High resolution magneto-Raman microscopy of Graphene at low temperatures and high magnetic fields

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Abstract The electronic properties of Graphene are very much reminiscent of high quality layered semiconductors devices. Such two dimensional layered systems do exhibit striking electrical and optical properties when submitted to strong magnetic fields and cryogenic temperatures. Very pure graphene can be obtained in form of tiny flakes of few μm^2 by exfoliating the surface of high quality natural single crystal of graphite. The operation of exfoliation leaves behind high quality flakes of graphene on the surface of the natural graphite substrate. The flake are however so small that they require the use of local probes techniques to detect them and measure their properties. We have designed and built a high resolution confocal microscope capable of measuring spectroscopic optical Raman properties of surfaces at low temperature and high magnetic fields. We have based our optical microscopes instruments not only on standard liquid helium bath magnet cryostat but also on an ultra-low vibration closed-cycle magnet cryostat system, and this in order to gain total independence from the need of liquid cryogens enhancing this way enormously the ease of use of the system as well as the logistics usually involved with liquid based cryostat.

In this presentation we report on sub-micro magneto-Raman scattering experiments performed on the surface of a freshly exfoliated single crystal of natural graphite [1]. Graphene flakes left on graphite are expected to be of very high electronic quality but are not easy to spot since they show no contrast in standard optical microscopy. In this work we image natural graphene flakes using high spatial resolution confocal Raman scattering microscopy in high magnetic fields (0-9 Tesla) at 7 K. Graphene flakes on graphite are revealed in the presence of a strong magnetic field, as first imaged in [1], when the E2g phonon energy coincides with the electron-hole separation between the valence and conduction Landau levels (-N,+M) of the Dirac cone. Resonant hybridization of the E2g phonon and the Dirac fermion magnetoexciton is a specific signature of graphene flakes [2-3] and display very rich Raman scattering spectra varying strongly as a function of magnetic field [1]. In the figures below the magnetic field evolution of Raman spectra are taken in region where the hybridization between the E2g phonon and the (-2,+1) and (-1,+2) magneto-exciton takes place. We map the Raman scattering over 7x7 µm with 600 nm spatial resolution on three different scattering bands namely i) centered, ii) blue-shifted and iii) red-shifted from the E2g phonon peak and this at 4.3 T (lower three images) and 5.3 T (upper three images). These two magnetic fields are chosen to be just bellow and just above the resonant conditions for hybridization. As expected at 4.3 T the graphene flake appears bright in the blue shifted image (lower right), it appears bright in the red shifted image at 5.3 T while it is darker in the Raman scattering mapping centered on the E2g (both center images). The upper right and lower left image have been shown here for completion with a much enhanced contrast.

References

[1] C. Faugeras et al., M. Amado, P. Kossacki, M. Orlita, M. Kühne, A.A.L. Nicolet, Yu. I. Latyshev, and M. Potemski, Phys. Rev. Lett. 107, 036807 (2011).
[2] C. Faugeras et al. Phys. Rev. Lett. 103, 186803 (2009).
[3] J. Yan et al. Phys. Rev. Lett. 105, 227401 (2010)

