Charge tuning of non-resonant magneto-exciton phonon interactions in graphene

Anna K Swan, Sebastian Remi, Bennett Goldberg
Boston University, 8 St Mary's street, Boston MA 02215
swan@bu.edu

Abstract

We explore the interactions between the G-band phonon and Landau level (LL) excitons far from energy resonance in graphene[1].

In a strong perpendicular magnetic field, the graphene electron energy bands split into discrete Landau levels. It has been shown earlier that when the G-band phonon energy overlaps with a LL exciton energy, the phonon and LL exciton can mix via electron-phonon coupling, and the interaction on resonance is well described by a single 2 level coupled mode description[2]. Anti-crossings at resonance have been observed by magneto Raman spectroscopy of decoupled surface layers of graphene on graphite [3-and more recently on exfoliated graphene on SiO_2 [6]

Here we show for the first time charge carrier density dependent magneto Raman measurements on single layer graphene field effect devices at constant magnetic fields. Contrary to previous magneto-phonon studies, we explore the Raman response in a magnetic field far from magneto-phonon resonance conditions. We demonstrate a linear dependence on the phonon frequency with the LL filling, ν, for the non-resonant regime, rather than the $\sqrt{\nu}$ behavior predicted for the on-resonance response. Notably, at a *constant* magnetic field, we observe pronounced splittings and slope changes of the G-band phonon energy as a function of ν. The coupling of the G-band phonon to magneto-excitons in single layer graphene displays kinks and splittings versus filling factor that are well described by Pauli blocking and unblocking of inter- and intra- Landau level transitions.

In order to model this dependence, we linearize the equation for the phonon shift as a function of magnetic field and charge density. We use values of the Fermi velocity and the electron-phonon coupling strength from zero magnetic field measurements of the phonon energy shifts due to electronic screening by charge carriers, Fig 1. We observe signatures of the logarithmic divergence of the phonon energy as a function of the charge carrier density at liquid He temperatures after vacuum annealing on single layers. Fig 1 (a). The fit of the measured dependence on charge density yields the values of the electron-phonon coupling strength,λ, and the Fermi velocity v_F. These values form zero magnetic field is then used, without further fitting, in the linear model to predict the dependence of the phonon energy in a magnetic field as function of the charge density, as is shown in fig 2c). The qualitative and quantitative agreement between spectra and a linearized model of electron-phonon interactions in magnetic fields is shown.

The reason for the kinks in the phonon energy is understood by considering the effect of filling and emptying LL, and hence blocking or turning on different symmetry allowed transitions. This is illustrated in Fig 2 (a) and (b). Fig 2 (a) shows the dependence on circular polarization of the incident laser light. In this example, the n=0 LL is fully filled, which blocks the σ- transitions between n=-1 to 0, and for all lower n's. On the other hand, the equivalent σ+ transition from n=0 to 1 is fully on. The effect on the phonon energy for the different σ+ transitions as a function of filling is shown in Fig 2 (b). The splitting of the Raman G band is caused by dichroism of left and right handed circular polarized light due to lifting of the G-band phonon degeneracy, as illustrated in Fig 2 a). The piecewise linear slopes are caused by the linear occupancy of sequential Landau levels. Fig 2 b) illustrates the dependence of ν on the different resonances. This is in contrast to earlier work on-resonance, where the Landau level filling dependence is proportional to $\sqrt{\nu}$. In contrast to on-resonance measurements, no single transition dominates the coupling, and several inter and intra-band transitions have to be considered to account for the experimental observations.

By focusing on the non-resonant regime we have discovered fine structure in the G-band optical phonon in single layer graphene at high magnetic fields as a function of charge density. The observed behavior is caused by coupling between the phonon and magneto-exciton far from resonance that result in a linear dependence on filling factor, in contrast to on-resonant coupling that leads to a square root dependence on filling factor.

References

[1] Remi, Goldberg and Swan Phys. Rev. Lett., Journal, **112** (2014), 056803.
[2] M. O. Goerbig, Rev. Mod. Phys. **83**, (2011) 1193.
[3] M. Kühne, C. Faugeras, P. Kossacki, A. A. L. Nicolet, M. Orlita, Y. I. Latyshev, and M. Potemski, Phys. Rev. B **85**, (2012), 195406.
[4] C. Faugeras, M. Amado, P. Kossacki, M. Orlita, M. Kühne, A. A. L. Nicolet, Y. I. Latyshev, and M. Potemski, Phys. Rev. Lett. **107,** (2011), 036807.
[5] J. Yan, S. Goler, T. D. Rhone, M. Han, R. He, P. Kim, V. Pellegrini, and A. Pinczuk, Phys. Rev. Lett. **105**, (2010), 227401.
[6] Y. Kim, J. M. Poumirol, A. Lombardo, N. G. Kalugin, T. Georgiou, Y. J. Kim, K. S. Novoselov, A. C. Ferrari, J. Phys. Rev. Lett., **112**, (2014) 056803-4
[7] Kono, O. Kashuba, V. I. Fal'ko, and D. Smirnov, Phys. Rev. Lett. **110**, (2013). 227402.

