



Theory of charged impurity scattering in two-dimensional graphene

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ABSTRACT

We review the physics of charged impurities in the vicinity of graphene. The long-range nature of Coulomb impurities affects both the nature of the ground state density profile and graphene's transport properties. We discuss the screening of a single Coulomb impurity and the ensemble averaged density profile of graphene in the presence of many randomly distributed impurities. Finally, we discuss graphene's transport properties due to scattering off charged impurities both at low and high carrier density.

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1. Introduction

Graphene is a two-dimensional (2D) sheet of carbon whose atoms arrange in a honeycomb lattice with nearest neighbor atoms forming strong sp_2 bonds. The electronic properties of this material are mostly determined by the p_z orbitals with each carbon atom contributing one electron to a Bloch band whose low-energy properties are adequately described by a Dirac–Weyl effective Hamiltonian. While the study of Dirac Fermions has emerged in several contexts in theoretical condensed matter physics, its experimental realization about three years ago, in the form of gated graphene devices [1–3], where the carrier density can be tuned continuously from electron-like carriers for positive bias to hole-like carriers for negative gate voltage, has prompted a prolific theoretical and experimental effort to understand the properties of this novel material.

Most of the excitement surrounding graphene stems from one of the following peculiar properties: (i) electrons and holes in graphene have a gapless linear dispersion relation in contrast to the parabolic dispersion of other more conventional electron gases; (ii) the carriers in graphene are chiral – a property that has striking consequences such as the “half-integer” quantum Hall effect [2,3]; and (iii) carriers in graphene live at an exposed almost perfect 2D surface that is amenable to surface probes [4–8] and surface manipulation [9,10]. In addition, we note that there is the potential of mass producing graphene through epitaxial growth

methods [11], and that graphene has remarkable mechanical properties [12,13] which only further enhance the interest.

In this perspective, we look at one important aspect of graphene which is the influence of disorder on its ground state and transport properties. We demonstrate that for graphene, charged (i.e. Coulomb) impurities behave qualitatively different from neutral impurities [14–16] and dominate graphene's transport properties at low carrier density. The importance of the Coulomb nature of graphene impurities was highlighted by Ando [17], where by calculating the intraband contribution to the polarizability and absorbing the interband (i.e. electron–hole) contribution into a redefinition of the dielectric constant [18] he showed that charged impurities could explain the conductivity being linear in density, as was seen in experiments [1,19]. Similar conclusions were obtained by Nomura and MacDonald [20] using a “complete screening” model (i.e. $r_s \rightarrow \infty$, see definition below), Cheianov and Fal'ko using a numerical Thomas–Fermi approximation [21] and in Ref. [22] using the full Random Phase Approximation (RPA). Analytic expressions for the RPA polarizability function calculated first in Ref. [23] and then in Refs. [24–26] revealed that for momentum transferred on the Fermi circle (i.e. $q = |\mathbf{k} - \mathbf{k}'| = 2k_F \sin \theta/2 \leq 2k_F$) the graphene dielectric function calculated using the RPA was identical to the much simpler Thomas–Fermi approximation at $T = 0$ (see Fig. 1). This then made it possible to calculate the RPA Boltzmann conductivity analytically [27], and the dependence of graphene's conductivity on the fine-structure constant $r_s \equiv e^2/(\hbar v_F \kappa)$ was recently verified experimentally [10]. The importance of Coulomb scattering in explaining the observed graphene transport properties soon prompted an interest in investigating the properties of a single

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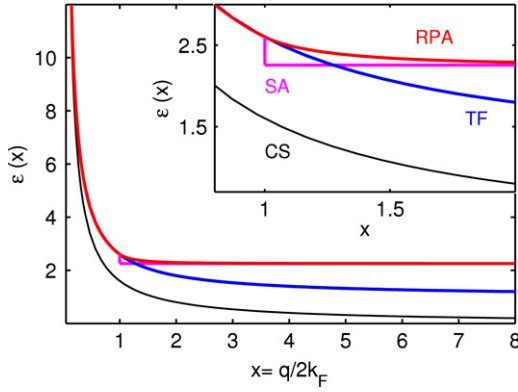


Fig. 1. The main panel shows different dielectric functions used in the literature, including the “Complete Screening” (CS), “Thomas–Fermi” (TF) and “Random Phase Approximation” (RPA). The inset is a blow-up at $q = 2k_F$ to show where the “Step Approximation” (SA) used in Ref. [27] differs from the exact result.

charged impurity embedded in graphene. Katsnelson [28] studied this problem using a Fermi–Thomas approximation, followed by studies in Refs. [29–35] which were mostly interested in effects beyond the RPA such as determining the critical impurity charge for which the Coulomb impurity forms bound states and the screening properties of graphene in the supercritical regime.

