Neutrality Point of Graphene with Coplanar Charged Impurities

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The ground state and the transport properties of graphene subject to the potential of in-plane charged impurities are studied. The screening of the impurity potential is shown to be nonlinear, producing a fractal structure of electron and hole puddles. Statistical properties of this density distribution as well as the charge compressibility of the system are calculated in the leading-log approximation. The conductivity depends logarithmically on α, the dimensionless strength of the Coulomb interaction. The theory is asymptotically exact when α is small, which is the case for graphene on a substrate with a high dielectric constant.

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A number of recent experimental [1–5] and theoretical [6–16] investigations have studied the effect of *charged* impurities on the properties of graphene [17] at its neutrality point (NP). It has been shown that in response to the random potential of impurities electron density becomes very inhomogeneous. A deeper physical understanding of these inhomogeneities is important for clarifying the true nature of disorder in this new material. Hence, an analytic approach to this problem is desirable. Below I present my results in this direction.

As a model, I assume that impurities with random charge $\pm e$ and total concentration n_i are distributed randomly on a graphene sheet of area A. The system resides in a medium of dielectric constant κ. The strength of the Coulomb interaction, $U(r) = e^2/\kappa r$, is characterized by the parameter $\alpha = e^2/\kappa \hbar v$ where v is the Fermi velocity.

My main results are as follows. For $\kappa \gg 1$ where the interaction is weak, $\alpha \ll 1$, the basic electronic properties of graphene at the NP can be computed to the leading order in $1/\mathcal{L}$, where $\mathcal{L} \gg 1$ is the solution of the equation

$$\mathcal{L} = \ln(1/(4\alpha\mathcal{L})). \tag{1}$$

I find that the charge compressibility χ_0 at the NP (measured in Ref. [5]) is given by

$$\chi_0 = \kappa/(2\pi e^2 R), \qquad R = \ell/(4\alpha\mathcal{L}), \tag{2}$$

where R is the screening length and $\ell = (2\alpha\sqrt{n_i})^{-1} \ll R$ has the meaning of a typical quantum uncertainty in the quasiparticle positions. Observables, such as the correlator $S(r) \equiv \langle n(\mathbf{r})n(0)\rangle$ of the local density become nontrivial on scales $r > \ell$. For such r, I find

$$S(r) = \frac{\mathcal{L}^2}{2\pi\ell^4}[3\vartheta\sqrt{1-\vartheta^2} + (1+2\vartheta^2)\arcsin\vartheta], \tag{3}$$

$$\vartheta(r) = \frac{K(r)}{K(\ell)}, \qquad K(\ell) = \frac{\pi}{2}\left(\frac{\hbar v}{\ell}\right)^2 \mathcal{L}. \tag{4}$$

Here $K(r) \equiv \langle\Phi(0)\Phi(\mathbf{r})\rangle$ is the correlator of the screened random potential Φ. I show that $K(r)$ behaves as

$$K(r) = \frac{\pi}{2}\left(\frac{\hbar v}{\ell}\right)^2 \times \begin{cases} \ln(R/r), & \ell \ll r \ll R, \\ 2(R/r)^3, & R \ll r. \end{cases} \tag{5}$$

Equation (5) agrees with the results of Ref. [13] in the limit $\alpha \ll 1$. Equation (3) is in a qualitative agreement with the numerical simulations [15] for $\alpha \sim 1$ (where my theory is at the border of validity).

Of special interest for transport and imaging experiments are the contours $n(\mathbf{r}) = 0$, which separate electron [$n(\mathbf{r}) > 0$] and hole [$n(\mathbf{r}) < 0$] regions. Only contours larger than some minimum size Λ^{-1} are important for the key observables (in imaging, Λ^{-1} is the spatial resolution; in transport, $\Lambda^{-1} \sim \ell\sqrt{\mathcal{L}}$, see below). To isolate them, we can separate the total density and the total potential into smoothly and rapidly varying parts,

$$n(\mathbf{r}) = \bar{n}(\mathbf{r}) + \delta n(\mathbf{r}), \qquad \Phi(\mathbf{r}) = \bar{\Phi}(\mathbf{r}) + \delta\Phi(\mathbf{r}), \tag{6}$$