Figures

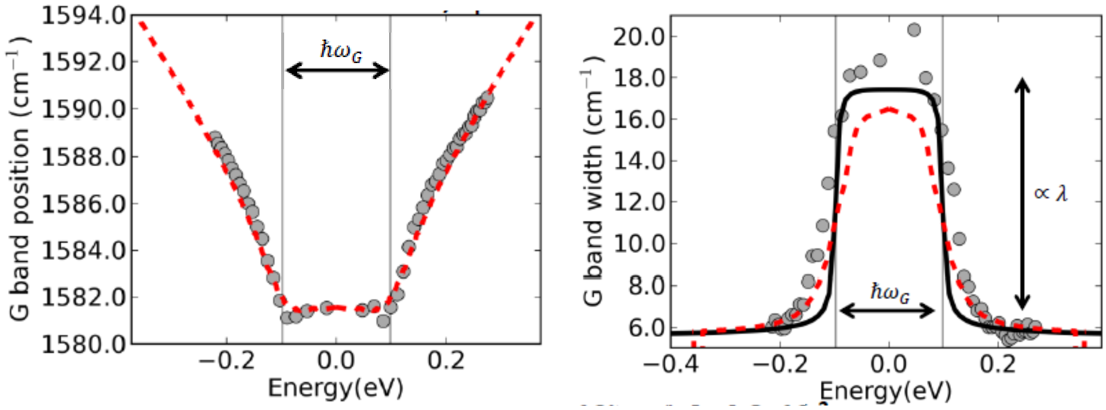

Fig. 1. G band position (a) and linewidth (b) as a function of of gate voltage at zero magnetic field. The fit gives the values for the electron phonon coupling strength from the slope in (a), and v_F from matching the phonon energy to the logarithmic divergence. Charge puddling δn is masking the depth of the divergence in a). $v_F=1.1*10^8$ cm/s, $\lambda= 4.8*10^{-3}$ and $\delta n=0.3*10^{12}/cm^2$.

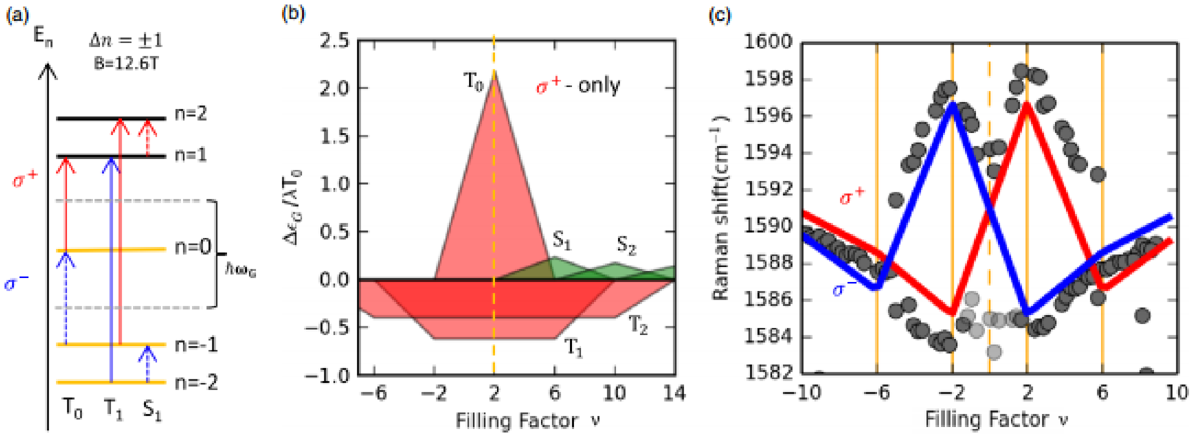

Fig 2. a) Schematic view of the Landau level spectrum at B=12.6T, filling factor ν=2 and the lowest Landau level transitions participating in magneto-phonon coupling. Filled electronic states are highlighted using orange color. Red and blue arrows show transitions allowed by the selection rule $\Delta|n|=\pm1$. Dashed arrows mark Pauli blocked transitions. Circular arrows represent the angular momentum involved in the transitions b) Relative strength and filling factor dependence of individual terms of the phonon self energy for σ+ transitions. Terms describing interband transitions are shaded red, intraband transitions are shaded green. c) Phonon energy as a function of the filling factor at B=12.6T. Vertical orange lines mark specific filling factors at ν =-6,-2,0,2,6 where the n=-1,0,1 levels are completely filled/depleted with charge carriers (ν=0 corresponds to half filling of n=0 level). The calculated magneto-phonon energies are plotted as solid red ($\Delta n=+1$) and solid blue ($\Delta n=-1$) lines.