It was understood by Refs. [22,36] that as one approached the Dirac point, one would soon encounter a situation where the gate voltage induced carrier density would be smaller than the fluctuation of carrier density induced by the charged impurities thereby breaking the graphene landscape into puddles of electrons and holes. Solving numerically for the conductivity using a finite-sized Kubo formalism for a limited range of impurity concentrations, Ref. [36] concluded that the Coulomb disorder model gave a *universal* minimum conductivity whose value did not depend on the charged impurity concentration, but that was larger than that expected for clean Dirac Fermions [28,37–40], while Ref. [22] argued that this would give rise to a *non-universal* minimum conductivity whose value depended on the concentration of charged impurities. Ref. [27] developed a mean-field approach to understand the properties of graphene at the Dirac point by calculating an effective carrier density self-consistently. This theory made quantitative predictions about the dependence of the minimum conductivity and rms carrier density on the charged impurity concentration and substrate dielectric constant, and in particular argued that cleaner graphene samples would have larger minimum conductivity. Ref. [41] then studied the ground state properties of graphene by minimizing an energy functional comprising kinetic energy, Hartree, exchange [25,42,43] and correlation [25,43,44] contributions in the presence of Coulomb disorder. This work made quantitative predictions about properties of the carrier density distribution, both at and away from the Dirac point, and enabled Ref. [45] to develop an effective medium theory to calculate the graphene’s conductivity through these inhomogeneous puddles, capturing quantitatively the minimum conductivity plateau that is seen in experiments [1,19,46]. We mention that underlying the existence of this minimum conductivity plateau is the high transmission of graphene p–n junctions, which has been the subject of theoretical [47,48] and experimental [49–51] studies. For the purposes of this paper we do not discuss quantum interference effects (see Ref. [52] and references therein) or the strongly interacting regime (see Ref. [53] and references therein).

The remainder of this paper is structured as follows. In Section 2 we discuss the problem of the screening of a single Coulomb impurity in the sub-critical regime as a useful toy model to understand the many-impurity problem that we address in

Section 3, where we study the case of many Coulomb impurities that are uncorrelated and distributed uniformly in order to study the ground state properties of graphene. In Section 4.1, we review the high-density Boltzmann transport theory, and discuss the Effective Medium Theory (EMT) in Section 4.2. In Sections 5–7 we briefly review the experimental situation, discuss graphene’s minimum conductivity, and recent theoretical work not covered in this review. We then conclude in Section 8.

2. Screening of a single Coulomb impurity

Following Ref. [28], one can construct the Thomas–Fermi screening of a single charged impurity. The goal is to calculate the screened Coulomb potential $V_s(r) = V_0(r) + V_{\text{ind}}(r)$, where the bare potential $V_0 = \hbar v_F r_s [r^2 + d^2]^{-1/2}$ and the induced potential is given by

$$V_{\text{ind}}(r) = (\hbar v_F r_s) \int \frac{d\mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|} \frac{n(r') - \bar{n}}{r + r'}, \quad (1)$$

where we can imagine tuning the back gate to ensure charge neutrality $\bar{n} = 0$. If one further assumes that the local carrier density is given by the Fermi–Thomas condition, $n[V(r)] = -V(r)^2 / [\pi (\hbar v_F)^2]$, one can write down a (one-dimensional) self-consistency equation for $\tilde{V}_s = V_s / \hbar v_F r_s$:

$$\tilde{V}_s(r) = \frac{1}{\sqrt{r^2 + d^2}} - \frac{4r_s^2}{\pi} \int \frac{dr'}{r + r'} K \left[\frac{4rr'}{(r + r')^2} \right] \tilde{V}_s^2(r'), \quad (2)$$

where $K[x]$ is the complete elliptic integral of the first kind. The screened potential induced for this single impurity using this method was discussed in Ref. [28]. This formalism can be generalized using the method developed in Ref. [41] to include the effects of exchange. The ground state carrier density can be obtained from the Thomas–Fermi–Dirac (TFD) energy functional

$$E[n] = \hbar v_F \left[\frac{2\sqrt{\pi}}{3} \int d^2 r \text{sgn}(n) |n|^{3/2} + \frac{r_s}{2} \int d^2 r \int d^2 r' \frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \frac{E_{xc}[n]}{\hbar v_F} + r_s \int d^2 r V_D(\mathbf{r})n(\mathbf{r}) - \frac{\lambda}{\hbar v_F} \int d^2 r n(\mathbf{r}) \right] \quad (3)$$