where $\bar{n}(\mathbf{r})$ and $\bar{\Phi}(\mathbf{r})$ contain only Fourier harmonics with $k < \Lambda$. Below I assume that $1/\ell \lesssim \Lambda \ll 1/R$. I refer to the contours $\bar{n}(\mathbf{r}) = 0$ as the *p-n* junctions (PNJ). I show that most of the PNJ are closed loops of diameter $d \sim \Lambda^{-1}$. They reside inside of successively larger loops, forming a self-similar set. The fractal dimension D_h of the PNJ contours is equal to $3/2$ at $d < R$ but becomes $7/4$ at $d > R$. Finally, the conductivity at the NP is

$$\sigma_{\mathrm{NP}} = (e^2/h)c\mathcal{L}, \qquad c = 0.50 \pm 0.05. \tag{7}$$

This result is supposed to be valid at temperatures high enough that weak localization effects [18] can be neglected but low enough that elastic scattering by impurities is still the dominant current relaxation mechanism.

A similar expression for σ_{NP} can be deduced from Ref. [13]. It has $c = 4/\pi$ for $\alpha \ll 1$, which is larger than my value of c by about a factor of 2. Numerical estimates of σ_{NP} at $\alpha \sim 1$ were reported in Ref. [16].

Let us turn to the derivation. Let $V(\mathbf{r})$ be the bare random potential due to impurities. In the k space it is given by $V_\mathbf{k} = U_\mathbf{k}N_\mathbf{k}$, where $N_\mathbf{k}$ and $U_\mathbf{k} = 2\pi\alpha\hbar v/k$ are the Fourier transforms of the impurity density and the Coulomb potential, respectively. At $0 < k \ll n_i^{1/2}$ we can

treat them as zero-mean Gaussian random variables with variances $\langle N_{\mathbf{k}} N_{\mathbf{k}'} \rangle = A n_i \delta_{\mathbf{k}, -\mathbf{k}'}$ and $\langle V_{\mathbf{k}} V_{\mathbf{k}'} \rangle = U_{\mathbf{k}}^2 \langle N_{\mathbf{k}} N_{\mathbf{k}'} \rangle$. It is easy to see then that the coarse-grained random potential $\bar{V}(\mathbf{r})$ is infrared divergent [19]:

$$\langle \bar{V}^2(\mathbf{r}) \rangle = \sum_{k < \Lambda} \langle V_{\mathbf{k}} V_{\mathbf{k}'} \rangle \simeq (\pi/2)(\hbar v/\ell)^2 \ln(\Lambda \sqrt{A}). \quad (8)$$

This divergence is of course cured once the bare potential is replaced by a screened one,

$$\Phi_{\mathbf{k}} = V_{\mathbf{k}} + n_{\mathbf{k}} U_{\mathbf{k}} = V_{\mathbf{k}} + (2\pi \alpha \hbar v/k) n_{\mathbf{k}}, \quad (9)$$

which is expected to have the following properties:

$$\Phi_{\mathbf{k}} = V_{\mathbf{k}}, \quad kR \gg 1; \quad \Phi_{\mathbf{k}} = V_{\mathbf{k}} \frac{kR}{1 + kR}, \quad kR \ll 1. \quad (10)$$

The last equation follows from the definition of the thermodynamic charge compressibility $\chi_0 = -n_{\mathbf{k}}/\Phi_{\mathbf{k}}$ and the relation between χ_0 and R, Eq. (2). I wish to show that R is given by the second formula in Eq. (2), in particular, that it is a function of the impurity concentration. This means that the screening is nonlinear; i.e., the response of electrons in graphene to long-wavelength Fourier harmonics $k < R^{-1}$ is nonperturbative in $V_{\mathbf{k}}$.

