

Carrier Transport in Two-Dimensional Graphene Layers

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Carrier transport in gated 2D graphene monolayers is considered in the presence of scattering by random charged impurity centers with density n_i . Excellent quantitative agreement is obtained (for carrier density $n > 10^{12} \text{ cm}^{-2}$) with existing experimental data. The conductivity scales linearly with n/n_i in the theory. We explain the experimentally observed asymmetry between electron and hole conductivities, and the high-density saturation of conductivity for the highest mobility samples. We argue that the experimentally observed saturation of conductivity at low density arises from the charged impurity induced inhomogeneity in the graphene carrier density which becomes severe for $n \lesssim n_i \sim 10^{12} \text{ cm}^{-2}$.

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Recent experimental observation of the density dependent 2D carrier transport in single monolayers of graphene is an important, perhaps seminal, development in low dimensional electronic phenomena in nanostructures [1–5]. First, the gated 2D graphene systems could potentially become multifunctional high speed and high-power transistors, thus introducing a prospective paradigm shift in the micro- (and nano-) electronics of the future. Second, carriers (both electrons and holes) have intriguing (and conceptually novel) linear “Dirac-like” bare kinetic energy dispersion spectra in 2D graphene monolayers due to the interesting honeycomb lattice structure of Carbon atoms. Graphene is, in fact, a carbon nanotube rolled out into a 2D sheet, and the band structure induced carrier spectrum in graphene monolayers is thus linear and chiral with a four-fold ground state degeneracy arising from spin and valley, as well as a Berry phase term arising from the inherent sublattice symmetry which also restricts the carriers from back scattering. This theoretical absence of back scattering has led to the speculation that, as a matter of principle, carrier mobilities in 2D graphene monolayers could be extremely high even at room temperature, and indeed, mobilities as high as $15\,000 \text{ cm}^2/\text{V s}$ have been reported from 1 K to room temperature. These mobility values are comparable to (much higher than) the mobility in the best available Si MOSFET samples at low (high) temperatures. It is, therefore, of considerable fundamental and technological interest to investigate the theoretical limit on the 2D graphene carrier mobility.

In this Letter, we present a theory for 2D graphene carrier transport taking into account scattering by random charged impurity centers, which is the most likely scattering mechanism limiting graphene conductivity. We also calculate the effect of short-range scattering (e.g., by lattice defects), which may be present in some graphene samples. Quantitative agreement between our theory and existing graphene experimental transport data is a strong indication that the dominant carrier scattering mechanism operational in 2D graphene monolayers is Coulomb scattering by random charged impurities located near the interface be-

tween the graphene layer and the substrate. We estimate the typical random charged impurity (effective) concentration to be approximately 10^{12} cm^{-2} in currently available graphene samples, and suggest that reducing this impurity concentration to the 10^{10} cm^{-2} density range—a difficult but not an impossible materials fabrications task—should increase 2D graphene mobility to the extremely large value of $\sim 2 \times 10^6 \text{ cm}^2/\text{V s}$ even at high temperatures. We argue that the experimentally observed “low-density” ($\lesssim 10^{12} \text{ cm}^{-2}$) saturation of 2D graphene conductivity (i.e., the observed “minimum conductivity” at nominally zero gate voltage) also arises from the presence of these charged impurities which lead invariably to large-scale density inhomogeneities in the 2D carrier system with the 2D density fluctuations being larger than the average density at low carrier density where the system therefore breaks into a random network of 2D electrons and hole conducting puddles, producing a finite conductivity at zero gate voltage. Around zero gate voltage, the 2D graphene layer is thus not a simple (homogeneous) zero-gap semiconductor (as intrinsic graphene is expected to be), but a spatially inhomogeneous semimetal with small random puddles of electron and hole 2D liquids depending on the details of the charged impurity configuration in the sample.

