Numbers of donors and acceptors from transport measurements in graphene

D. S. Novikov
W. I. Fine Institute of Theoretical Physics, University of Minnesota, Minneapolis, MN 55455, USA
(Dated: February 5, 2008)

A method is suggested to separately determine the surface density of positively and negatively charged impurities that limit the mobility in a graphene monolayer. The method is based on the exact result for the transport cross-section, according to which the massless carriers are scattered more strongly when they are attracted to a charged impurity than when they are repelled from it.

PACS numbers: 81.05.Uw 72.10.-d 73.63.-b 73.40.-c

The discovery of graphene opened up a number of research directions. The outstanding properties of this material originate from its massless electron-hole symmetric carrier dispersion \( \epsilon(p) = \pm vp \), where the Fermi velocity \( v \approx 10^6 \text{ m/s} \). Recent transport measurements show that the graphene mobility is approximately independent of the transport directions.

Indeed, such an asymmetry is absent in the exact result for the transport cross-section, according to which the massless carriers are scattered more strongly when they are attracted to a charged impurity than when they are repelled from it.

\[ U(r) = -\frac{Ze^2}{r} = -\hbar v \times \frac{\alpha_0}{r} \quad (1) \]

where the dimensionless impurity strength
\[ \alpha_0 = \frac{Ze^2}{\hbar v} \quad (2) \]

is proportional to the energy-dependent carrier wavelength \( \lambda_c = 2\pi \hbar v/|\epsilon| \). The dimensionless function \( C(\alpha_e) \), which is the transport cross-section in the units of the carrier wavelength, is plotted in Fig. 1.

The transport cross-section is strongly asymmetric with respect to sign \( \alpha \times \text{sign} \epsilon \), i.e. the sign of the potential as seen by the carrier: A donor (\( \alpha > 0 \)) scatters conduction electrons (\( \epsilon > 0 \)) more effectively than it scatters holes (\( \epsilon < 0 \)). The attraction-repulsion asymmetry is commonplace [e.g. Problem 6 to Sec. 132 in Ref. 1].

Physically, one may expect the particle to spend more time around an attractive potential center and thereby
be more significantly deflected (this intuition, strictly speaking, applies to the massive particles). However, for the practically important 2D and 3D Coulomb scattering in a parabolic band, the corresponding exact solutions are somewhat exceptional in a sense that they lack such an asymmetry. Remarkably, for the “relativistic” carrier dispersion, characteristic of graphene, this generally expected asymmetry is recovered.

The result (5) applies to the half-filled \( \pi \)-electron band. In this case the RPA screening (3) is scale-invariant, preserving the functional form of the potential (1). At finite carrier density \( k_F^2/\pi \), corresponding to the Fermi momentum \( k_F \), the RPA screening is not scale-invariant anymore (6,10). This means that the screened potential has the form (1) only at distances shorter than the screening length \( l_s \simeq Z/\alpha k_F \), and is cut-off at larger distances. Scattering in such an effective potential is not tractable exactly, and the momentum relaxation rate is practically calculated only in the Born approximation (6,9,10,11). The latter becomes asymptotically exact for small \( |\alpha| \ll 1 \) at large distances \( r \sim l_s \gg 1/k_F \), but misses non-perturbative effects at distances shorter than \( l_s \sim 1/k_F \) when the potential is large, \( \alpha \sim 1 \).

To investigate the role of the asymmetry, below I neglect the additional screening at finite \( k_F \), and use the scale-invariant screening (6) by the filled valence band only. Such an approach may be justified noting that, while the divergent Coulomb phase shifts \( \delta_j \) in \( 2k_F \) (same for all the angular momentum channels) comes from the distances \( k_F r \gg 1 \), the relative scattering phases \( \delta_j \), that contribute to the momentum relaxation via the transport cross-section, accumulate on the scale \( l_j = j/k_F \). This way the lowest phase shifts \( \delta_j \) are practically acquired within the screening length \( l_s \gg l_j \), while the rest are strongly reduced by the momentum relaxation at finite \( k_F \). Fortunately, the dominant contribution to the cross-section \( \Sigma \) comes from the channels with \( j \sim 1 \) (Fig. 1 inset), that are least damaged by the additional screening at finite \( k_F \). In particular, the lowest shift \( \delta_{1/2} \) is behaving qualitatively differently from the rest, causing the pronounced attraction-repulsion asymmetry. Such an anomalous behavior is to be expected, as this is the channel in which the criticality, \( |\alpha| = j \), is first reached.