Figures

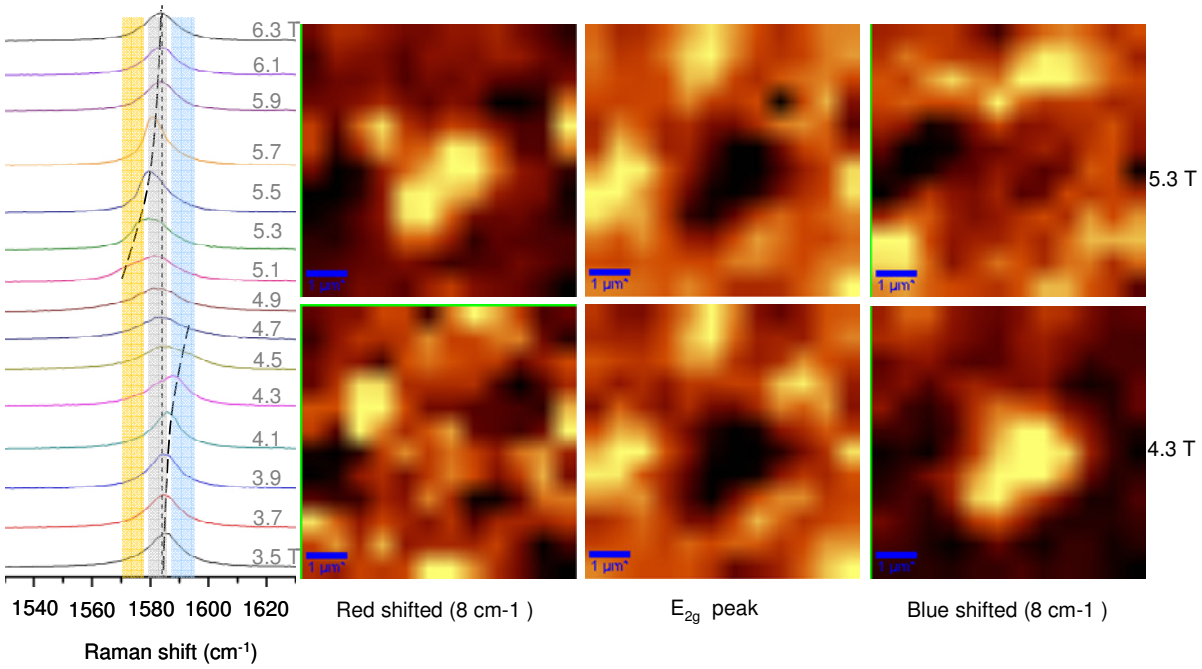

Figure 1*:* Left: series of Raman spectra of the E2G phonon peak evolving as a function of the magnetic field. The spectra are shifted vertically for clarity. The spectra shows a contribution in the Raman peak that clearly displays an avoided crossing behavior centered at about 4.7 T. Right: series of images of the graphene flakes a two different fields (5.3 and 4.3T) and three different Raman bands (see text for details)

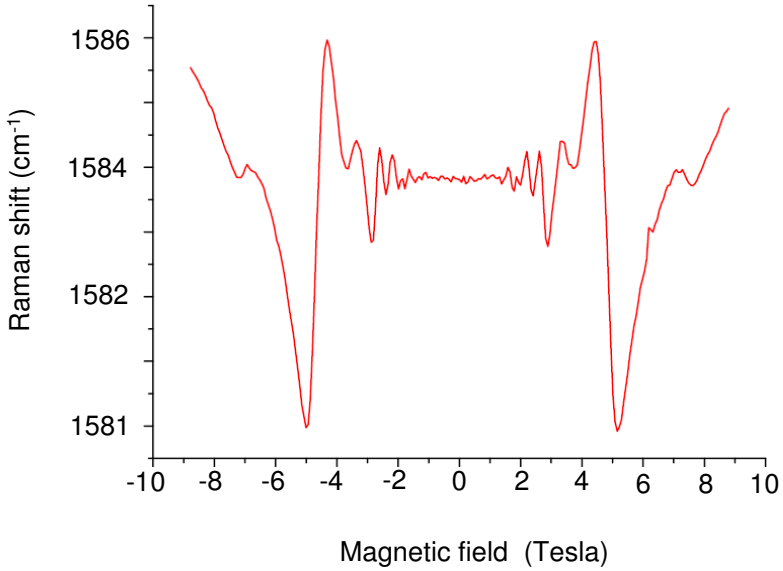

Figure 2: Evolution of the center of mass of the Raman peak of the E2G band as a function of the magnetic field between -9T and +9T. The oscillations are a clear signature of the graphene flake. The same type of measurement done away from the graphen shows no shifts within the spectral resolution.