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 of graphene with current flowing is assumed to be an almost 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 point.¹⁻³ Theory predicts a specific universal conductivity⁴ for this point, whereas the experimental conductivity exceeds this prediction by three times. However, an adequate understanding of the physical mechanisms at the compensation point will be important in the proposed future applications of graphene in electronic devices. Responsible for this discrepancy may be the concept of noninteracting quasiparticles, well established in normal Fermi liquids and assumed in Ref. 4 but questionable when applied to the graphene directly. The electronelectron interaction in normal Fermi liquids cannot change the current because of the Galilean symmetry, and the electron liquid flows as a whole. The rate of electron-electron scattering is also 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, neutral graphene is different. In its Brillouin zone there are two points where the electron dispersion acquires a conelike shape, the relativistic massless Dirac dispersion.⁵ This feature is simply understood by using the tight-binding model on the honeycomb lattice,⁶ which may represent the band structure of graphene. At the cone apexes the two crystal bands of graphene meet. The valence band is filled, whereas the conduction band is empty. At low-temperature electronic excitations of two types, particles and holes, proliferate in the vicinity of the two Dirac points. The special band structure of graphene makes the physics of one electron in graphene Lorentz invariant, with the role of the ultimate light velocity being played by the band cone angle. However, the entire graphene system, including the ions coupled by the instantaneous Coulomb interaction, is not Lorentz invariant. Thus, in neutral defectless graphene the current can relax via the Coulomb interaction alone. As the Fermi circle degenerates into two points, the rate of Coulomb scattering is no longer weak, $\sim T$.

An electric field has a different effect in zero-gap semiconductors than in Fermi liquids. In the Fermi liquid the total momentum of electrons increases in time as a product of the electric field and the total charge of electrons. However, in the neutral semiconductor, the total momentum of the valence and the conduction electrons remains zero in the electric field. This occurs due to exact compensation of the electric-field force and the force from the ions, which is conveniently described as a rate of umklapp momentum change in the valence electrons reaching the boundary of the Brillouin zone. All of these special features of graphene call for a study of the role of the Coulomb interaction on the evolution of graphene charge carriers. One important motivation for such a study is the recent explanation⁷ of a linear dependence of the conductivity as a function of the doping charge away from the compensation point³ using the theory of scattering on charged impurities. In this paper, the conductivity of defectless graphene is found at the compensation point for weak Coulomb interaction assuming that charge carriers constitute an almost ideal rare plasma of particles and holes.

The main precondition for nonconservation of the current is proximity to the compensation point. Noninteracting particles and holes separate in the electric field, although in 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, the precise value of the conductivity is determined by the Coulomb interaction. A microscopic process that changes the current is shown in Fig. 1. A pair consisting of particle 1 and hole 2 has zero net momentum and nonzero net current. The electron velocity in state 2 is opposite to the electron velocity in state 1. However, a hole is the absence of an electron, therefore the total current of the pair (1,2), is nonzero. The Coulomb interaction scatters the pair (1,2) into a new position (3,4)with the same total momentum and energy. And the net pair current in the new state (3,4) is reversed.



FIG. 1. (Color online) Scattering of the particle-hole pair (1,2) into the particle-hole pair (3,4), which conserves the momentum and the energy but changes the current. Excitation currents are shown by arrows. The *x* axis is the momentum and the *y* axis is the energy. The matrix element for this specific process is zero but variation of the momenta makes it nonvanishing.