To calculate the conductivity, consider for simplicity the case when all the donors have the same valence and are characterized by the strength \( +\alpha \), while all the acceptors have the opposite valence and the strength \( -\alpha \); hereon \( \alpha > 0 \). Let \( f_{\pm}(p) \) be the distribution functions for the particles and holes correspondingly. With the electron interaction effects approximated via the screening (3), the kinetic equations for each carrier type decouple,

\[
e\hat{\epsilon} \cdot \frac{\partial f_{\pm}}{\partial p} = -\frac{\delta f_{\pm}}{\tau_{\pm}(E_p)}, \quad e\hat{\epsilon} \cdot \frac{\partial f_{\mp}}{\partial p} = -\frac{\delta f_{\mp}}{\tau_{\mp}(E_p)},
\]

where \( e < 0 \) is the electron charge, and

\[
\tau_{\pm}^{-1} = v \left[ n_+^+ \Lambda_+^{\pm} + n_-^- \Lambda_{-\tau} \right], \quad \tau_{\mp}^{-1} = v \left[ n_+^+ \Lambda_+^{\mp} + n_-^- \Lambda_{-\tau} \right].
\]

Here \( \Lambda_+^\pm = \lambda \times C(\pm \alpha) \) are the right and left parts of the cross-section in Fig. 1. The rates (7) have straightforward meaning: The conduction electrons scatter off the donors with the enhanced cross-section \( \Lambda_+^{\pm} \) and off the acceptors with the reduced cross-section \( \Lambda_{-\tau} \), while for the holes the situation is reversed. The Coulomb transport times (7) are proportional to the quasiparticle energy,

\[
\tau_{\pm}(E) = \frac{|\epsilon|}{2\pi \hbar v^2} \times \frac{1}{n_+^+ C(+\alpha) + n_-^- C(-\alpha)}, \quad \tau_{\mp}(E) = \frac{|\epsilon|}{2\pi \hbar v^2} \times \frac{1}{n_+^+ C(+\alpha) + n_-^- C(-\alpha)}.
\]

From the kinetic equations (6) find the deviations

\[
\delta f_{\pm} = \pm e(\hat{\epsilon} \hat{p}) v \tau_{\pm}(E) \left[ -\partial_{\epsilon} f_{\pm}(0) \right], \quad \hat{p} \equiv p/\hbar
\]

From the distribution functions from the equilibrium Fermi distribution \( f_{\pm}^{eq}(\epsilon) = 1/[e^{(\epsilon + \mu)/T} + 1] \), where \( \mu \) is chemical potential relative to the half-filled \( \pi \) band. The resulting electric current [here \( N_f = 2_{\text{spin}} \times 2_{\text{valley}} = 4 \) independent polarizations]

\[
eJ = e N_f v \int \frac{d^2 p}{(2\pi \hbar)^2} \hat{p} \left( \delta f_+ - \delta f_- \right) = \sigma \hat{\epsilon}
\]

corresponds to the dc conductivity

\[
\sigma = \frac{N_f e^2}{h} \int_0^\infty \epsilon \left[ \frac{\tau_+(E)}{2\hbar} \left( -\partial_{\epsilon} f_+^{eq}(0) \right) + \frac{\tau_-(-E)}{2\hbar} \left( -\partial_{\epsilon} f_-^{eq}(0) \right) \right].
\]
Last, I express the conductivity \((\sigma)\) via the carrier number densities. Using integration by parts, the electron and hole densities \(n^\pm\) can be cast in a similar form,

\[
n^\pm(\mu) = \int \frac{N_f d^3 p}{(2\pi)^3} f_0^\pm(\epsilon_p) = \frac{N_f}{4\pi^2 v^2} \int_0^\infty e^2 d\epsilon \left[ -\partial_\epsilon f_0^\pm \right].
\]

