Raman and Electronic-Raman Scattering Measurements under High Magnetic Fields and Cryogenic Temperatures

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Introduction

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 at cryogenic temperatures and under strong magnetic fields.

Very pure graphene can be obtained in form of tiny flakes of few µm² by exfoliating the surface of high quality natural single crystal of graphite. In many cases the flakes are however so small that a micro-Raman setup is necessary to localize them and to measure their properties. We have designed and built a high resolution confocal microscope capable of Raman spectroscopy of surfaces at low temperature and high magnetic fields.

Experimental Setup

The attocube cryogenic Raman spectroscopy system combines a high resolution, low temperature confocal microscope with an ultra-sensitive Raman spectrometer. This innovative product enables state-of-the-art confocal Raman measurements at cryogenic environments and can be combined with liquid Helium cryostats (magnetic fields up to 16 T) or cryogen-free cryostats (magnetic fields up to 9 T). The attoRAMAN is a ready-to-use system and comes typically with a Raman laser source (frequency doubled or tripled Nd:YAG [532 nm, 355 nm] or HeNe [632.8 nm]), an ultra-high throughput spectrometer including a Peltier-cooled, back-illuminated CCD, and a state-of-the-art Raman controller/software package.

The microscope uses a set of attocube xyz-positioners for coarse positioning of the sample over a range of several mm. Developed particularly for cryogenic applications, the piezo scanner ANSxy100 provides a scan range of 30 x 30 µm² even at liquid helium temperature.

Measurement Results

Raman spectra have been acquired on a single exfoliated graphene flake with a 532 nm laser source at 7 K. A typical result is presented in Fig. 3: the green spectrum was acquired at B = 0 T field and shows the characteristic E2g and D bands. With high magnetic field applied (B = 9 T) an electronic Raman band appears at around 1800cm^{-1}, the energy of which depends on the field strength.

In the presence of magnetic field the continuous spectrum of electronic excitations becomes discrete and splits into excitonic Landau levels characteristic of a 2D system. By sweeping the magnetic field, optically active inter-Landau level transitions are tuned in resonance with the E2g phonon excitation, resulting in avoided crossings in the field evolution of the graphene Raman spectra. The signature of electron-phonon hybridization is presented in the following measurements.

Field evolution of Raman spectra on graphene

We map the Raman scattering signal over an area of 7 x 7 µm² centered onto a single graphene flake with 0.5 µm spatial resolution. Fig. 4 shows different scattering bands – namely red-shifted (left), centered (center) and blue-shifted (right) from the E2g phonon peak at B = 4.3 T and B = 5.3 T. These two magnetic fields are chosen to be just below and just above the resonant condition for hybridization. As expected, at B = 4.3 T the graphene flake appears bright in the blue shifted image (lower right), it appears bright in the red shifted image at B = 5.3 T, while it is dark in the Raman scattering mapping centered on the E2g (both center images).

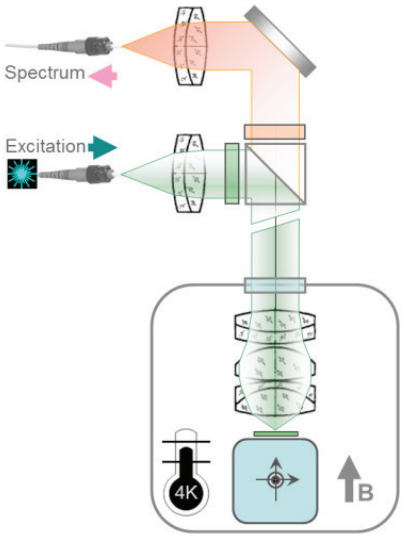

Figure 1: *Schematics of the confocal microscope.*

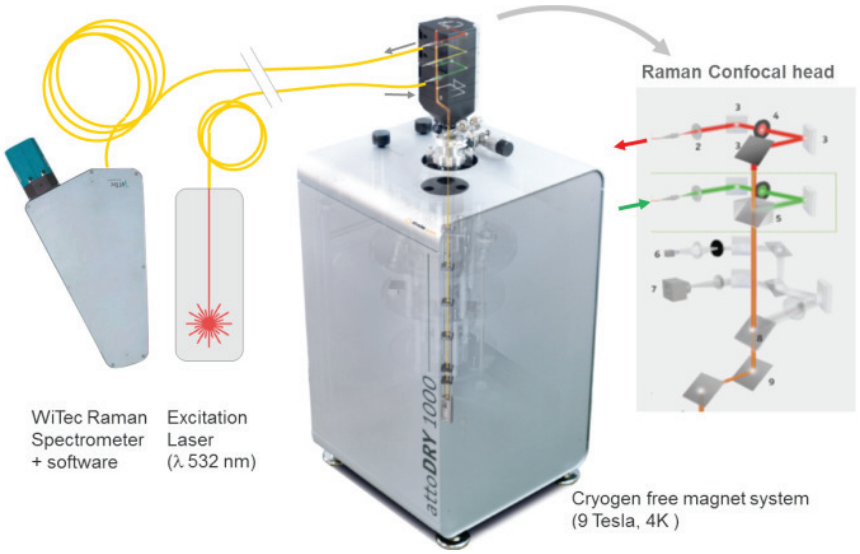

Figure 2: *Low temperature and high magnetic field Raman setup in the configuration with a 9 T cryogen-free cryostat.*

