Charge transfer between graphene and fullerene C_{60}

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Abstract: Graphene consists of only one layer of carbon atoms, thus the system is under the influence of surrounding area; it had been probed experimentally that electrical properties of graphene undergo limitations when it is on top of SiO2/Si substrate [1], which is the common substrate used in optical observations and in field effect transistors measurements. Another way to modify electrical properties of graphene, is doping it with other atoms (substitutional doping), adsorbed molecules on its surface (surface transfer doping) [2] or electrochemical top gating [3]. These processes permit us to obtain p and n-type graphene. p-type doping drives the Fermi level of graphene below the Dirac points, and n-type drives the Fermi level above the Dirac points. Functionalization of graphene increases applications of this material, particularly in thin film transistors, optoelectronics and transparent conducting films, for example.

On the other hand, fullerene C_{60}, a three dimensional structure of carbon atoms, presents some interesting physical properties, it can create electron-hole pairs after excitation with light. In this context we are interested in charge transfer between these two materials, graphene (or few layers of graphene, FLG) and C_{60} films deposited on its surface. To this end, we performed Raman spectroscopy and in situ electrical measurements.

Raman spectroscopy is a powerful tool in characterization of graphene; the shapes, intensities and positions of peaks are used to determine the number of layers, strain, doping, disorder, types of edge, and so on. Raman fingerprint of graphene is characterized by three mean peaks: D peak at around 1350 cm^{-1} due to the breathing modes of six-atom rings and requires defects for its activation; G peak at 1585 cm^{-1} corresponds to the high frequency E_{2g} phonon at the Brillouin zone center, and the D peak overtone 2D peak at around 2690 cm^{-1} [4]. In surface transfer doping of graphene samples, position of G and 2D peaks, Pos(G) and Pos(2D) respectively, upshift for p-type doping but for n-type doping, Pos(G) downshifts while Pos(2D) upshifts [2].

We obtained FLG films by CVD technique, using 25 μm thick copper foils, methane as the carbon precursor and ambient pressure. After CVD process, FLG film was removed from the Cu foil by etching in an aqueous solution of iron nitrate, then the samples were transferred to SiO_2/Si substrates to be analyzed by Raman scattering, and glass substrates were used to perform electrical measurements. We used a Nicolet Almega XR Spectrometer, 532 nm of laser excitation for Raman measurements. The FLG sample mainly consists of graphene and many isolated regions with few layers. We selected just graphene zones in FLG films for Raman measurements. In Figure a), Pos(2D) versus Pos(G) of graphene are plotted, with and without fullerene C_{60} deposited on graphene surface. As it is shown in this figure, Pos (2D) of graphene upshifts about 20 cm^{-1}, and this is because of the electrical influence of C_{60} [5], on the other hand, Pos(G) upshifts about 10 cm^{-1}, indicating p type doping.

For electrical characterization two parallel silver strips were evaporated on FLG film to form Ohmic contacts, then fullerene C_{60} film was thermally evaporated on FLG, at the same time electrical conductivity was in situ monitored in darkness. In Figure b) current versus thickness of C_{60} on FLG is shown. When molecules of C_{60} are depositing on FLG, the current increases up to a thickness of about 70 Å; for thickness higher than this value, current do not change any more. After evaporation of C_{60} on FLG, the electrical conductivity decreases upon exposition of the sample to monochromatic light of 514.5 nm (see figure c). This was an ex situ measurement in a different vacuum chamber.

Since we observe both Pos(G) and Pos(2D) upshift, we can suppose that electrons flow from graphene to C_{60}, thus C60 induce p-doping in graphene [6]. The intensity ratio of G and 2D peaks (I(2D)/I(G)) is also sensitive to doping [3], I(2D)/I(G) decreases for increasing charge concentration, and the same variations have been observed for our samples, from 3.5 in pristine graphene to 1.5 in graphene with fullerene C_{60} film

Our tentative description, in order to reconcile electrical observations with Raman results, is as follows. We know that using CVD technique, contaminants from solvent of copper, or water originates p type graphene samples. Thus, Fermi level is below the Dirac point in our pristine samples. Since in situ measurements show an increase of conductivity when C_{60} is deposited on FLG film surface; which means that the hole density increases, now Fermi level of FLG film downshift even more with respect to Fermi level in initial sample. When light is on, a decrease in the electrical conductivity means that hole concentration are decreasing, and now Fermi level upshifts, but never above the Dirac point of initial FLG film, thus maintaining the p-type doping character.