Formally, $n(\mathbf{r})$ and $\Phi(\mathbf{r})$ can be sought by minimizing the total energy $E[n] + \mathcal{E}[n]$, where $E[n] = A^{-1} \sum_{\mathbf{k}} V_{-\mathbf{k}} n_{\mathbf{k}} + (1/2) U_{\mathbf{k}} |n_{\mathbf{k}}|^2$ is the electrostatic energy and $\mathcal{E}[n]$ is the sum of kinetic, exchange, and correlation energies [20]. Since the functional $\mathcal{E}[n]$ is nonlocal, nonlinear, and strictly speaking, unknown, further steps require approximations. We are aided by the parameter $\mathcal{L}$; the condition $\mathcal{L} \gg 1$ ensures separation of two length scales: (i) the typical quantum uncertainly ℓ over which the nonlocality of $\mathcal{E}[n]$ is important and (ii) the screening length $R \gg \ell$ at which the nonlinear screening sets in. As a result, a *local* density approximation $\mathcal{E} = \int d\mathbf{r} \varepsilon(\bar{n}(\mathbf{r}))$ is valid and the total energy is minimized when

$$\bar{\Phi}(\mathbf{r}) = -\varepsilon'(\bar{n}(\mathbf{r})). \quad (11)$$

The kernel $\varepsilon(n)$ can be computed by integrating out the rapid density fluctuations $\delta n(\mathbf{r})$ within the linear-response theory [6] for $\alpha \ll 1$. The result is [21]

$$\varepsilon = \frac{2\sqrt{\pi}}{3} \hbar v |\bar{n}|^{3/2} + \delta \varepsilon, \quad \delta \varepsilon = -\frac{3\varepsilon}{4|\bar{n}|\ell^2} \ln\left(\frac{k_F}{\Lambda}\right). \quad (12)$$

Here $k_F(\mathbf{r}) = \sqrt{\pi|\bar{n}(\mathbf{r})|}$ is the local Fermi momentum. Equation (11) can now be easily inverted to give

$$\bar{n}(\bar{\Phi}) \simeq -\frac{\bar{\Phi}|\bar{\Phi}|}{\pi(\hbar v)^2} - \frac{\mathrm{sgn}(\bar{\Phi})}{2\ell^2} \ln\frac{|\bar{\Phi}|}{\hbar v \Lambda}, \quad |\bar{\Phi}| \gg \hbar v \Lambda. \quad (13)$$

Later, we will also need the derivative of function $\bar{n}(\bar{\Phi})$:

$$\chi(\bar{\Phi}) \equiv -\frac{d\bar{n}}{d\bar{\Phi}} \simeq \frac{2}{\pi} \frac{|\bar{\Phi}|}{(\hbar v)^2} + \frac{1}{2\ell^2 |\bar{\Phi}|}, \quad |\bar{\Phi}| \gg \hbar v \Lambda. \quad (14)$$

We have reduced the original *quantum* problem to the nonlinear integral equation (13) for the *classical* quantity $\bar{n}(\mathbf{r})$ [Note that $\bar{\Phi}[\bar{n}]$ is given by Eq. (9).] I call this a renormalized Thomas-Fermi approximation (RTFA) [22].

In conventional gapped semiconductors, this would be about as far as one could go analytically before having to resort to heuristic estimates [23], computer simulations [19], or variational methods [24]. The reason is as follows: the energy minimization is constrained by $\bar{n}(\mathbf{r}) \geq 0$, leading to a finite area fraction of depletion regions $\bar{n}(\mathbf{r}) = 0$. Therein Eq. (11) is replaced by $\bar{\Phi}(\mathbf{r}) \geq 0$ [23], so that no analog of Eq. (13) exists, making the problem analytically intractable. In graphene, there is no constraint on the sign of density. Instead, the conditions for validity of Eq. (12) are $\delta \varepsilon \ll \varepsilon$ and $\Lambda \ll k_F$, which are typically met everywhere except small area regions near the PNJ (see below).

Yet another serendipity is that the divergence of the bare potential is only logarithmic, cf. Eq. (8). The enables us to compute the nonperturbative long-range response with a logarithmic accuracy. Indeed, in view of Eq. (10), $\Phi_{\mathbf{k}}$ and $V_{\mathbf{k}}$ are linearly related at all $\mathbf{k}$, except perhaps $k \sim 1/R$. But this "difficult" range of intermediate k makes an $\mathcal{O}(\mathcal{L}^{-1})$ contribution to the total $\bar{\Phi}(\mathbf{r})$. To this order in $\mathcal{L}$ we can treat $\bar{\Phi}(\mathbf{r})$ as a Gaussian random potential. Its probability distribution function (PDF) is uniquely determined by the correlator $\bar{K}(r) \equiv \langle \bar{\Phi}(0)\bar{\Phi}(\mathbf{r}) \rangle$, to calculate which we *can* use Eq. (10). Since $\bar{K}(r) \simeq K(r)$ for $r\Lambda \gg 1$, this immediately leads to Eq. (5).

