

RAMAN SPECTROSCOPY OF GRAPHENE IN DIFFERENT DIELECTRIC ENVIRONMENTS

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Graphene, a monolayer of sp^2 hybridized carbon atoms, has a unique electronic structure with a zero gap and quasiparticles behaving like massless Dirac fermions [1]. Its extremely high carrier mobility makes it a promising candidate for future electronics [1]. However, its carrier mobility has been shown to vary from sample to sample and to be limited to $\sim 10000 \text{ cm}^2/\text{Vs}$ [2,3]. Furthermore, unintentional doping and excess charges up to 10^{13} cm^{-2} have been reported for pristine graphene, deposited on silicon substrate covered with silicon oxide (Si/SiO_x), [4]. Thus, charged impurities seem to be the dominant source of charge scattering in graphene. For this reason, it is fundamental to investigate the origin of charged impurities. Since graphene is unprotected from the environment, the strength of scattering by charged impurities should be strongly dependent on the dielectric environment [3]. On the other side, charged impurities could be in the substrate. Indeed, the highest carrier mobility ($2 \times 10^5 \text{ cm}^2/\text{Vs}$) has been measured on suspended graphene [5].

Raman spectroscopy is a widely used method for the study of graphene: it can identify graphene [6], monitor the charge carrier concentration [7,8] and probe disorder [9] and edges [10]. In particular, the Raman G and 2D bands positions and intensities are sensitive to the amount of excess charges [4,11] and they can be used to distinguish between disorder and doping effects [9].

Here we use Raman spectroscopy in order to investigate the effect of the dielectric environment and the substrate on the charged impurities of graphene. Graphene samples were produced by micro-mechanical exfoliation of graphite and deposited on Si/SiO_x and Calcium Fluoride (CaF_2) substrates. Water, ethanol and chloroform, which have a relative permittivity of 80, 30 and 4.8 respectively, have been used as a top dielectric layers: a drop of dielectric was placed on graphene deposited on Si/SiO_x , and then the sample was covered with a glass slide to avoid evaporation of the dielectric during the Raman measurement. We observed strong variations in the Raman spectra during the first 30 minutes after immersion; then the spectra stabilize and no variations are observed until evaporation of the dielectric. We show that ethanol mainly introduces disorder in graphene, since graphene tends to roll and wrinkle, after interaction with ethanol; water and chloroform, in contrast, mainly produce chemical doping. In particular a strong p-doping has been observed in the case of chloroform, where an upshift of 15 cm^{-1} has been measured for the 2D peak position. Thus, a change in the environment can produce strong variations in the Raman spectrum, but the strength of this variation is not directly correlated with the change in permittivity.

We finally show the Raman spectrum of graphene deposited on CaF_2 . The analysis of the Raman fit parameters shows that graphene deposited on CaF_2 is un-doped and without disorder, and virtually comparable with suspended graphene. This suggests that the charged impurities are mainly related with the Si/SiO_x substrate.

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