

Conductivity of defectless graphene

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The direct current conductivity of defectless, perfect crystal graphene is found at the neutrality point at zero temperature and in the limit of large dielectric constant of the substrate. The nonequilibrium steady state with weak current is assumed to be an ideal, rare plasma of particle and hole excitations adequately described by the Boltzmann kinetic equation.

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Conductivity of graphene, a flat monolayer of carbon atoms, as a function of doping charge shows a pronounced minimum at the neutrality, compensation point¹⁻³. The theory predicts for this point a universal conductivity⁴, whereas the experimental conductivity exceeds this prediction by few times. This discrepancy persists for long time now and may justify an additional study of zero gap semiconductors, with graphene being an example. Needless to say, adequate understanding of the physical mechanisms at the compensation point is also important in some of proposed future applications of graphene in electronic devices. In my view, the problem is that a concept of non-interacting quasiparticles, well established in normal Fermi liquids, is being translated onto the graphene without careful consideration. In normal Fermi liquids the Galilean invariance makes the electron-electron interaction ineffective to relax the current and the Fermi liquid can flow as a whole. Also the rate of electron-electron scattering is relatively small $\sim T^2/\epsilon_F$ for large Fermi surfaces. Hence, the transport at low temperature is determined by disorder. On the other hand, the neutral graphene is different. In its Brillouin zone there exist two points where the electron dispersion acquires a cone-like shape, exactly the relativistic massless Dirac dispersion⁵. This feature is simply understood using the tight binding model on the honeycomb lattice⁶, that may represents the band structure of the graphene. At the cone apexes the two crystal bands of graphene meet. The valence band is filled at the compensation point whereas the conduction band is empty. At low temperature the electronic excitations of two types: particles and holes, are present in the vicinity of these two apexes. This special crystal band structure transform the Galilean invariance into Lorentz one but instantaneous Coulomb interaction breaks it and the current is not conserved. Therefore, in defectless graphene at compensation point the current can relax in the process of the Coulomb interaction alone. As the Fermi circle degenerates into two points the rate of Coulomb scattering is no longer weak $\sim T$. All this special features of graphene call for a study of the role of Coulomb interaction on the evolution of graphene charge carriers. One impor-

tant motivation for such study is the recent success in the theory of scattering on charged impurity⁷ to explain a linear dependence of the conductivity as a function of the doping charge away from the compensation point³.

In this paper, the conductivity of defectless graphene is found at the compensation point for weak Coulomb interaction and assuming that charge carriers constitute almost ideal, rare plasma of particles and holes. The result contradicts the universal conductivity⁴ for non-interacting Dirac fermions. To clarify this I studied numerically unitary evolution of non-interacting fermions in the valence band of graphene under the influence of n -cycle pulse of alternating electric field E . After the pulse ceases the resulting fermion transitions into the conduction band can be examined. The excitation energy is found to be independent of the pulse length n , and is proportional to $E^{5/2}$. Thus, the scattering free evolution of non-interacting graphene in ac-field leaves behind no traces of Joule heat.

The main precondition for non-conservation of the current is being close to the compensation point. Non-interacting particles and holes do separate in the electric field, although, in the momentum space they both move in the same direction. However, a neutral particle-hole cloud coupled by strong Coulomb forces behaves like a collection of pairwise neutral atoms. The response of these to the electric field is, initially, a polarization rather than a current. Therefore, a precise value of the conductivity is determined by the mutual Coulomb interactions. A microscopic process that changes the current is shown in Fig.1. A pair of a particle 1 and a hole 2 has zero net momentum and non-zero net current. The electron velocity in the state 2 is opposite to the electron velocity in the state 1. But a hole is the absence of an electron, therefore, the total current of the pair (1,2) is non zero. Coulomb interaction scatters the pair (1,2) into a new position (3,4) with the same total momentum and energy. We observe that the net pair current in the new state (3,4) is reversed.