Finally, we effectively found that fullerene C_{60} induce p type doping on graphene. Of course more measurements are needed in order to understand and describe the behavior of our system under illumination.

References

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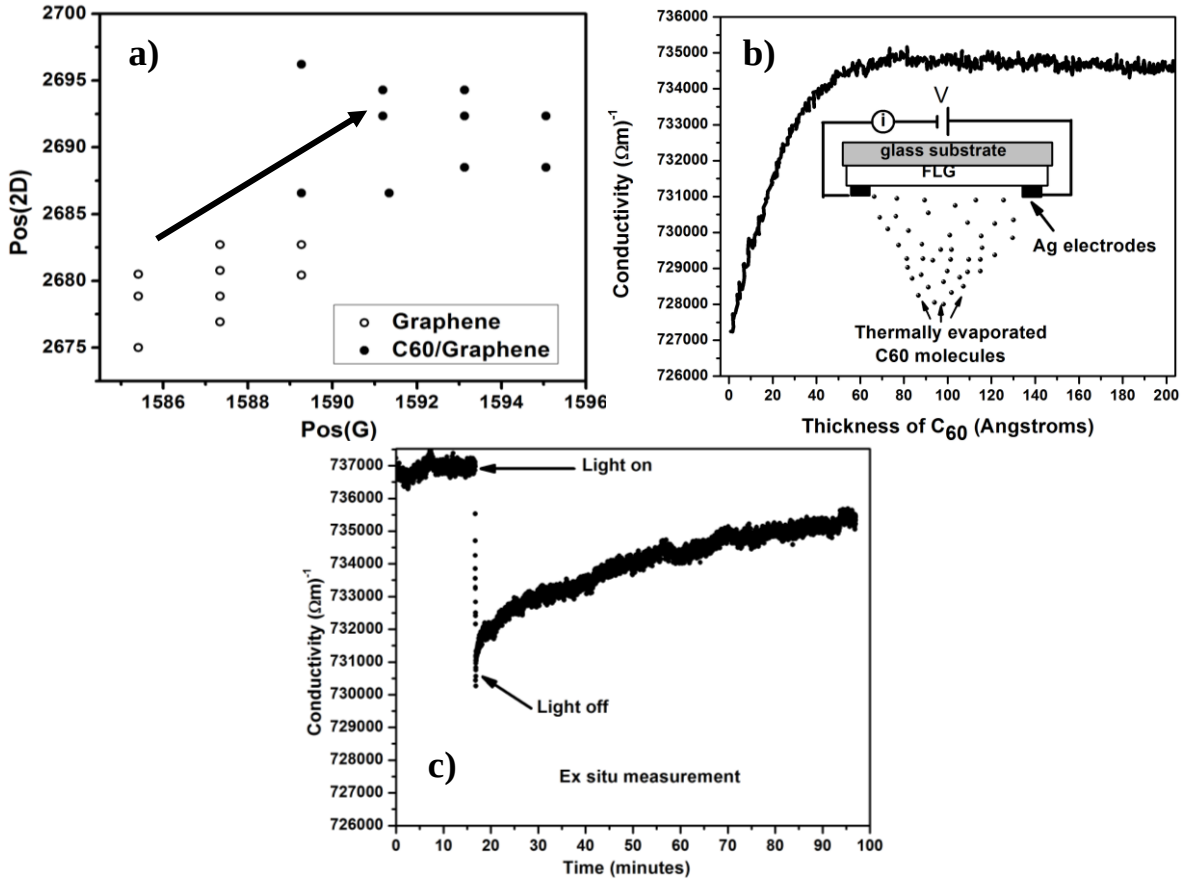
Figures



a) Position of G and 2D peaks of graphene with and without thermally evaporated fullerene C_{60}; b) Conductivity of FLG film versus thickness of C_{60} in darkness; c) Behavior of electrical conductivity of FLG film with C_{60} under illumination; for this experiment we used a 514.5 nm of laser excitation and 10 mW of Intensity.