Unconventional Plasmon-Phonon coupling in Graphene

Marinko Jablan[1], Marin Soljacic[2], and Hrvoje Buljan[1]

[1]Department of Physics, University of Zagreb, Bijenicka c. 32, 10000 Zagreb, Croatia
[2]Physics Department, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge MA 02139, USA
mjablan@phy.hr

The interaction of collective vibrations of crystal lattice (phonons) and electrons has fundamental implications on properties of materials. This interaction takes an unusual form in graphene [1] leading to breakdown of Born-Oppenheimer approximation [2] or the anomaly of the optical phonon [3]. In light of this peculiar electron-phonon interaction, and recent interest in collective excitations of electrons in graphene – plasmons (e.g., see [4] and Refs. therein), here we consider plasmon-phonon coupling in graphene. We predict [4] that plasmon-phonon coupling occurs with a peculiar crossing of polarizations: longitudinal plasmons couple exclusively to transverse optical phonons, whereas graphene's transverse plasmons couple only to longitudinal optical phonons. This is calculated within the framework of the self-consistent linear response formalism.

To understand physical mechanism behind this unusual crossing of polarizations, let us look at the band structure of graphene. Near high symmetry K point (shown in Fig. 1b.) electrons are described by Dirac cones with effective Hamiltonian given by the tight-binding approximation [1]:

$$H_e = \hbar v_F \vec{\sigma} \cdot \vec{k} \quad (1).$$

If we now imagine a collective oscillation of electrons, i.e. a plasmon, we can describe it with a mean field vector potential $\vec{A}$ (scalar potential is gauged to zero) which acts on the electrons in a self-consistent way. To quantify this interaction we use a general substitution $\hbar \vec{k} \rightarrow \hbar \vec{k} + e\vec{A}$ in Eq. (1), so that electron-plasmon interaction can be written as:

$$H_{e-pl} = e v_F \vec{\sigma} \cdot \vec{A} \quad (2).$$

Turning our attention to phonons let us first assume that $\vec{u} = (\vec{u}_A - \vec{u}_B)/\sqrt{2}$ denotes a small displacement of the atoms A and B in the basis as shown in Fig. 1c. Within tight-biding approximation this results in the interaction Hamiltonian given by (see [3] and Refs. therein):

$$H_{e-ph} = -\sqrt{2} \frac{\beta \gamma}{b^2} \vec{\sigma} \times \vec{u} = e v_F \vec{\sigma} \cdot \vec{A}_{eff} \quad (3).$$

When comparing this to Eq. (2) one can see that Eq. (3) is equivalent to the presence of an effective vector potential $\left| \vec{A}_{eff} \right| \propto \left| \vec{u} \right|$ which simply shifts original Dirac cone to a new position as show in Fig. 1d.

The peculiar property of graphene is that vector $\vec{A}_{eff}$ is perpendicular to the displacement vector $\vec{u}$ so that the oscillation of logitudinal (transverse) optical phonons creates an effective vector potential, and thereby an effective electric field, in the transverse (longitudinal) direction. On the other hand, since longitudinal (transverse) plasmons are accompanied by longitudinal (transverse) electric field this will lead to the mentioned crossing of polarization of the two collective excitations. The plasmon-phonon coupling will be greatest when phonon energy and momentum match that of the appropriate plasmon mode since then the effective electric field created by phonon will have a huge response due to collective motion of electrons. These points are clearly shown in Fig. 2.

Finally we note that the frequency shift of the transverse optical phonon due to interaction with collective electron excitations (Fig. 2a.) is much larger than the one where phonon interacts with single particle excitations which was recently measured in the Raman experiment [2]. In this context plasmon-phonon coupling can serve as a magnifier for exploring the electron-phonon interaction in graphene, while it also offers novel electronical control over phonon frequencies. To point out the technological implications we note that there has been an increased interest for plasmons in graphene in the context of plasmonics and metamaterials and we have shown in another paper [5] that plasmon-phonon interaction plays an important role there.

References

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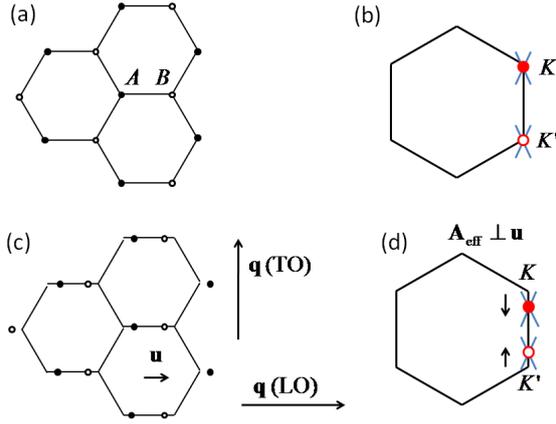**Fig. 1.** (a) Bravais lattice of graphene with two atoms *A* and *B* in the basis. (b) Brillouin zone with Dirac cones at *K* and *K'* points. (c) displacement of basis atoms corresponding to the profile of optical phonon mode and (d) the resulting movement of the Dirac cones which can be described trough the action of effective vector potential $\vec{A}_{eff} \perp \vec{u}$. We also show in figure (c) the orientation of wave vector $\vec{q}$ for different polarizations of phonon modes.
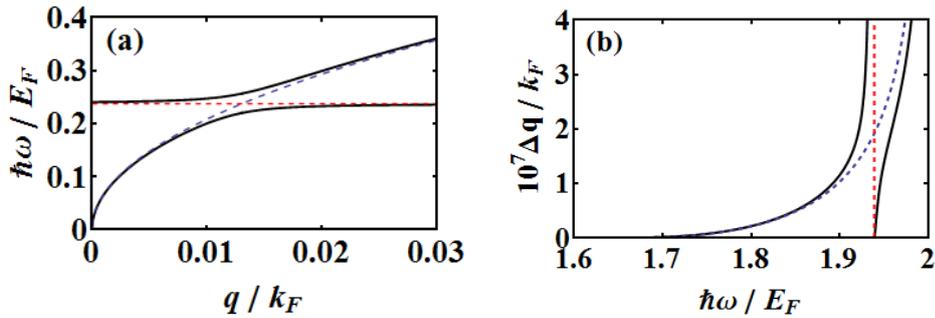

Fig. 2. Dispersion relation for coupled: (a) longitudinal plasmon-transverse optical phonon mode at doping value $E_F = 0.82eV$, and (b) transverse plasmon-longitudinal optical phonon mode for doping value $E_F = 0.1eV$. The coupled (uncoupled) modes are depicted by solid (dashed) lines and note that phonons are effectively dispersion-less for these wave vectors. Finally note that, since dispersion relation of transverse plasmons is very close to the light line, we plot $\Delta q = q - \omega/c$ on ordinate axis in figure (b).