Consider infinite and perfect graphene on top of dielectric substrate at zero temperature. Applying an electric field E will create particle and hole excitations due to the

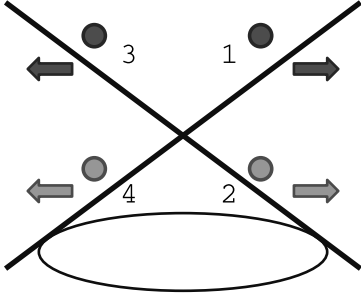


FIG. 1: Scattering of the particle-hole pair (1,2) into the particle-hole pair (3,4) that conserves the momentum and energy but changes the current. Excitation velocities are shown by arrows. x-axis is the momentum and y-axis is the excitation energy. The matrix element for this specific process is zero but variation of momenta makes it non-vanishing.

Schwinger mechanism. The work of E on these excitations is the Joule heat that will induce lattice vibrations near the graphene layer and will eventually escape into the bulk. In the balance, a steady distribution of particle and hole excitations in graphene will be established. We assume this state to be an almost ideal, rare plasma with the excitation distribution given by the Fermi-Dirac function for some effective temperature T^* .

The graphene Hamiltonian in the long wavelength limit includes the Coulomb part and the crystal band part⁵:

$$\hat{H} = c(\hat{\tau}^z \hat{\alpha}^x p_x + \hat{\alpha}^y p_y), \quad (1)$$

where $\mathbf{p} = (p_x, p_y)$ is momentum and $\hat{\alpha}^x, \hat{\alpha}^y, \hat{\tau}^z$ are the Pauli matrices, the first two act in the representation space of the crystal point group and the last one acts in the valley space. $c \approx 10^8$ cm/s is the characteristic band velocity that determines the cone angle⁵. The total degeneracy of electron states in graphene is $N = 4$ due to the spin and valley. The Hamiltonian (1) is diagonalized by unitary transformation: $(1 + \hat{\alpha}^y) \exp(i\hat{\tau}^z \hat{\alpha}^z \phi/2)/\sqrt{2}$ into two crystal bands, two halves of the cone: $\epsilon_{\tau\sigma}(\mathbf{p}) = \tau\alpha|\mathbf{p}|$, where $\tau, \alpha = \pm 1$ are eigenvalues, with α specifying the two crystal bands. At the compensation point the electron state of graphene is determined by a dimensionless Coulomb coupling:

$$g = \frac{e^2}{\kappa\hbar c} \quad (2)$$

where κ is the half of the dielectric constants of the substrate and the vacuum. For graphene on top of SiO₂ substrate $g \approx 0.8$ whereas for graphene suspended in vacuum: $g \approx 2$. We find the dispersion in the second order of perturbation in the Coulomb interaction:

$$\epsilon(\mathbf{p}) = c_R |\mathbf{p}| \left(1 - g_R^2 \left(\frac{N}{24} - 0.01308 \right) \log \log \frac{Q}{|\mathbf{p}|} \right) \quad (3)$$

in the long wavelength limit $|\mathbf{p}| \ll Q$, where Q is the size of the Brillouin zone. Although in 3D zero-gap semiconductor the renormalization group is called upon to

described the critical indices⁸, in 2D graphene RG degenerates to a simple Hartree-Fock renormalization of the cone velocity and the Coulomb coupling⁹:

$$c_R = c \left(1 + \frac{g}{4} \log \frac{Q}{|\mathbf{p}|} \right) \quad g_R = \frac{e^2}{\kappa\hbar c_R} \quad (4)$$

In addition to the non-equilibrium state of graphene with the current described by the electron distribution function in momentum space: $F_\alpha(\mathbf{p})$, we imagine also an 'equilibrium' state with relaxed, zero current but with the same excitation energy. We disregard here graphene states with particle-hole coherence of any kind [see e.g.¹⁰], since this may lead to the time dependence of the coherence order parameter averaging out its effect. In the end $F_\alpha(\mathbf{p})$ will not depend on either spin or valley indices. Since the total momentum of the scattered electrons is conserved in the crystal [neglecting the Umklapp processes], we search for the graphene state with zero total momentum. In this state there are on average as many holes as particles in every small momentum cell. If the distribution function for the particles is $F_+(\mathbf{p})$ then the distribution function for the holes $1 - F_-(\mathbf{p})$ is the same. Therefore, there holds the particle-hole symmetry: $1 - F_\alpha(\mathbf{p}) = F_{-\alpha}(\mathbf{p})$. In the 'equilibrium' state the electron distribution function:

