Chemical Potential of Inhomogeneous Single Layer Graphene

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Many of the unique electrical properties of single layer graphene (SLG) have been attributed to the linear dispersion relation and linear density of states (DOS) of graphene. Several experiments conducted on SLG found the predicted non interacting DOS with a small correction to the inverse compressibility of graphene because of electron-electron interaction. In most of these studies the primary technique is based on measuring the change in the graphene chemical potential, $\delta\mu$, under periodic modulation of the gate voltage, δV_g, which is responsible for the change in the hole carrier density, δp, for example. Because the DOS and the compressibility are given by $\frac{\partial p}{\partial \mu}$, δp is usually extracted from either the gate bias or the Hall data. However, as the system is tuned towards the Dirac point, the charge carrier of the graphene breaks into puddles, in which electrons (n) and holes (p) coexist. Moreover, because μ is not a linear function of the carrier density, the gate bias induces unequal changes in the electron and hole densities. Consequently, for those experiments that directly measure the quantum capacitance, which is proportional to the compressibility, the density assignment is inadequate in the nonhomogeneous regime. Thus, these two obstacles may lead to inaccurate estimates of the electron-electron exchange and correlation contributions, which are of significant physical importance because of their many body origins.

A common method for the extraction of the densities of p and n is based on the two band model for the Hall effect. However, in the inhomogeneous regime, this method is inadequate because this model for the Hall effect was derived for situations in which these two carriers share the same region in space. However, this is not the case for the graphene. Although electrons and holes coexist near the charge neutrality point (CNP), at zero temperature they never share the same region in space. As will become evident later in this study, the disorder potential strength is $\approx 100\,\mathrm{meV}$; thus, even at room temperature, our assumptions that the fraction of minority carriers is negligible in the same puddle as the majority carriers is well justified.

Alternatively, it is possible to directly measure the chemical potential, and to derive from it the inverse compressibility and the DOS. For gated, p-type graphene far from the CNP, the relation between the external voltage and the chemical potential is given by

$$V_g = \frac{ep}{c_g} + \mu(p)/e + \phi_0, \tag{1}$$

where c_g is the geometric gate-graphene capacitance per unit area, and ϕ_0 is the electrical potential attributed to residual doping. Usually, the geometric capacitance is much smaller than the quantum capacitance, $c_q = e^2\frac{\partial p}{\partial \mu}$, and the chemical potential is a small contribution to Eq. 1; this small contribution makes measurements of the DOS challenging. However, if one dramatically increases c_g, either by using a thinner oxide layer, using a high dielectric material, or using both, the quantum term will no longer be negligible. In Fig. 1a, we depict the relative contributions of the geometric and quantum terms with respect to the total energy (eV_g), as a function of the carrier density, for two extreme cases. The first case, plotted in light blue (c_g) and magenta (c_q), reflects the relative contributions for a $300\,\mathrm{nm}$ SiO_2 gate oxide in which the quantum part is negligible. The second case (blue and red) shows the same two relative contributions for $2\,\mathrm{nm}$ of HfO_2, which was one of the gate oxides that we used in this study. Clearly, one can see that for densities below $2\cdot10^{12}\,cm^{-2}$, the quantum term is more than 50% of the total energy, and drops slowly to $\sim 30\%$ as the density increases to $10^{13}\,cm^{-2}$.

In this study [1] we prepare several types of devices, all compose of single layer graphene (SLG) on top of highly p-doped silicon substrates with a thin oxide layer, of 2 and $15\,\mathrm{nm}$ HfO_2, $10\,\mathrm{nm}$ SiO_2, and 50 and $100\,\mathrm{nm}$ Si_3N_4. Typical transfer characteristics are presented in Fig. 1b. Many of these devices have high transconductance ($g_m = \partial I/\partial V_g/W$, where W is the device width) and the highest g_m we measured at room temperature and under ambient conditions was $700\,\mu S/\mu m$ at $V_{DS} = 1$V. After subtracting the contact resistance of the device, one finds $g_m \simeq 11\,mS/\mu m$, which, is the highest transconductance ever measured for SLG to the best of our knowledge. The normalized g_m, which takes into account the geometrical dimensions of the device and the bias voltage at which the measurements were performed, is given by $g_{mN} = Lg_m/V_{DS} = \mu c_g$ [2], and was found to be $2.8/44\,mFV^{-1}s^{-1}$ w/wo contact resistance. From direct measurements of the mobility, using the Hall effect, one obtains $c_g = 1.6 \pm .1\,\mu Fcm^{-2}$, which is in good agreement with the expected theoretical value of $1.7\,\mu Fcm^{-2}$. This gate capacitance is 130 times larger than that of the usual $285\,\mathrm{nm}$ SiO_2 based capacitor, and is the largest capacitance published to date in SLG.