For our purpose, intrinsic graphene is essentially a zero-gap semiconductor (with the Fermi level E_F precisely at $E = 0$), with linear chiral carrier kinetic energy dispersion given by $E = \hbar\gamma k$, where k is the 2D carrier wave vector and γ is the constant (i.e., independent of carrier density) Fermi velocity. The intrinsic situation (i.e., with zero gate voltage) has no free carriers at $T = 0$ as a matter of principle (but, as a matter of practice the system breaks up into spatially inhomogeneous conducting puddles of 2D electron and hole droplets due to the potential fluctuations induced by the extrinsic random charged impurity centers which are invariably present in any real 2D graphene sample). The application of an external gate voltage (positive or negative) leads to free carriers (electrons or holes) in the system. We start by assuming the system to be a homogeneous 2D carrier system of electrons (or holes)

with a carrier density n induced by the external gate voltage V_g with $n = \kappa_s V_g / (4\pi t)$, where κ_s is the dielectric constant of the substrate and t is the substrate thickness (i.e., the distance of the gate from the graphene layer). The chemical potential (or, equivalently, the Fermi energy E_F) at $T = 0$ is given by $E_F = \hbar\gamma k_F$, where the 2D Fermi wave vector k_F depends on the carrier density through $k_F = (4\pi n / g_s g_v)^{1/2}$ with g_s (g_v) being the spin (valley) degeneracy of graphene. (We choose $g_s = g_v = 2$ throughout this Letter, although it is possible in some special situations for these degeneracies to be lifted.)

For large carrier densities, where the system is homogeneous, we expect the full Boltzmann transport theory developed below to be exact, hence providing an accurate determination of the charge impurity density n_i by comparing our theory with experimental conductivity data, since n_i just sets the overall scale of the conductivity in the theory. This in turn provides an estimate for the breakdown of our transport theory (typically around $n \lesssim n_i$) and the onset of the inhomogeneous puddles of electron and hole carriers. The experiments of Ref. [2] show that the characteristic density for this breakdown (i.e., the onset of inhomogeneity in our opinion) is $n_c \sim n_i \approx 10^{12} \text{ cm}^{-2}$. An independent estimate (giving the same order of magnitude) can be made by considering the typical local density fluctuations induced by a single charge located inside the substrate at a distance d from the interface. This density fluctuation becomes global in the presence of multiple impurities which can be seen by considering multiple charged impurities distributed in a random Poisson manner in a 2D plane at a distance d above the graphene monolayer. One finds that the density fluctuation δn around the gate-induced average 2D carrier density n is given by $\delta n^2 = n_i / (8\pi d^2)$ confirming that transport close to the Dirac point is dominated by a spatially inhomogeneous carrier density. The transport through such 2D puddles is essentially a one dimensional random network for each of the four spin/valley channels providing a conductivity $\sigma \sim e^2/h$, although a more complete theory is necessary to provide any quantitative comparison with experiments for $n \lesssim n_c \sim n_i$.

We now proceed to describe in detail the microscopic transport properties at high carrier density using the Boltzmann transport theory [6]. We calculate the mobility in the presence of randomly distributed Coulomb impurity charges near the surface with the electron-impurity interaction being screened by the 2D electron gas in the random phase approximation (RPA). The screened Coulomb scattering is the only important scattering mechanism in our calculation. There are additional interface-scattering mechanisms unrelated to the Coulomb centers (e.g., surface roughness scattering), but such interface scattering is believed to be less important in the case of graphene (see below).

We also neglect all phonon scattering effects mainly because both the reduced phase space imposed by chirality

and the low temperatures cause phonon scattering to be negligible in the regime of current interest. Given that 2D graphene is essentially a weakly interacting system with effective $r_s \sim 0.75$, a constant independent of carrier density (r_s here is just the effective fine structure constant), we expect our Boltzmann theory to be a quantitatively and qualitatively accurate description of graphene transport for all practical purposes.

In Boltzmann theory, the conductivity for graphene is given by $\sigma = (e^2/h)(2E_F \langle \tau \rangle / \hbar)$ which comes from the massless chiral Dirac spectrum, where $\langle \tau \rangle$, the energy averaged finite temperature scattering time, is given by $\langle \tau \rangle = \int d\epsilon_k \epsilon_k \tau(\epsilon_k) (-\frac{\partial f}{\partial \epsilon_k}) / \int d\epsilon_k \epsilon_k (-\frac{\partial f}{\partial \epsilon_k})$, where $f(\epsilon_k)$ is the Fermi distribution function. By considering sublattice symmetry of graphene, the energy dependent scattering time $\tau(\epsilon_k)$ for our model of randomly distributed impurity charge centers is given in the leading-order theory by