$$f_\alpha(\mathbf{p}) = 1/(\exp(\alpha|\mathbf{p}|/\langle p \rangle) + 1) \quad (5)$$

makes the collision integral to vanish for any momentum scale parameter: $\langle p \rangle$, which is related to the effective temperature of the electrons in the 'equilibrium' state of the graphene: $T^* = c_R \langle p \rangle$, where c_R in Eq.(4) is evaluated at $|\mathbf{p}| = \langle p \rangle$. The distribution function Eq.(5) satisfies the electron-hole symmetry: $f_{-\alpha}(\mathbf{p}) = 1 - f_\alpha(\mathbf{p})$. We rescale isotropically the momentum space in the vicinity of the cone apexes and set $\langle p \rangle = 1$.

The Boltzmann kinetic equation defines the steady state distribution function balancing the two processes - the acceleration of the excitations in the electric field and their redistribution in the course of collisions¹¹:

$$e\vec{E} \frac{\partial F_\alpha}{\partial \mathbf{p}} = St_\alpha(\mathbf{p}) \quad (6)$$

In the lowest order of the Coulomb coupling g , the second order Fermi golden rule, the collision integral reads [for short notations α_3, α_4 of the out-going electrons are inverted]:

$$St_{\alpha_1}(\mathbf{p}_1) = \sum_{\alpha_2\alpha_3\alpha_4} \iiint \text{Tr}_\tau (|V_{\alpha_1\alpha_2}^{\alpha_3\alpha_4}(\mathbf{p}_1\mathbf{p}_2\mathbf{p}_3\mathbf{p}_4)|^2) \times \\ (2\pi)\delta(\sum_{i=1}^4 \alpha_i \epsilon(\mathbf{p}_i)) \left(\prod_{i=1}^4 F_{\alpha_i}(\mathbf{p}_i) - \prod_{i=1}^4 F_{-\alpha_i}(\mathbf{p}_i) \right) \times \\ (2\pi)^2 \delta(\mathbf{p}_1 + \mathbf{p}_2 - \mathbf{p}_3 - \mathbf{p}_4) \frac{d^2\mathbf{p}_2 d^2\mathbf{p}_3 d^2\mathbf{p}_4}{(2\pi)^6} \quad (7)$$

Below we use interchangeably the notation: $\mathbf{p}_1 = \mathbf{p}$, $\mathbf{p}_2 = \mathbf{p}'$, $\mathbf{p}_3 = \mathbf{p}' + \mathbf{q}$ and $\mathbf{p}_4 = \mathbf{p}' - \mathbf{q}$. Since the excitation

plasma is assumed to be rare in the limit $\langle p \rangle \ll Q$, with the Debye screening radius being large $R_D \sim \hbar^2 c_R^2 / e^2 T^*$, the Coulomb matrix element is weakly screened:

$$V_{\alpha_1 \alpha_2}^{\alpha_3 \alpha_4}(\mathbf{p}_1 \mathbf{p}_2 \mathbf{p}_3 \mathbf{p}_4) = \frac{1}{2} \left(\frac{2\pi e^2}{\kappa |\mathbf{p}_1 - \mathbf{p}_3|} \frac{1 - z_1 z_3^*}{2} \frac{1 - z_2 z_4^*}{2} \delta_{\tau_1 \tau_3} \delta_{\tau_2 \tau_4} - \frac{2\pi e^2}{\kappa |\mathbf{p}_2 - \mathbf{p}_3|} \frac{1 - z_2 z_3^*}{2} \frac{1 - z_1 z_4^*}{2} \delta_{\tau_1 \tau_4} \delta_{\tau_2 \tau_3} \right) \quad (8)$$

where the notation $z_i = \alpha_i(p_i^x + ip_i^y)/|\mathbf{p}_i|$ is used. τ indices run over the total $N = 4$ spin-valley degeneracy space of the graphene. The square of the matrix element Eq.(8) includes two terms - the direct and exchange ones. The exchange term vanishes when two scattering excitations have different spins or valleys.