In the limit of homogenous graphene, for example, p-type, the carrier density is measured by the Hall effect,

and $\mu(p)$ is found from Eq. 1. However, as the density decreases, puddles forms, and the densities are unknown. In a recent paper Li *et al.* [3] developed a theoretical model for transport in inhomogeneous graphene. They introduced a single parameter, s, which described the standard deviation of the electrostatic potential fluctuations of the graphene landscape with Gaussian distribution. Consequently, the density of states of both the electrons and holes becomes finite at the Dirac point, and electrical transport may prevail. This model also predicts the temperature dependence of the carrier densities, the chemical potential, and the total conductivity based on the effective-medium theory of conductance in composite mixtures [4]. Thus, incorporating the model of Li *et al.* with generalization of Eq. 1 for the two types of charge carriers, and with our conductivity data, we can self-consistently solve this set of equations and extract the fluctuation parameter, s; the charge carriers densities; the chemical potential; and the inverse compressibility.

The results of this analysis are plotted in Fig. 2a, and b before (inset) and after (main) annealing for the p-type branch. Similar results were found for the n-type branch as well. The red curve in Fig. 2a presents the experimental data and the blue line the expected theoretical result. As evident, the agreement is very good, and at low densities where electron and hole puddles coexist, the chemical potential and the inverse compressibility have been modified due to the electrostatic disorder potential, which may be described well by a Gaussian disorder distribution.

In addition, from our measurements and analysis we extract the temperature dependence of the charge carriers densities, the sheet resistance, $\rho_\square$, and the disorder strength parameter, s. Surprisingly, unlike many theoretical and experimental studies, non-monotonic sheet resistance was observed and was primarily attributed to the temperature dependence of the disorder potential itself. By extracting the T dependence of the electron and hole densities from the overall $\rho_\square(T)$, one can accurately study the temperature dependence of the different scattering mechanisms that affect the resistivity of single layer graphene.

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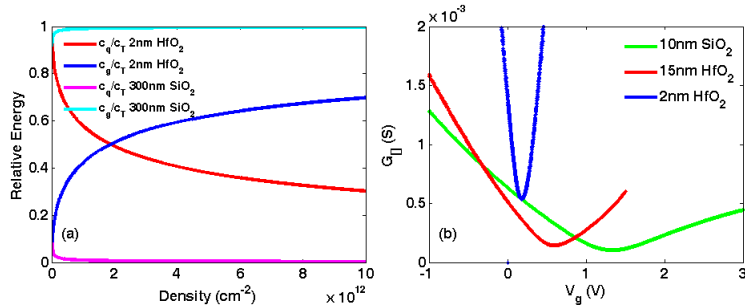[4] S. Kirkpatrick, *Rev. Mod. Phys.*, 45:574–588, 1973.

FIG. 1. (Color online) (a) Relative energy contribution with respect to carrier density according to Eq. 1 calculated at zero temperature and $\phi_0 = 0$. (b) Sheet conductance ($G_\square$) vs gate voltage for three different substrates.
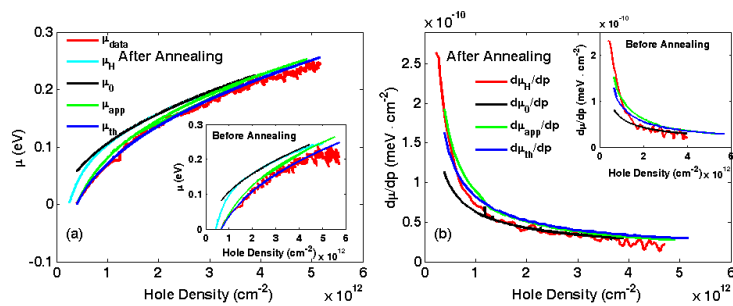

FIG. 2. (Color online) (a) Chemical potential and (b) inverse compressibility vs hole densities before (inset) and after (main) annealing.