$$\frac{1}{\tau(\epsilon_k)} = \frac{\pi}{\hbar} \sum_{a, \mathbf{k}'} n_i^{(a)} \left| \frac{v^{(a)}(q)}{\epsilon(q)} \right|^2 (1 - \cos^2 \theta) \delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'}), \quad (1)$$

where $n_i^{(a)}$ is the concentration of the a -th kind of impurity center, $q = |\mathbf{k} - \mathbf{k}'|$, $\theta \equiv \theta_{\mathbf{k}\mathbf{k}'}$ is the scattering angle between the scattering in and out wave vectors \mathbf{k} and \mathbf{k}' , $\epsilon_{\mathbf{k}} = \hbar\gamma|\mathbf{k}|$, and $v^{(a)}(q, d)$ is the matrix elements of the scattering potential between an electron and an impurity. For Coulomb interaction, we use $v^{(c)}(q, d) = 2\pi e^2 \times \exp(-qd) / (\tilde{\kappa}q)$ where d is the location of the charge

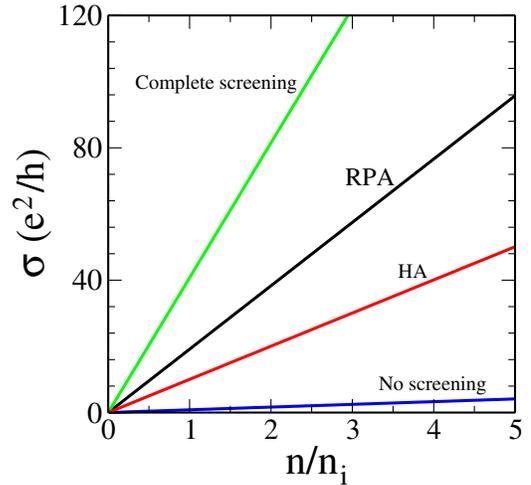


FIG. 1 (color online). Graphene conductivity limited by Coulomb scattering calculated using different approximation schemes with dielectric constant $\kappa = 2.5$. The complete screening approximation (Ref. [8]) does not depend on dielectric constant and overestimates the conductivity, whereas using the unscreened Coulomb interaction would have conductivity less than $4e^2/h$ for the entire range of gate voltages used in the experiment. RPA is the main approximation that we use in this Letter.

impurity in the substrate measured from the interface and for short-range point defect scatterers, $v^{(p)}(q, d) = v_0$, a constant. In Eq. (1), $\varepsilon(q)$ is the 2D finite temperature static RPA dielectric (screening) function appropriate for graphene [7], given by

$$\varepsilon(q) = 1 + \frac{q_s}{q} \begin{cases} 1 - \frac{\pi q}{8k_F} & q \leq 2k_F \\ 1 - \frac{\sqrt{q^2 - 4k_F^2}}{2q} - \frac{q \sin^{-1} 2k_F/q}{4k_F} & q > 2k_F \end{cases} \quad (2)$$

where $q_s = 4e^2 k_F / (\hbar \bar{\kappa} \gamma)$ is the effective graphene 2D Thomas-Fermi wave vector and $\bar{\kappa} = \kappa(1 + \pi r_s/2)$ is the effective dielectric constant. We can immediately observe from Eq. (1) that $\tau^{-1} = \tau_c^{-1} + \tau_s^{-1}$, where the Coulomb scattering time $\tau_c \sim \sqrt{n}$ dominates at low density over the short-range scattering time $\tau_s \sim 1/\sqrt{n}$.

We emphasize that in order to get quantitative agreement with experiment, the full RPA dielectric function needs to be calculated. In Fig. 1, we show comparison between calculated conductivities in the RPA screening used in our calculation with other approximation schemes including the completely screened (“strong screening”) approximation [8] valid only for $r_s \gg 1$, which gives a larger value for the conductivity that is independent of the background lattice dielectric constant κ . We note that for graphene $r_s \sim 0.75$ making the strong screening approximation invalid. We also show results for Hubbard approximation (HA) screening which includes local field corrections approximately. A recent theory [9] has considered the temperature dependence of conductivity within the Thomas-Fermi approximation.