Next, Gaussian statistics implies ergodicity. Therefore, the density correlation function can be written as

$$S(r) = \int df df' P_{\Phi\Phi}(r, f, f') \bar{n}(f) \bar{n}(f'), \quad (15)$$

where $P_{\Phi\Phi} \equiv \langle \delta(f - \bar{\Phi}(0)) \delta(f' - \bar{\Phi}(\mathbf{r})) \rangle$ is the two-point PDF of $\bar{\Phi}$ and function $\bar{n}(f)$ is given by Eq. (13). Using the standard expression [25] for $P_{\Phi\Phi}$ in terms of $\bar{K}$ and a bit of algebra [21], I get Eq. (3). At $r \gg R$, it simplifies to $S(r) \simeq \langle\chi\rangle^2 K(r)$, where $\langle\chi\rangle \equiv \int df P_{\Phi}(f) \chi(f)$, which implies that $\chi_0 = \langle\chi\rangle$. Substituting here Eq. (14) and computing the average over the Gaussian field $\bar{\Phi}$ using Eq. (5), I get $\chi_0^2 = 8K(\ell)/(\pi^3 \hbar^4 v^4)$. Finally, since $K(\ell)$ is given by Eq. (4), we recover the desired Eq. (2).

It is also possible to compute the PDF P_n of $\bar{n}$. The Gaussian statistics of $\bar{\Phi}$ combined with Eq. (13) entails

$$P_n(\bar{n}) = \frac{\ell}{2\sqrt{\pi \mathcal{L}|\bar{n}|}} \exp\left(-\frac{|\bar{n}|\ell^2}{\mathcal{L}}\right), \quad |\bar{n}|\ell^2 \gg 1. \quad (16)$$

Note, however, that this equation is invalid at small densities, $|\bar{n}| < 1/\ell^2$, where the RTFA fails. At such $\bar{n}$ the divergence of $P_n(\bar{n})$ basically saturates [21]. These low-density regions are usually found near the PNJ.

Geometrically, the PNJ are the isolines $\bar{\Phi}(\mathbf{r}) = 0$ of a surface with the "height" profile $\bar{\Phi}(\mathbf{r})$. The zero height is the percolation threshold, and so all but one of the PNJ are closed loops. These loops are characterized by a certain fractal dimension D_h. For loops of diameter d in the range $\Lambda^{-1} \ll d \ll R$, in which $\bar{\Phi}(\mathbf{r})$ surface is logarithmically rough [Eq. (5)], we have the exact result [26] $D_h = 3/2$, which means that closed-loop PNJ typically have the perimeter length of

$$p \sim d^{3/2}\ell^{-1/2}, \qquad \Lambda^{-1} \ll d \ll R. \qquad (17)$$

As d increases beyond R, the correlator $K(d)$ rapidly decays [Eq. (5)], and so D_h crosses over [26] to the usual uncorrelated percolation exponent [25] of $7/4$.

Let us now discuss electron transport. Away from the NP where electron density n is large and homogeneous, transport can be studied by means of the usual kinetic equation. The final results for conductivity $\sigma(n)$ and transport mean-free path $l(n)$ are [6,8,12,13]

$$\sigma(n) = \frac{8}{\pi}\frac{e^2}{h}\ell^2|n|, \qquad l(n) = \frac{4}{\pi^{3/2}}\ell^2|n|^{1/2}. \qquad (18)$$

Our goal is to compute the conductivity σ_{NP} at the NP where $n = n(\mathbf{r})$ is inhomogeneous.