Consider an infinite layer of perfect graphene on top of a dielectric substrate at zero temperature. Applying an electric field E will create particle and hole excitations due to the Schwinger mechanism. The work of E on these excitations is the Joule heat, which will induce lattice vibrations near the graphene layer and which will eventually escape into the bulk. In the end, a steady distribution of 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 part,⁵

$$\hat{H} = c(\hat{\tau}^{z}\hat{\alpha}^{x}p_{x} + \hat{\alpha}_{y}p_{y}), \qquad (1)$$

where $\mathbf{p} = (p_x, p_y)$ is momentum and $\hat{\alpha}^x, \hat{\alpha}^y, \hat{\tau}^z$ are the Pauli matrices, with the first two acting in the representation space of the crystal point group and the last one acting in the valley space. $c \approx 10^8$ cm/s is the characteristic band velocity that determines the cone angle.⁵ The total degeneracy of electronic states in graphene is N=4 due to the spin and the valley. The Hamiltonian (1) is diagonalized by a unitary transformation $(1 + \hat{\alpha}^y)\exp(i\hat{\tau}\hat{\alpha}^z\phi/2)/\sqrt{2}$ into two crystal bands and 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 electronic state of graphene is determined by a dimensionless Coulomb coupling,

$$g = \frac{e^2}{\kappa \hbar c},\tag{2}$$

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

$$\boldsymbol{\epsilon}(\mathbf{p}) = c_R |\mathbf{p}| \left[1 - gg_R \left(\frac{N}{24} - 0.013 \ 08 \right) \ln \ln \frac{Q}{|\mathbf{p}|} \right], \quad (3)$$

in the long-wavelength limit $|\mathbf{p}| \leq Q$, where Q is the size of the Brillouin zone. Although in three-dimensional (3D) zerogap semiconductors, the renormalization group describes nontrivial critical indices,⁸ in two-dimensional (2D) graphene it degenerates into a simple Hartree-Fock renormalization of the cone velocity and the Coulomb coupling,⁹

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

In addition to the nonequilibrium state of graphene with 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 the graphene states with the particle-hole coherence [see, e.g., Ref. 10], since it 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 part of momentum space. Hence, the distribution function for particles $F_+(\mathbf{p})$ is the same as the distribution function for holes, $1-F_-(\mathbf{p})$, or in general $1-F_{\alpha}(\mathbf{p})=F_{-\alpha}(\mathbf{p})$. In the equilibrium state of graphene, the electron distribution function,

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

nullifies the collision integral for any momentum scale $\langle p \rangle$, which is related to the effective temperature $T^* = c_R \langle p \rangle$. Here c_R , defined in Eq. (4), is evaluated at $|\mathbf{p}| = \langle p \rangle$. The equilibrium 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 static Boltzmann kinetic equation determines the distribution function, which balances two processes—the acceleration of the excitations in the electric field and their redistribution in the course of mutual collisions:¹¹

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

In the lowest second order of the Coulomb coupling g the collision integral reads [for brevity, α_3, α_4 of the outgoing electrons are inverted]

$$St_{\alpha_{1}}(\mathbf{p}_{1}) = \sum_{\alpha_{2}\alpha_{3}\alpha_{4}} \int \operatorname{Tr}_{\pi} \left[|V_{\alpha_{1}\alpha_{2}}^{\alpha_{3}\alpha_{4}}(\mathbf{p}_{1}\mathbf{p}_{2}\mathbf{p}_{3}\mathbf{p}_{4})|^{2} \right]$$
$$\times (2\pi) \delta \left[\sum_{i=1}^{4} \alpha_{i} \epsilon(\mathbf{p}_{i}) \right] \left[\prod_{i=1}^{4} F_{\alpha_{i}}(\mathbf{p}_{i}) - \prod_{i=1}^{4} F_{-\alpha_{i}}(\mathbf{p}_{i}) \right]$$
$$\times (2\pi)^{2} \delta(\mathbf{p}_{1} + \mathbf{p}_{2} - \mathbf{p}_{3} - \mathbf{p}_{4}) \frac{d^{2}\mathbf{p}_{2}d^{2}\mathbf{p}_{3}d^{2}\mathbf{p}_{4}}{(2\pi)^{6}}.$$
(7)

