited charge carriers

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The equilibrium phonon properties of graphene are well described in terms of anharmonicity and electron-phonon interactions, and Raman scattering is the main avenue to their characterization [1]. The interaction with light initially generates an out of equilibrium distribution of (hot) electrons with respect to the (cold) phonon bath. Relaxation to thermal equilibrium occurs within a few picoseconds and, on the laboratory timescale, continuous wave laser sources commonly used for high resolution spontaneous Raman scattering probe equilibrated carrier-phonon distributions. Sub picosecond photoexcitation provides a way to impulsively localize energy in the graphene's electronic subsystem. While the response of hot charge carriers to such ultrafast perturbation has been thoroughly investigated [2,3], unravelling the behaviour of optical phonons under strongly out of equilibrium conditions remains a challenge. Using a 3-ps laser excitation, which trades off between impulsive stimulation and spectral resolution, we show how the Raman response of graphene can be detected in presence of an electronic subsystem temperature largely exceeding that of the phonon bath. The period and lifetime of both the G and 2D phonons as function of the carriers' temperature in the range 1700-3100 K indicates a broadening of the Dirac cones. We explain this by modelling the electron-phonon scattering in a highly excited transient regime, which is critical for the emerging field of graphene-based nanophotonics and optoelectronics.

References

[1] A. C. Ferrari, D. M. Basko, Nat. Nanotechnol., 8 (2013), 235
[2] C.H. Lui, K.F. Mak, J. Shan, T.F. Heinz, Phys. Rev. Lett., 105 (2010), 127404
[3] D. Brida et al., Nat Commun, 4 (2013), 1987.

Figure

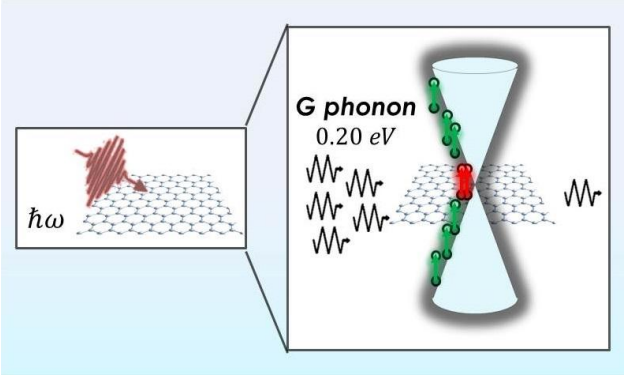

Figure 1: Picosecond excitation determines the smearing out of the Dirac cone due to the large electronic temperature. Consequently, the e-ph coupling is modified by new intraband processes which enhance the phonon absorption cross section, determining a lifetime reduction of the G phonon.