Our main result is the quantitative agreement with experiments in the regime where the conductivity is linear in density. We note, however, that electron-hole asymmetry with a superlinear conductivity is ubiquitous in the experimental data. Changing the sign of bias voltage has no effect on the concentration of charged impurities, but (and especially at high voltages) it changes the average distance

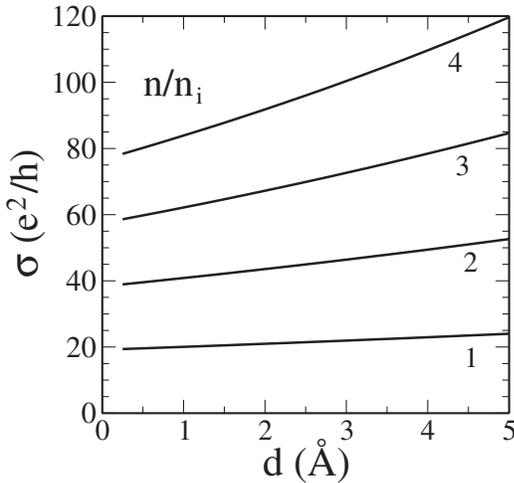


FIG. 2. The effect of remote scatters. Here d is the distance between the 2D graphene layer and the 2D impurity layer.

between 2D graphene sheet and the impurity in the substrate, particularly since the random charged impurities could be both positive and negative in unequal numbers. In our model, we approximate that the impurities are confined in a 2D plane located at a distance d from the interface. We find that a small shift of $d \sim 1-2 \text{ \AA}$ is sufficient to explain the superlinear behavior seen in the experiments. As shown in Fig. 2, we note that for fixed d , the difference between the actual conductivity and the linear value at $d = 0$ is negligible at low density, $n/n_i \sim 1$, but becomes important for $n/n_i \gtrsim 3$.

Another feature of the experiments is that for very high mobility samples, one finds a sublinear conductivity [4]. Such high quality samples presumably have a small charge impurity concentration n_i , and it is therefore likely that point defects here play a more dominant role. Point defects (which would be dominant for either large carrier density n or low charge impurity concentration n_i) give rise to a constant conductivity in contrast to charged impurity scattering which produces a conductivity roughly linear in n/n_i . Any realistic graphene sample is somewhere in the crossover between these two limits. Our formalism (see Eq. (1)) can include both effects, where zero range scatterers have an effective point defect density of n_p .

We estimate that for most samples, $n_p/n_i \ll 1$, while for the highest mobility samples, $n_p/n_i \sim 0.2$. Shown in Fig. 3 is the graphene conductivity calculated including both charge impurity (n_i) and zero range point defect (n_p) scattering. For small n_p/n_i , we find the linear conductivity that is seen in most experiments, while for large n_p/n_i , we see the flattening out of the conductivity curve (which in the literature [3] has been referred to as the sublinear conductivity). We believe this high-density flattening of

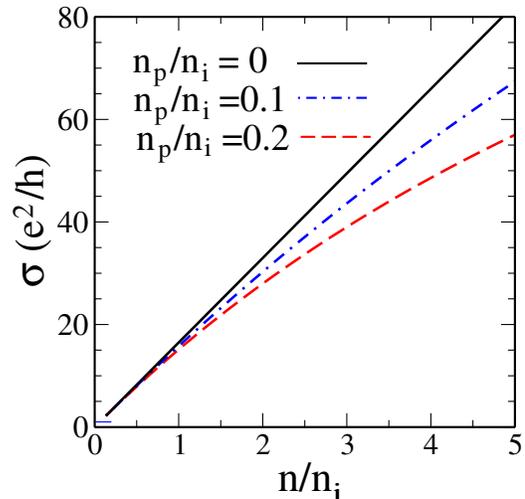


FIG. 3 (color online). Graphene conductivity calculated using a combination of short and long range scatterers. One finds that sublinear conductivity at high density is likely to be seen in samples with a small Coulomb impurity density and high mobility.