 τ indices run over N=4 spin-valley degeneracy space. 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 $\langle p \rangle \ll Q$ and the Debye screening radius is large $R_D \sim \hbar^2 c_R^2 / e^2 T^*$, the Coulomb matrix element is negligibly screened

$$V_{\alpha_{1}\alpha_{2}}^{\alpha_{3}\alpha_{4}}(\mathbf{p}_{1}\mathbf{p}_{2}\mathbf{p}_{3}\mathbf{p}_{4}) = \frac{1}{2} \left(\frac{2\pi e^{2}}{\kappa |\mathbf{p}_{1} - \mathbf{p}_{3}|} \frac{1 - z_{1}z_{3}^{*}}{2} \frac{1 - z_{2}z_{4}^{*}}{2} \delta_{\tau_{1}\tau_{3}} \delta_{\tau_{2}\tau_{4}} - \frac{2\pi e^{2}}{\kappa |\mathbf{p}_{2} - \mathbf{p}_{3}|} \frac{1 - z_{2}z_{3}^{*}}{2} \frac{1 - z_{1}z_{4}^{*}}{2} \delta_{\tau_{1}\tau_{4}} \delta_{\tau_{2}\tau_{3}} \right),$$
(8)

where the notation $z_i = \alpha_i (p_i^x + i p_i^y) / |\mathbf{p}_i|$ is used. The square of the matrix element [Eq. (8)] consists of two terms—the direct and the exchange terms. The exchange term vanishes when two scattering excitations have different spins or valleys.

In the state of graphene with current flowing, the general form for the electron distribution function is

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

where $\chi(\mathbf{p})$ is the perturbation due to the electric field. It likely has to satisfy the condition $\chi(\mathbf{p}) \rightarrow 0$ as $|\mathbf{p}| \rightarrow 0$. Any $\chi(\mathbf{p})$ in Eq. (9) explicitly conserves the number of electrons, their total energy, and their total momentum.¹¹ We linearize the Boltzmann kinetic Eq. (6) with respect to $\chi(\mathbf{p})$ (Ref. 11). The linearized collision integral becomes a matrix, which is symmetric due to a detailed balance of the direct and timereversed processes in the steady state. The linearized current in response to \vec{E} reads

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

where the band velocity $\vec{v}_{\alpha} = \alpha c_R \vec{p} / |\mathbf{p}|$. For exactly linear dispersion and for the collinear orientation of four vectors representing the momenta of two electrons before and after the collision, $\mathbf{p} || \mathbf{p}' || \mathbf{q}$, the change in the energy during the collision ΔE , given by the argument of the delta function in Eq. (7), is zero [many technical details of what follows are thoroughly discussed in Ref. 12], i.e., $\alpha_1 |\mathbf{p}| + \alpha_2 |\mathbf{p}'| + \alpha_3 |\mathbf{p}_3| + \alpha_4 |\mathbf{p}_4| = 0$ for any $|\mathbf{p}'|$ and $|\mathbf{q}|$ provided three conditions are met: $\alpha_1 = \alpha_2 \operatorname{sgn}(\mathbf{p} \cdot \mathbf{p}_2) = -\alpha_3 \operatorname{sgn}(\mathbf{p} \cdot \mathbf{p}_3) = -\alpha_4 \operatorname{sgn}(\mathbf{p} \cdot \mathbf{p}_4)$. Choosing the direction of the vector \mathbf{p} as x and expanding around the collinear configuration we find

$$\Delta E = c_R \left[\frac{p_y'^2}{2p'} - \frac{q_y^2}{2(p+q)} - \frac{(p_y' - q_y)^2}{2(p'-q)} + \frac{g_R}{4} \sum_i p_i \ln|p_i| \right],$$
(11)

where $p_i = p_{ix}$. The integration of the energy delta function $\delta(\Delta E)$ in the kinetic equation Eq. (7) with respect to the *y* components of all momenta gives the Jacobian $\sqrt{pp_2p_3p_4}/|p|$, provided $pp_2p_3p_4>0$, as well as the large logarithm $2 \ln(1/g_R)$, neglecting terms of the order of one. In this large logarithm approximation,