In the graphene state with current the electron distribution function can be sought in the form:

$$F_\alpha(\mathbf{p}) = \frac{1}{\exp \left[\alpha |\mathbf{p}| + \alpha (e\vec{E} \cdot \vec{p}) \chi(|\mathbf{p}|)/|\mathbf{p}| \right] + 1} \quad (9)$$

where $\chi(\mathbf{p})$ defines the perturbation of the distribution in electric field. It is better to satisfy the condition $\chi(\mathbf{p}) \rightarrow 0$ as $|\mathbf{p}| \rightarrow 0$. Also, $\chi(\mathbf{p})$ in Eq.(9) explicitly conserves the number of electrons, their total energy and their total momentum¹¹. We linearize the Boltzmann kinetic equation Eq.(6) with respect to $\chi(\mathbf{p})$ ¹¹. The linearized collision integral becomes a matrix, which is symmetric due to a detailed balance of the direct and time-reversed processes in the steady state. The linear response current reads:

$$\vec{j}[\chi] = -N \frac{e^2}{\hbar} \sum_{\alpha} \int \frac{\chi(|\mathbf{p}|)}{|\mathbf{p}|^2} \vec{p} (\vec{p} \cdot \vec{E}) f_\alpha(\mathbf{p}) f_{-\alpha}(\mathbf{p}) \frac{d^2 \mathbf{p}}{(2\pi)^2} \quad (10)$$

where the band velocity $\vec{v}_\alpha = \alpha c_R \vec{p}/|\mathbf{p}|$. For exact linear dispersion and for the collinear orientation of all momenta $\mathbf{p}||\mathbf{p}'||\mathbf{q}$ the argument of the energy delta-function in Eq.(7) becomes degenerate [many technical details of what follows are thoroughly discussed in Ref.¹²], i.e. $\alpha_1|\mathbf{p}| + \alpha_2|\mathbf{p}'| + \alpha_3|\mathbf{p} + \mathbf{q}| + \alpha_4|\mathbf{p}' - \mathbf{q}| = 0$ for any $|\mathbf{p}'|$ and $|\mathbf{q}|$ provided three conditions are met: $\alpha_1 = \alpha_2 \text{sgn}(\mathbf{p} \cdot \mathbf{p}_2) = -\alpha_3 \text{sgn}(\mathbf{p} \cdot \mathbf{p}_3) = -\alpha_4 \text{sgn}(\mathbf{p} \cdot \mathbf{p}_4)$. Taking the direction of the vector \mathbf{p} as x and expanding around the collinear configuration of four momenta we find:

$$\Delta E = \frac{p_y'^2}{2p'} - \frac{q_y^2}{2(p+q)} - \frac{(p_y' - q_y)^2}{2(p' - q)} + \frac{g}{4} \sum_i p_i \log |p_i| \quad (11)$$

where $p_i = p_{ix}$ and where the effect of the renormalization of the velocity $\delta c = (g/4) \log(Q/\langle p \rangle)$ is omitted. It suffices to approximate the last term in Eq.(11) as $\pm g$. The integration of the energy delta-function $\delta(\Delta E)$ with respect to the y momentum components gives the Jacobian $\sqrt{pp_2p_3p_4}/|p|$, provided $pp_2p_3p_4 > 0$, and the large

logarithm $2 \log(1/g)$. Thus, in the large logarithm approximation:

$$\frac{1}{2\pi} \log \left(\frac{1}{g} \right) \gg 1 \quad (12)$$

the linearized Boltzmann kinetic equations reads:

$$\lambda \iint_{-\infty}^{+\infty} \frac{\chi(|p|) + \chi(|p'|) - \chi(|p+q|) - \chi(|p'-q|)}{(e^p + 1)(e^{p'} + 1)(e^{-p-q} + 1)(e^{-p'} + 1)} \times \frac{\sqrt{pp'(p+q)(p'-q)} dp' dq}{q^2} = \frac{-|p|}{(e^p + 1)(e^{-p} + 1)} \quad (13)$$