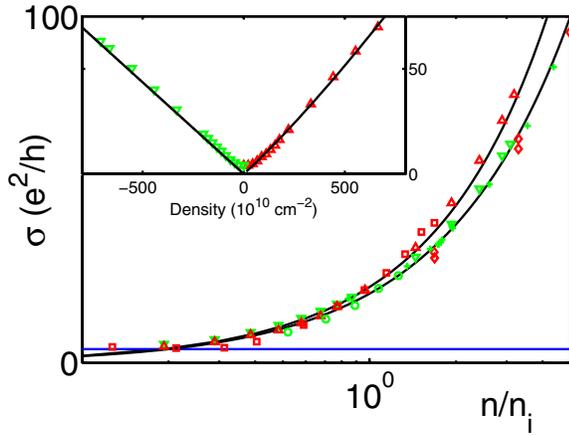


FIG. 4 (color online). Figure shows comparison of theory (Eq. (1)) with experimental data of Ref. [2] (up and down triangles, $n_i = 2.3 \times 10^{12} \text{ cm}^{-2}$), Ref. [5] (circles and squares, $n_i = 3.4 \times 10^{12} \text{ cm}^{-2}$), and Ref. [4] (diamonds and crosses, $n_i = 0.43 \times 10^{12} \text{ cm}^{-2}$) for electrons and holes, respectively. Solid lines (from bottom to top) show the minimum conductivity of $4e^2/h$, theory for $d = 0$, and $d = 2 \text{ \AA}$. The inset shows Ref. [2] data on a linear scale assuming that the impurity shifts by $d = 2 \text{ \AA}$ for positive voltage bias.

the graphene conductivity [4] is a nonuniversal crossover behavior arising from the competition between two kinds of scatterers, n_p and n_i .

Finally, in Fig. 4, we show the comparison of our theory (with only charged impurity scattering) to recent experiments [2,4,5]. We determine n_i (which simply sets the overall scale) by fitting the data at high density to the result of Eq. (1) and observe very good agreement between all three experimental data sets and our transport theory. We note that the order of magnitude for $n_i \sim 10^{12} \text{ cm}^{-2}$ agrees with independent estimates (e.g., the low-density $n \lesssim n_i$ conductivity saturation around zero gate voltage). The one-parameter (i.e., n_i) quantitative agreement between our theory and three independent sets of experimental data is one of our most important findings.

We emphasize that our Drude-Boltzmann transport theory is valid only in the high-density, $n > n_i$, regime, and therefore only the agreement between theory and experiment in the high-density regime is meaningful. Indeed, our theory predicts that conductivity should vanish linearly as carrier density goes to zero, whereas in reality, the graphene conductivity at low density (i.e., gate voltage), $n < n_i$ in our model, becomes approximately a density independent constant of the order e^2/h . Our theory provides no quantitative or qualitative explanation for this low-density behavior, where we believe that the physics of strong density inhomogeneity (random puddles of 2D electron and holes) induced by the charged impurities in the insulating dielectric dominates, and our Drude-Boltzmann

physics simply does not apply. We cannot, however, rule out that the low-density regime is dominated by localization physics [10], or the Dirac cone physics of intrinsic graphene [11] since our theory does not apply in the $n < n_i$ low-density regime. We note, however, that recent numerical work [8,12] on quantum transport in the low-density regime is consistent with the picture of a percolation transition between electron and hole carriers with the implication that the zero-carrier density intrinsic regime may simply be experimentally inaccessible. However, what we have convincingly established in this Letter is that the high-density, $n > n_i$, 2D graphene transport is dominated by impurity scattering which can be theoretically described by a microscopic Drude-Boltzmann model.

In conclusion, we have developed a detailed microscopic transport theory for 2D graphene layers. We have shown that charged impurities in the substrate are the dominant source of scattering. We have argued that the low-density regime is dominated by density fluctuations caused by these charge impurities and that a picture of inhomogeneous puddles of conducting electrons and conducting holes is necessary to understand the finite value of conductivity near zero bias. Finally, the Fermi temperature of 2D graphene being around 1300 K for $n \sim 10^{12} \text{ cm}^{-2}$, there is essentially no temperature dependence in 2D graphene conductivity in the 0–300 K temperature range arising from charged impurity scattering, a fact that is in excellent agreement with experimental observation [13].

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