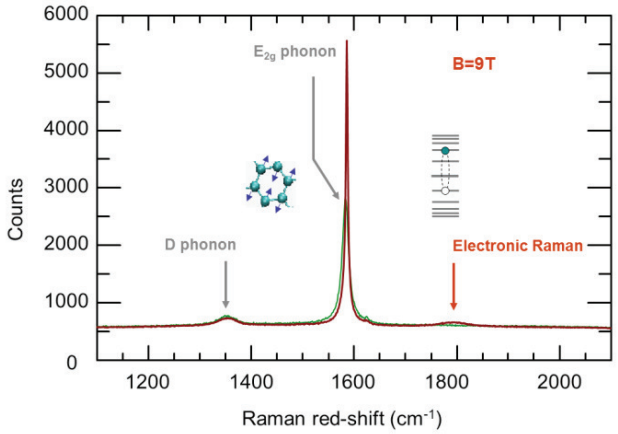

Figure 3: *Raman spectra of graphene acquired at 7 K and at B = 0 T (red) and B = 9 T (green) showing a magnetic field dependent band at 1800 cm^{-1}.*

The lower right side of Fig. 4 shows the magnetic field evolution of Raman spectra recorded in the center of the graphene flake where the hybridization of E2g phonon and (-2, +1) and (-1,+2) magneto-exciton takes place. For comparison, the E2g phonon band on the graphite substrate just a few micrometers away doesn't show any dependence on the magnetic field (left side).

The evolution of the E2g band while sweeping the magnetic field from -9 T to +9 T is shown in Fig. 5. As expected the data show a series of avoided crossing when the E2g phonon energy is resonant with the electron-hole separation between the valence and conduction Landau levels (-N, +M) of the Dirac cone. Furthermore, the field dependent electronic Raman features in the spectra and their field dependence are clearly resolved.

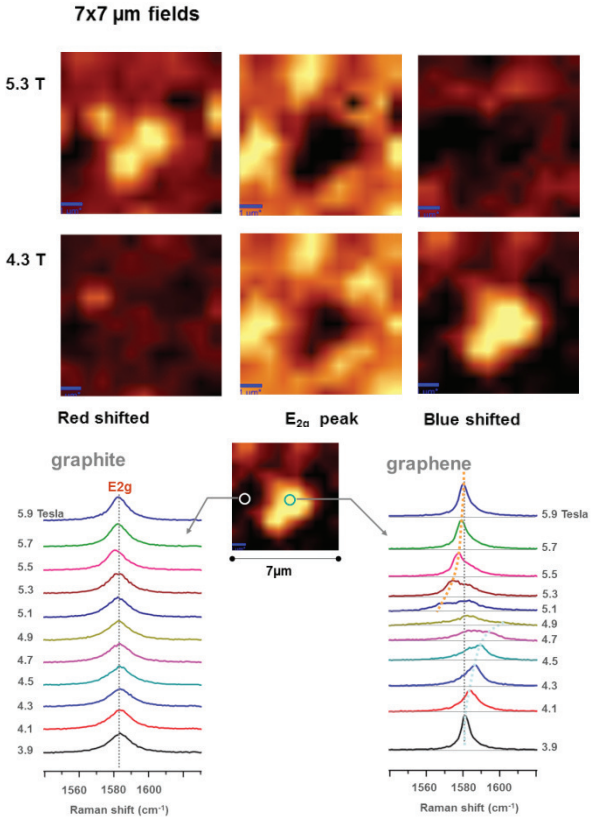

Figure 4: *Raman mapping of a graphene flake on graphite. The magnetic fields values (B = 4.3 T and B = 5.3 T) are chosen to be just below and just above the resonant condition for electron-phonon hybridization.*

Summary

Magneto-Raman measurements were performed with unprecedented spatial resolution (0.5 µm) on an exfoliated single crystal of natural graphite, exposed to magnetic fields of up to 9 T at 7 K. The data were recorded on a single graphene flake and clearly show electron-phonon resonances in the spectra. Resonant hybridization of the E2g phonon with excitons is a specific signature of graphene flakes which display very rich Raman scattering spectra varying strongly as a function of magnetic field [1].

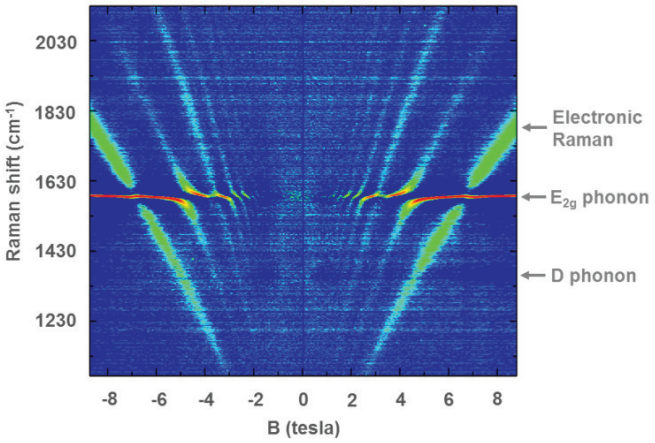

Figure 5: *Micro-Raman spectra of a graphene flake as function of the magnetic field. The spectrum at B = 0 T has been subtracted.*

References

[1] C. Faugeras, M. Amado, P. Kossacki, M. Orlita, M. Kühne, A.A.L. Nicolet, Yu. I. Latyshev, and M. Potemski, *„Magneto-Raman Scattering of Graphene on Graphite: Electronic and Phonon Excitations"*, Phys. Rev. Lett. **107**, 036807 (2011).

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