$$\frac{1}{2\pi} \ln\left(\frac{1}{g_R}\right) \gg 1, \qquad (12)$$

the exchange term vanishes and the linearized Boltzmann kinetic equation reads

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

where $\lambda = 2Ng_R^2 \ln(1/g_R)$ is the Coulomb integral, and the condition pp'(p+q)(p'-q) > 0 is enforced in the integrand. The Debye screening mass makes the integral in Eq. (13) converge as the principal value in the vicinity of q=0. Due to several symmetries of the integral in Eq. (13): $(p \leftrightarrow p', q \leftrightarrow -q), p \leftrightarrow -p-q$, and $p' \leftrightarrow -p'+q$, Equation

(13) is a symmetric operator. Thus, Eq. (13) is the variation of the functional, $\mathcal{R}[\chi] - \Sigma[\chi]$, where

$$\mathcal{R}[\chi] = \frac{\lambda}{8} \int \int \int_{-\infty}^{+\infty} \frac{\sqrt{pp'(p+q)(p'-q)}}{q^2} \frac{dpdp'dq}{(2\pi)^2} \\ \times \frac{[\chi(|p|) + \chi(|p'|) - \chi(|p+q|) - \chi(|p'-q|)]^2}{(e^p + 1)(e^{p'} + 1)(e^{-p-q} + 1)(e^{-p'+q} + 1)},$$

$$\Sigma[\chi] = -\int \frac{p\chi(p)}{(e^p + 1)(e^{-p} + 1)} \frac{dp}{2\pi}.$$
 (14)

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

Equation (13) is contradictory and has no solution since the integral over all p, applied to the left-hand side, is zero. It means that the leading large logarithm approximation is insufficient and we have to return to the Boltzmann kinetic Eq. (6). We let its solution be a sum of homogeneous and nonhomogeneous, in the momentum space, modes: $\chi(\mathbf{p}) = \chi_0$ $+\chi_1(\mathbf{p})$. For the linearized collision integral, the kinetic Eq. (6) reads

$$\frac{\delta \Sigma}{\delta \chi} = St_{\alpha}(\mathbf{p})[\chi(\mathbf{p})] = \frac{\delta \mathcal{R}}{\delta \chi}[\chi_1(\mathbf{p})] + \chi_0 St_{\alpha}(\mathbf{p})[1], \quad (15)$$

where $St_{\alpha}(\mathbf{p})[\chi_1(\mathbf{p})] \approx \delta \mathcal{R} / \delta \chi[\chi_1(\mathbf{p})]$ and $St_{\alpha}(\mathbf{p})[\chi_0] = \chi_0 St_{\alpha}(\mathbf{p})[1]$. We note that $St_{\alpha}(\mathbf{p})[1]$ is independent of α . We let a function $\Phi(\mathbf{p})$ be defined according to $Ng_R^2 \Phi(\mathbf{p}) = St_+(\mathbf{p})[1]$. It is obviously isotropic: $\Phi(\mathbf{p}) = \Phi(p)$. The leading order of the collision integral for the homogeneous mode is g_R^2 without the large logarithm since $\mathcal{R}[\chi_0]=0$. The homogeneous mode arises in the process of a parallel shift of all momenta $|\mathbf{p}+\mathbf{a}|=\mathbf{p}+(\mathbf{p}\cdot\mathbf{a})/|\mathbf{p}|$. In the large logarithm limit [Eq. (12)], the nonhomogeneous mode is relatively small $|\chi_1(\mathbf{p})| \ll |\chi_0|$. Integrating Eq. (15) with respect to p eliminates the nonhomogeneous term $\delta \mathcal{R} / \delta \chi[\chi_1(\mathbf{p})]$ and leaves the linear kinetic equation for the homogeneous mode χ_0 only,

$$Ng_R^2 C\chi_0 = \int_0^{+\infty} \frac{-|p|dp}{(e^p + 1)(e^{-p} + 1)} = -\ln(2), \qquad (16)$$

with the solution $\chi_0 = -\ln(2)/NCg_R^2$. Here,

$$C = \int_0^{+\infty} \Phi(p) dp \tag{17}$$

is the momentum average of the collision integral with the weight 1/p.