where the first term in Eq. (3) is the kinetic energy, the second term is the Hartree part of the Coulomb interaction, the third term is the exchange–correlation energy and the fourth term is the energy due to disorder, where V_D is the disorder potential and the last term is added to set the average carrier density, $\langle n \rangle$, through the chemical potential λ . The correlation term is much smaller than the exchange and, to very good approximation [25,43,44], is proportional to the exchange. Therefore, hereafter, we neglect the correlation contribution by assuming $\delta E_{xc} / \delta n = \Sigma(n)$, where $\Sigma(n)$ is the Hartree–Fock self-energy [25,43,42]. The energy functional equation (3) is quite general and can be tailored by properly choosing V_D and its coupling to $n(\mathbf{r})$, to consider different sources of disorder. For the single-impurity problem, the solution can also be cast as a one-dimensional integral equation:

$$\begin{aligned} \frac{V_s(r)}{\hbar v_F} &= \frac{r_s}{\sqrt{r^2 + d^2}} + 4r_s \int \frac{dr'}{r + r'} K \left[\frac{4rr'}{(r + r')^2} \right] n(r') \\ &= -\text{sgn}(n) \sqrt{\pi |n(r)|} \left[1 + \frac{1}{4} \ln \left(\frac{4\Lambda}{\sqrt{\pi |n(r)|}} \right) \right] \\ &\quad + r_s \left(\frac{2C + 1}{2\pi} + \frac{1}{8} \right), \end{aligned} \quad (4)$$

where $\Lambda = 1/(0.25 \text{ nm})$ is the band energy cutoff and $C \approx 0.916$.

3. Ground state properties at the Dirac point

The single-impurity problem discussed in the previous section is a much simpler problem because rotational symmetry makes the problem one dimensional. Adding many impurities also brings further complications: while the carrier density induced by a single impurity is negligible, this is not the case for many impurities, where although the average density can be tuned to zero via an external gate potential, the scale of the density fluctuations is set by the impurity concentration [22]. As shown in Fig. 1, the RPA screening properties of graphene are very different at the Dirac point (i.e. $k_F \rightarrow 0$) and at finite density; therefore, theoretical frameworks constructed to work at the Dirac point are bound to fail when there are such large density fluctuations. In this section, we present two different approaches to describe the Dirac point. The first is a mean-field theory where an effective density n^* is obtained by solving self-consistently for the density induced by the fluctuations of the screened impurity potential (that itself depends on the density). The second is a generalization of the energy functional method discussed above for a single impurity to the much more complicated case of many Coulomb impurities.

3.1. Self-consistent approximation (SCA)

For any microscopic single-impurity potential $\phi(r, n)$, the probability distribution, $P(V)$, of the total potential, V , is $P(V) = \langle \delta(V - \sum_{i=1}^{N_{\text{imp}}} \phi(r_i, n)) \rangle_{r_i}$, where $\langle \dots \rangle_{r_i}$ is the average over all possible disorder configurations. Assuming that the impurities' positions are uncorrelated, one can compute expressions for all moments of the induced disorder potential [54,55]. For example, the connected moment $\langle V^k \rangle_c = n_{\text{imp}} \int d^2r [\phi(r, n)]^k$. The self-consistent approximation involves obtaining the effective carrier density n^* by equating the second moment of the disorder potential with the square of the corresponding Fermi energy $\langle V^2 \rangle_c = (E_F[n^*])^2 = \pi(\hbar v_F)^2 n^*$. This self-consistent approximation then allows us to compute any correlation function at the Dirac point, although closed form analytic results are often elusive. To make analytical progress, one can map

$$\langle V(r)V(0) \rangle = n_{\text{imp}} \int d\mathbf{q} [\phi(q, n^*)]^2 e^{i\mathbf{q}\cdot\mathbf{r}} \quad (5a)$$

$$\approx \frac{n_{\text{imp}}(\hbar v_F)^2 K_0[r_s, d\sqrt{n^*}]}{2\pi(\xi[r_s, d\sqrt{n^*}])^2} \exp\left[\frac{-n_{\text{imp}}r^2}{2(\xi[r_s, d\sqrt{n^*}])^2}\right], \quad (5b)$$

where analytic expressions for K_0 and ξ were reported in Ref. [56]. A numerical evaluation of Eq. (5a) and the Gaussian approximation Eq. (5b) is shown in Fig. 2. Within the Gaussian approximation one finds that $n_{\text{rms}} = \sqrt{\langle V^2 \rangle_c} / [\pi(\hbar v_F)^2] \approx n^* \sqrt{3 + [n_{\text{imp}}\pi\xi^2]^{-1}} \approx \sqrt{3}n^*$, where in the last equation we further assume that $n_{\text{imp}}\pi\xi^2 \sim r_s^{-4} \gg 1$. This result for n_{rms} is particularly useful when comparing the self-consistent approximation with other methods.

