Reversible optical doping of graphene [1]

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Abstract

The ultimate surface exposure provided by graphene makes it the ideal sensor platform but also exposes its intrinsic properties to any environmental perturbations. Raman spectroscopy has been used here to demonstrate the influence of the laser power and the surface chemistry of the SiO_2/Si substrate on the charge carrier density of exfoliated graphene in air.

Raman spectroscopy is being considered as a high-throughput technique to characterize graphene and to study its fundamental physical properties. This technique is highly sensitive to both the electronic and phononic structure of graphene and can probe the changes of these properties under external parameters such as doping, chemical modifications, or strain. The most intense Raman active bands in graphene are the G and 2D bands. In this work, we studied their evolutions in terms of shifts, widths and intensities as a function of the laser power impinging on graphene.
Results obtained on the same graphene flake with two different environments are compared in figure 1: (i) supported on a hydrophilic SiO_2/Si substrate, and (ii) suspended over a trench etched into this substrate. The relative variations of the 2D and G band positions as a function of the laser power are displayed. This specific representation disentangles doping and strain effects [2]. It is demonstrated that the charge carrier density of graphene exfoliated on a SiO_2/Si substrate can be finely and reversibly tuned between hole (p-type) and electron (n-type) doping, across neutral state (i-type) with visible photons. This effect is significantly affected by the substrate cleaning method and completely suppressed in suspended graphene. The observed phenomenon has a subsecond characteristic time and does not involve the chemical modification of graphene. The results were interpreted in terms of a local and reversible perturbation of the chemical equilibrium established between graphene, the substrate and the ambient atmosphere by laser-induced heating. Raman mapping experiments were also performed and allowed to show that these laser-induced doping variations are uniform across the graphene surface.

One technical implication of our study for the entire scientific community using Raman spectroscopy of graphene as a routine characterization technique is that it should be considered as potentially invasive as far as electronic properties are concerned. On the other hand, the ability to tune the charge carrier density with visible photons opens a wide set of opportunities to develop optically gated graphene electronic devices and a new approach to graphene optoelectronics. Finally, this effect should allow studying the interplay between graphene properties and the environment and triggering laser-assisted functionalization of graphene leading to more advanced devices.

References

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Figures

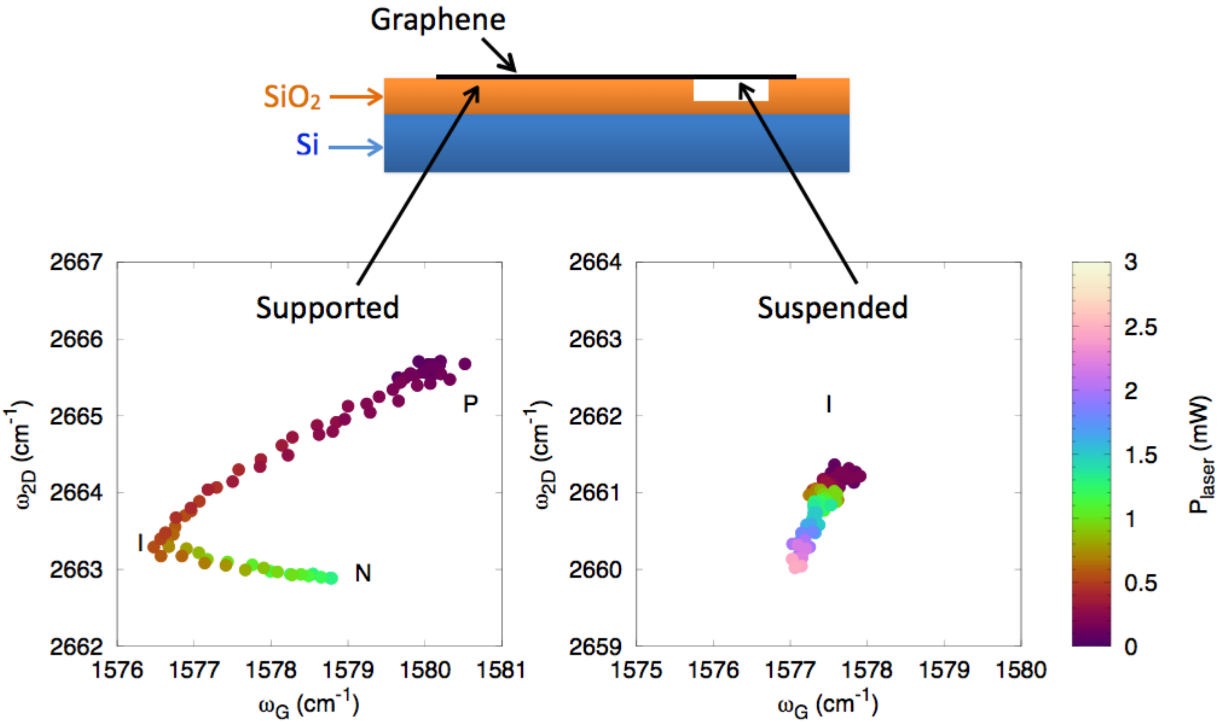

Figure 1 Comparison of the relative evolutions of the 2D band position (ω_{2D}) versus the G band position (ω_G) as a function of the laser power (P_{laser}) for supported and suspended graphene flakes. The color code of each point corresponds to the incident P_{laser} as displayed on the right hand side color bar. The supported flake is p-doped at low P_{laser}, it becomes quasi-neutral around 0.5 mW and n-doped for higher P_{laser}. The suspended graphene flake is neutral and stays neutral with the increasing P_{laser}. The measured shifts for the suspended flake are only due to laser heating effects. Each plot includes both increasing and decreasing power sweeps demonstrating the reversibility of this photodoping effect.