The first step is to show that we can define the conductivity $\bar{\sigma}(\mathbf{r})$ and the mean-free path $\bar{l}(\mathbf{r})$ locally. Under the assumed condition $1/R \ll \Lambda \lesssim 1/\ell$ for typical $\bar{n}(\mathbf{r}) \sim \mathcal{L}/\ell^2$ we have $\nabla l(\bar{n}) \sim \Lambda l(\bar{n}) \lesssim 1$. Hence, function $l(\bar{n}(\mathbf{r}))$ is slowly varying and Eq. (18) for the uniform density can be used: $\bar{l} = l(\bar{n})$. In turn, the local conductivity is given by the Einstein relation $\bar{\sigma}(\mathbf{r}) = e^2\chi(\mathbf{r})v\bar{l}(\mathbf{r})/2$. To be careful one should check this zeroth order result by calculating higher order corrections due to spatial fluctuations of the collision term in the kinetic equation. Skipping the details [21], I just announce the conclusion: if $|\bar{n}| \gg 1/\ell^2$, then the fluctuations of $\delta l^{-1}(\mathbf{r})$ are self-averaging on the scale of $l(\bar{n})$. In other words, corrections to $l(\bar{n})$ are parametrically small. For simplicity, I will ignore them together with corrections to $\chi(\mathbf{r})$ [the second term in Eq. (14)] to obtain

$$\bar{\sigma}(\mathbf{r}) \simeq \sigma(\bar{n}(\mathbf{r})), \qquad |\bar{n}(\mathbf{r})| \gg 1/\ell^2. \qquad (19)$$

Problems arise at $|\bar{n}| \lesssim 1/\ell^2$ where the corrections to Eq. (19) exceed $\sigma(\bar{n})$. In the classical regime $\bar{\sigma} \gg e^2/h$, we could have handled this by adopting a model form

$$\bar{\sigma}(\mathbf{r}) = \sigma_0\zeta(\mathbf{r}), \qquad |\bar{n}| \lesssim 1/\ell^2, \qquad (20)$$

where $\zeta(\mathbf{r}) > 0$ is some random function with the correlation length $\Lambda^{-1} \sim \ell$ and the typical value of the order of unity [27]. However, in graphene $\sigma_0 \equiv \sigma(1/\ell^2) = (8/\pi)e^2/h$ is so low that the very concept of local conductivity is potentially jeopardized by quantum interference and localization effects. Fortunately, Eq. (20) is saved by the special geometry of the regions where it is intended to be used. These regions typically form ribbons of width $x \sim \ell$ that follow the PNJ loops. These low-conductance

"ribbons" are connected to high-conductance "reservoirs" on both sides of the PNJ. In such a geometry the importance of the quantum effects is controlled [30] by the total conductance G_p across the perimeter length p of the PNJ. For $p \gg \ell$, it is given by

$$G_p \sim \sigma_0\frac{p}{\ell} \gg \frac{e^2}{h}, \qquad (21)$$

in which case the semiclassical model (20) is justified. Again we have succeeded in reducing the complicated quantum problem to a simpler classical one: finding the macroscopic conductivity σ_{NP} of an inhomogeneous medium with local conductivity given by Eqs. (19) and (20) as a function of the local density $\bar{n}(\mathbf{r})$.

A rigorous upper bound on σ_{NP} is the spatially averaged conductivity. Because of ergodicity of Gaussian fields, the averaging can be done over $\bar{n}$ instead. Using Eqs. (16) and (18), I obtain:

$$\sigma_{\mathrm{NP}} < \langle\bar{\sigma}\rangle = \int d\bar{n}P_n(\bar{n})\bar{\sigma}(\bar{n}) \simeq (4\mathcal{L}/\pi)e^2/h. \qquad (22)$$

(Incidentally, this enables me to conclude that Ref. [13] overestimates σ_{NP} in the limit $\alpha \ll 1$.)

Now I present the argument crucial to my theory of transport. It shows that σ_{NP} is *not* sensitive to the details of the model form (20). Instead, it is determined by the typical local conductivity, and so is not far below the upper bound (22). This statement should be contrasted with other theoretical views on the subject. Since the low-conductivity regions (20) reside at the percolation contour, it has been suggested [1,13,31] that the transport at the NP may be governed by percolation [25]. In that picture the current paths are severely constrained in order to avoid crossing the PNJ as much as possible and σ_{NP} depends on their average transparency [31].