3.2. Energy Functional Minimization (EFM)

To study graphene's transport properties for a distribution of charged impurities, we use Eq. (3), taking V_D to be the potential generated by a random 2D distribution $C(\mathbf{r})$ of impurity charges placed at a distance d from the graphene layer of size $L \times L$. We assume $C(\mathbf{r})$ to be on average zero and uncorrelated, and perform our calculations on a 200 nm \times 200 nm square sample with a 1 nm spatial discretization. Close to the Dirac point, for a single-disorder realization, we find that the carrier density breaks up into electron–hole puddles. Since we are interested in disorder averaged quantities, we examine several

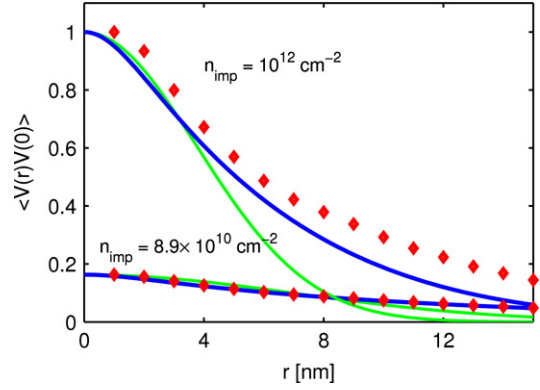


Fig. 2. Spatial correlation function for the screened potential at the Dirac point for $r_s = 0.8$ and $d = 1$ nm. Red diamonds are the results obtained by minimizing graphene's energy functional (Section 3.2). The lines are the SCA results using Eq. (5a), blue lines, and its Gaussian approximation, Eq. (5b). All the results are normalized via the value of $\langle V(0)V(0) \rangle$ for $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$.

disorder realizations (500–1000) and denote disordered averaged quantities by angled brackets. To characterize the density profile, we calculate the disorder averaged density–density correlation function $\langle \delta n(\mathbf{r})\delta n(0) \rangle$, from which we can extract the root mean square $n_{\text{rms}} = \sqrt{\langle \delta n(0)\delta n(0) \rangle}$, and the typical correlation length, ξ , defined in this section as the full width at half maximum (FWHM) of $\langle \delta n(\mathbf{r})\delta n(0) \rangle$. We find [41], for typical graphene samples, that $n_{\text{rms}} \approx \langle n \rangle$ for dopings as high as 10^{12} cm^{-2} and that close to the Dirac point, for $n_{\text{imp}} \lesssim 10^{10} \text{ cm}^{-2}$, n_{rms} including exchange is three times smaller than without. We find the correlation length ξ to be of the order of 10 nm; see Fig. 3. This value suggests that the electron–hole puddles are quite small. However, a closer inspection reveals that, close to the Dirac point, the density profile is characterized by two distinct types of inhomogeneities [45]: wide regions (i.e. big puddles spanning the system size) of low density containing a number of electrons (holes) of order $n_{\text{rms}}L^2$; and few narrow regions, whose size is correctly estimated by ξ , of high density containing a number of carriers of order 2. This picture is confirmed by the results shown in Fig. 4, in which the disorder averaged area fraction, A_0 , over which $|n(\mathbf{r}) - \langle n \rangle| < n_{\text{rms}}/10$ is plotted as a function of n_{imp} . We see that A_0 is of order 1/3, and we also find that the area fraction over which $|n(\mathbf{r}) - \langle n \rangle|$ is less than 1/5 of n_{rms} is close to 50% for $n_{\text{imp}} \lesssim 10^{11} \text{ cm}^{-2}$. The combination of the relatively high density in the peaks/dips and the fact that in the low-density regions $n(\mathbf{r})$ varies over scales much bigger than 10 nm guarantees that the inequality $\sqrt{\pi n} [|\nabla n|/n]^{-1} \gg 1$ is satisfied over the majority of the graphene sample and therefore justifies the use of the EFM theory. EFM should be a reasonable quantitative theory for existing graphene samples at all values of the carrier density.

3.3. Comparison of SCA and EFM

Here we compare the results from the Self-Consistent Approximation (SCA) of Section 3.1 and the Energy Functional Minimization (EFM) formalism discussed in Section 3.2. Fig. 2 shows the disordered averaged spatial correlation function at the Dirac point for the screened disorder potential $V = V_D + (1/2) \int d^2r' n(\mathbf{r}') / |\mathbf{r} - \mathbf{r}'|$, where the (red) diamonds are obtained by minimizing Eq. (3). The solid (blue) line shows the same quantity calculated using the self-consistent approximation (SCA). The EFM approach and the SCA give a similar behavior for $\langle V(r)V(0) \rangle$, characterized by an algebraic, $\propto 1/r^3$, decay at large distances. The green solid line shows a Gaussian approximation which captures much of the quantitative details of the screened disorder potential correlation function, but not the power-law $1/r^3$ decay.