Combining Eqs. (5), (10), and (11), the conductivity

\[
\sigma = \frac{(e^2/h)n^+}{n^+_i C(\alpha)} + \frac{(e^2/h)n^-}{n^-_i C(-\alpha)}.
\]

The asymmetry of the cross-section \(C(\alpha)\) translates into the asymmetry in the dependence of the conductivity \((\sigma)\) on the net carrier density \(n = n^+ - n^-\). Introducing the charge carrier imbalance \(\delta n = n^+ - n^-\), as well as the symmetric and antisymmetric parts of the cross-section

\[
C_{s,a}(\alpha) = \frac{1}{2} C(\alpha) \pm C(-\alpha),
\]

one can represent the conductivity \((\sigma)\) in the form

\[
\sigma = \frac{c}{h n_i C_s} n \times \frac{1 - (\delta n/n)c}{1 - c^2},
\]

where

\[
n_i = n^+_i + n^-_i.
\]

Here \(n_i = n^+_i + n^-_i\) is the total surface density of charged impurities. Thus measuring the conductivity asymmetry for, say, opposite gate voltages relative to the charge neutrality point, such that \(\delta n \approx \pm n\), one can use fitting to Eq. (14) in order to determine the concentrations \(n^+_i\) of donors and acceptors separately, since the ratio \(c = (\delta n = -n - \sigma_{\delta n = n})/(\delta n = n + n = n = n)\). Including the effect of the corrugations (ripples) simply shifts the symmetric part \(n_i C_s \rightarrow n_i C_s + C_r\) in Eq. (14), where \(\sigma_r = (e^2/h)n/C_r\) is the conductivity due to the ripples only. This still allows one to determine the donor-acceptor imbalance \(n^+_i - n^-_i\). To find \(n^+_i + n^-_i\) one needs to independently gauge the ripple parameter \(C_r\) by varying the number of the charged impurities. As expected, the Born approximation \(C_r \rightarrow C^{\text{Born}} = \pi a^2/2\) has no asymmetry \((C_r \rightarrow 0)\), thus it entirely misses the effect.

Fig. [1] shows the odd and even components \([13]\) of the exact cross-section, together with their polynomial fits

\[
a_c \approx 1.68 \alpha^3 + 7.69 \alpha^5 - 21.4 \alpha^7,
\]

\[
c_s \approx 1.54 \alpha^2 + 3.33 \alpha^4,
\]

\[
C_a/C_s \approx 1.37 \alpha - 0.6 \alpha^3.
\]

For \(|Z| = 1\) impurities in SiO\(_2\) \((\alpha \approx 0.37)\) the asymmetry \(C_a/C_s \approx 0.46\) should be fairly pronounced. Recent dc measurements of graphene monolayers on SiO\(_2\) substrate\([6]\) show a noticeable asymmetry in the conductivity \(\sigma(n)\). The proposed explanation is that the acceptors prevail over donors. This provides an alternative to the suggestion\([12]\) that the asymmetry is caused by the gate-induced displacements of impurities. Further experiments with controlled numbers of adsorbed donors and acceptors may distinguish between the two scenarios.

To conclude, a novel method for characterizing the electrostatic environment of a graphene sample is proposed. The method is based on the attraction-repulsion asymmetry of the exact transport cross-section for scattering off the charged impurities, and could potentially allow to separate the scattering effects of the donors and of the acceptors from those of the microscopic ripples.

I thank L. Glazman, B. Shklovskii and A. Shytov for helpful discussions. This work was supported by DOE Grant DE-FG02-06ER46310.

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18. As \(l_i\) is the impact parameter for the particle with the angular momentum \(j = k_F l_i\), the asymptotic form of the scattering states\([12]\) (defining \(\delta l_i\)) becomes valid for \(r \gg l_i\).
19. The sum in Eq. (15) converges as \(\alpha^2 \sum j (j^2) \) for \(j \gg 1\), since \(\delta j \sim \alpha j\) according to the Stirling’s formula.
20. This value may decrease somewhat when the additional screening by the free carriers is taken into account.