The function $\Phi(p)$ can be found in the closed form as an integral. We parametrize the four momentum vectors of scattering electrons by their amplitudes $p_i = \alpha_i |\mathbf{p}_i|$ as well as the amplitude of the transferred momentum q. In this parametrization, the mutual angles between the vectors \mathbf{p}_i are defined up to the fourfold discrete flip transformation. The exchange

part of $\Phi(\mathbf{p})$ can be read off from Eqs. (7) and (8), whereas the direct part allows for further integration over q:

$$\Phi_{d}(p) = \int \int_{-\infty}^{+\infty} \frac{1}{(e^{p}+1)(e^{p_{2}}+1)(e^{p_{3}}+1)(e^{p_{4}}+1)} \\ \times \left[\sqrt{us} - (2Q^{2}-u-s)\operatorname{Arcth} \sqrt{\frac{u}{s}} \right] \\ + 2\sqrt{Q^{2}-u}\sqrt{Q^{2}-s} \operatorname{Arcth} \sqrt{\frac{(Q^{2}-u)s}{(Q^{2}-s)u}} \frac{dp_{2}dp_{3}}{2\pi u},$$
(18)

where $p_4 = -p - p_2 - p_3$ is fixed due to energy conservation, and the parametrization $Q = p + p_3 = -p_2 - p_4$, $u = 4pp_3$, and $s = 4p_2p_4$ [with $Q^2 > |u|, |s|$] is used, satisfying the condition us > 0 in Eq. (18).

We estimate numerically both the direct $C_d \approx 0.689$ and the exchange $C_{ex} \approx -0.24/N$ parts of $C = C_d + C_{ex}$. The distribution of excitations in the state with current flowing is the same as that without current but translated in parallel in momentum space by a vector proportional to the electric field. It does not vanish at the momentum origin, though. However, our numerical diagonalization of the operator \mathcal{R} reveals soft modes with the eigenfunctions $\chi_1(\mathbf{p})$ being localized at $|\mathbf{p}| < g_R$. These soft modes can produce a crossover of the solution to zero at $\mathbf{p}=0$ and, in the limit $g_R \rightarrow 0$, the net effect of neglecting this crossover becomes negligible.

The current in Eq. (10) is found by neglecting the small $\chi_1 \mod \vec{j}[\chi_0] = -N \ln(2)(e^2/2\pi\hbar)\chi_0\vec{E}$. Thus finally, the conductivity of defectless graphene is found:

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

in the limit of a large dielectric constant of the substrate, $g_R \rightarrow 0$, and in the limit of large logarithm $\ln(1/g_R)/(2\pi)$ $\rightarrow \infty$. The conductivity Eq. (19) depends logarithmically on the effective temperature T^* . To determine it, inclusion of specific mechanisms of energy relaxation due to, for example, the electron-phonon interaction is necessary. 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 realistic electron-phonon models. For the Coulomb coupling g_R ≈ 0.35 the conductivity Eq. (19) corresponds to the experimental minimum conductivity around $\rho_{max} \approx 4$ k Ohm. Numerical estimation of the kinetic equation shows that the large logarithm approximation begins to suppress the nonhomogeneous mode χ_1 at around $g_R < 0.2$, whereas for g_R ~ 0.35 an increase in the conductivity in Eq. (19) by 30% or so is expected.

As the gate voltage breaks the particle-hole symmetry and the graphene accumulates 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 cannot be controlled by the Coulomb interaction alone because it conserves the total momentum. Hence, some defects violating the translational symmetry are 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 in the minimum conductivity for graphene suspended in vacuum since g_R is larger. The same result is reported in Ref. 13.

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