where $\lambda = 2Ng_R^2 \log(1/g)$ is the Coulomb integral, and the condition $pp'(p+q)(p'-q) > 0$ is enforced in the integrand. The exchange term vanishes in the leading logarithm approximation. The Debye screening mass makes the integral in Eq.(13) to converge as the principle value in the vicinity of $q = 0$. Due to few symmetries of the integral in Eq.(13): ($p \leftrightarrow p'$, $q \leftrightarrow -q$), ($p \leftrightarrow -p - q$ and $p' \leftrightarrow -p' + q$), the Eq.(13) is a symmetric operator. Thus, the Boltzmann kinetic equation is the variation of the functional: $\mathcal{R}[\chi] - \Sigma[\chi]$, where

$$\mathcal{R}[\chi] = \frac{\lambda}{8} \iiint_{-\infty}^{+\infty} \frac{\sqrt{pp'(p+q)(p'-q)} dp dp' dq}{q^2} \frac{1}{(2\pi)^2} \times \frac{(\chi(|p|) + \chi(|p'|) - \chi(|p+q|) - \chi(|p'-q|))^2}{(e^p + 1)(e^{p'} + 1)(e^{-p-q} + 1)(e^{-p'} + 1)} \quad (14)$$

$$\Sigma[\chi] = - \int \frac{p\chi(p)}{(e^p + 1)(e^{-p} + 1)} \frac{dp}{2\pi}$$

The existence of this positively defined functional $\mathcal{R}[\chi]$ proves that the conductivity is positive. Indeed, in the minimum: $\mathcal{R}[\chi] - \Sigma[\chi] < 0$ because $\mathcal{R}[0] - \Sigma[0] = 0$. The conductivity $\sigma = \Sigma[\chi] > \mathcal{R}[\chi] > 0$.

The equation (13) is contradictory and has no solution as the integral over all p applied to the left hand side is zero. It means that the leading large logarithm approximation is insufficient. However, from the all next order terms of the Boltzmann kinetic equation we need only their combined action on the homogeneous, in the momentum space, mode $\chi(\mathbf{p}) = \chi_0 = \text{const}$, which is being neglected by the leading term Eq.(13). The 'leading' order of this 'subleading' term is g_R^2 without the large logarithm. The mode χ_0 arises in the process of parallel shift of all momenta $|\mathbf{p} + \mathbf{a}| = |\mathbf{p} + (\mathbf{p} \cdot \mathbf{a})/\mathbf{p}|$. We write the 'subleading' term as a projection: $Ng_R^2 |\Phi(\mathbf{p})\rangle \langle \Phi(\mathbf{p})|$, onto a function $\Phi(\mathbf{p})$. The function $\Phi(\mathbf{p})$ can be found in the closed form as an integral. We parameterize the four momenta of the two scattering electrons by $p_i = \alpha_i |\mathbf{p}_i|$. These also define the mutual angles of the momenta upto to the four-fold discrete flip transformations. The collision integral gives the function $\Phi(\mathbf{p}) = St_\alpha(\mathbf{p})[\chi = 1]$, independent of α , and we set $\alpha = +1$. Its exchange part can be read off from Eq.(7,8) whereas the direct part allows for further transformation:

$$\Phi_d(p) = \iint_{-\infty}^{+\infty} \frac{1}{(e^p + 1)(e^{p_2} + 1)(e^{p_3} + 1)(e^{p_4} + 1)}$$

$$\left(\sqrt{us} - (2Q^2 - u - s) \operatorname{Arcth} \sqrt{\frac{u}{s}} + 2\sqrt{Q^2 - u} \sqrt{Q^2 - s} \operatorname{Arcth} \sqrt{\frac{(Q^2 - u)s}{(Q^2 - s)u}} \right) \frac{dp_2 dp_3}{2\pi u} \quad (15)$$