I show that in the model under study the closed-loop PNJ contours are *not* resistive enough for the percolation effects to develop. Indeed, the percolation approach [31] would apply only if there existed a wide range of loop diameters $d \gg \ell$ such that the conductance G_p of the loop perimeter were much lower than the conductance G_d of their interior. Suppose first that $d \ll R$ for such loops. Let us show that it leads to a contradiction. If the current avoids crossing the perimeter, it has to flow in narrow channels of some width $w \ll d$ due to the fractal geometry of the loop. Hence, $G_d \sim \bar{\sigma}_{\mathrm{int}}w/d \ll \sigma_{\mathrm{int}}$ where σ_{int} is the typical local conductivity inside the loop. To estimate the latter, I note that from the general properties of Gaussian random fields [25] the potential $\bar{\Phi}$ at a distance r from the PNJ ($\bar{\Phi} = 0$) has the variance

$$\langle\bar{\Phi}^2\rangle_{\mathrm{PNJ}} = K(\ell) - \frac{K^2(r)}{K(\ell)} \simeq \frac{\pi}{4}\left(\frac{\hbar v}{\ell}\right)^2\ln\left(\frac{r}{\ell}\right), \qquad (23)$$

so that the corresponding density is $\bar{n} \sim \ln(r/\ell)/\ell^2$. Using $r \sim d$ and Eq. (18), I get $\sigma_{\mathrm{int}} \sim \sigma_0\ln(d/\ell)$. In comparison, $G_p \sim \sigma_0(d/\ell)^{3/2}$, cf. Eqs. (17) and (21). Hence,

$G_p \gg G_d$, which contradicts the assumption made (the same is true for $d \gg R$). I conclude that in the present model the percolation-type transport is not realized.

In the absence of strong ramification of current paths by the PNJ, I expect the numerical coefficient c in Eq. (7) to be of the order of unity. To estimate it more accurately I use the effective medium theory (EMT). Indeed, the EMT is usually adequate for systems where percolation effects are unimportant. Within the EMT, σ_{NP} is determined from a certain nonlinear equation, the two most popular versions of which were originally proposed in Refs. [32,33]:

$$\left\langle \frac{\sigma_{NP}}{\bar{\sigma} + (\mathcal{D} - 1)\sigma_{NP}} \right\rangle = \frac{1}{\mathcal{D}} \quad \text{(Bruggeman)}, \qquad (24)$$

$$\int_0^\infty \frac{dz}{e^z} \ln\left\langle \exp\left[\frac{z}{\mathcal{D}} \frac{\bar{\sigma}}{\sigma_{NP}}\right] \right\rangle = \frac{1}{\mathcal{D}} \quad \text{(Hori)}. \qquad (25)$$

Here $\mathcal{D} = 2$ is the space dimension. Equations (24) and (25) can be viewed [33] as an approximate resummation of the infinite diagrammatic series for the macroscopic conductivity using, respectively, the self-consistent single-site approximation and the cumulant expansion.

The averages in Eqs. (24) and (25) are dominated by typical $\bar{\sigma}$, so it suffices to use $P_n(\bar{n})$ from Eq. (16). A straightforward numerical solution of these equations then gives: $\sigma_{NP}(\text{Bruggeman}) = 0.48e^2/h$ and $\sigma_{NP}(\text{Hori}) = 0.55e^2/h$. Taking the difference of the two as a measure of their accuracy, I arrive at Eq. (7).

Finally, I briefly comment on experimental implications of the presented theory. For simplicity, I have assumed the dielectric constant of the medium to be the same on both sides of graphene. It is more realistic to have graphene at the interface of a half-space with dielectric constant $\kappa_1 \sim 1$ and a film of dielectric constant $\kappa_2 \gg 1$ and thickness D, whose other side is covered by a metallic gate. In this geometry κ is replaced by $(\kappa_1 + \kappa_2)/2$ while $\mathcal{L}$ becomes $\min(\ln(R/\ell), \ln(D/\ell))$. Small α can be achieved experimentally using ice [4], ethanol, and other dielectrics [34]. However, it is difficult to make $\mathcal{L}$ larger than $\mathcal{L} \sim 5$, for which Eq. (7) predicts $\sigma_{NP} \sim 2.5e^2/h$. This is 2 to 3 times lower than the measured [1–4] σ_{NP}. This may indicate that in experiment the charged impurities are either not exactly coplanar with graphene, or are correlated, or are not the only source of disorder. The last possibility is further corroborated by a very modest increase in mobility [34] away from the NP upon a large *in situ* increase of κ.

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