where $p_4 = -p - p_2 - p_3$ due to the energy conservation, and the parameterization: $Q = p + p_3 = -p_2 - p_4$, $u = 4pp_3$, $s = 4p_2p_4$ [with $Q^2 > |u|, |s|$] is used, satisfying the condition $us > 0$ in the Eq.(15). As long as $\Phi(p)$ is known and the homogeneous mode χ_0 is singled out: $\chi(\mathbf{p}) = \chi_0 + \chi_1(\mathbf{p})$, the Boltzmann kinetic equation reads:

$$Ng_R^2 \Phi(p) \chi_0 + \frac{\delta \mathcal{R}}{\delta \chi} [\chi_1] = -\frac{|p|}{(e^p + 1)(e^{-p} + 1)} \quad (16)$$

In the large logarithm limit Eq.(12), the function $\chi_1(\mathbf{p})$ is relatively small $|\chi_1(\mathbf{p})| \ll |\chi_0|$. Integrating Eq.(16) with respect to p eliminates all non-homogeneous modes in the kinetic equation that determines χ_1 and leaves the equation for χ_0 only:

$$Ng_R^2 C \chi_0 = \int_0^{+\infty} \frac{-|p| dp}{(e^p + 1)(e^{-p} + 1)} = -\log(2) \quad (17)$$

Solution is: $\chi_0 = -\log(2)/NCg^2$, where C is the average of the collision integral over the homogeneous mode:

$$C = \int_0^{+\infty} \Phi(p) dp \quad (18)$$

We estimate numerically the direct $C_d \approx 0.69$ and the exchange $C_{ex} \approx -0.1/N$ parts of $C = C_d + C_{ex}$. The distribution of excitations with the current is the same as without the current but translated in parallel in the momentum space by a vector proportional to the electric field. This solution, though, does not vanish in the momentum origin. We estimate numerically that a gap in the collision integral at the origin: $\Delta\delta(p)\delta(p')$, gives a necessary crossover of $\chi(\mathbf{p})$ to zero on the scale $|p| < g$, and as $g \rightarrow 0$ this feature has negligible effect.

The current Eq.(10) in this state is found neglecting small χ_1 contribution: $\vec{j}[\chi] = -N \log(2)(e^2/2\pi\hbar) \chi_0 \vec{E}$. Thus, the conductivity of the defectless graphene is:

$$\sigma = \frac{e^2 \log^2(2)/C}{\hbar 2\pi g_R^2} \quad (19)$$

in the limits of large dielectric constant of the substrate $g \rightarrow 0$ and, also, the large logarithm $\log(1/g)/(2\pi) \rightarrow \infty$. The conductivity Eq.(19) depends logarithmically on the effective temperature T^* . Inclusion of the specific mechanisms of energy relaxation due to say electron-phonon interaction is necessary to determine it. At any rate, T^* is small in the limit of weak electric field $T^* \sim E^\gamma$. However, the power γ can be as small as $\gamma = 1/3$ in the realistic electron-phonon models. For the Coulomb coupling $g_R \approx 0.35$ the conductivity Eq.(19) corresponds to the the experimental minimum conductivity around $\rho_{max} \approx 4$ kOhm. Numerical estimation of the Boltzmann kinetic equation shows that the large logarithm approximation begins to work at around $g_R < 0.2$ whereas for $g_R \sim 0.35$ an increase of the conductivity in Eq.(19) by 30% or so has to be expected.

As the gate voltage breaks the particle-hole symmetry and graphene accommodates a net charge: $e(N_h - N_e)$, the total momentum is no longer conserved: $d\vec{P}/dt = e(N_h - N_e)\vec{E}$. This runaway evolution of the excitation distribution can not be controlled by their mutual Coulomb interactions alone, as they conserve the momentum, and some defects violating the translational symmetry is required to stabilize the steady state.

In conclusion, the minimum conductivity of defectless graphene, Eq.(19), is found in the limit of weak Coulomb interaction. The result agrees with experiments on single layer graphene on the SiO₂ substrate and predicts a decrease of the minimum conductivity for graphene suspended in vacuum since g_R is larger.

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