



Electronic Contribution to Conductivity in Graphene

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The exceptional transport characteristics, coupled with high thermal, mechanical and chemical stability, provide wide opportunities for practical application of graphene. Temperature dependence of graphene electrical conductivity is hereby analyzed in wide temperature range of 15–400 K, by solving semiclassical Boltzmann equation in the approximation of relaxation time. Basic relaxation mechanisms in graphene—electron scattering on impurities and electron–phonon interaction—are accounted with corresponding relaxation times introduced phenomenologically. The received results are compared with the experimental data from the referent sources.

Keywords: Graphene, Electrical Conductivity, Boltzmann Equation, Times Relaxation.

1. INTRODUCTION

In the past several decades much attention has been devoted to electronic properties of low-dimensional systems in the condensed matter physics. Interest in such systems arises mainly due to the fact that completely new quantum effects appear with decrease of the system dimensionality. Quantum properties of such systems are standardly described by Schrödinger equation of charged quasiparticles behaving nonrelativistically with finite effective mass. However, significant interest recently appeared for quite different class of systems with zero effective mass charged quasiparticles described by Dirac equation. The first representative of such two-dimensional (2D) systems is graphene, representing one layer of carbon atoms ordered within hexagone lattice.^{1,2}

More than 70 years ago, Landau and Pailerls showed that strictly 2D crystals could not exist as thermodynamically unstable. Their theory implies that divergent contribution to thermalfluctuations in low-dimensional crystal lattices gives rise to atomic displacements comparative with interatomic distances at any finite temperature. Fortunately, there is a way to escape this problem. Interactions with 3D structures stabilize 2D crystals during their growths. Therefore it is possible to obtain 2D crystalsintercalated in between or formed on top of atomic layers of corresponding 3D crystals. In such form graphene does exist within graphite, which can be considered built of graphene layers interacting via van der Waals forces. A group from the University

of Manchester, lead by Geim i Novoselov, succeeded in obtaining the graphene in 2004, thus starting revolution in this field. Their group also obtained 2D-crystals of some other materials: boronitride, dichalcogenide, and high-temperature superconductor Bi–Sr–Ca–Cu–O. Their choice of convenient substrate was silicon-dioxide, material widely used in semiconductor industry.²

Shortly after obtaining first samples of graphene, numerous unique electrical, chemical and mechanical characteristics of graphene were discovered, like unusual half-integral quantum Hall effect and Barry phase π , which undoubtedly demonstrate existence of Dirac fermions in graphene and distinguish it in respect to usual 2D electronic systems with finite effective mass of charge carriers. It appears that its charge carriers have additional internal degree of freedom similar to chirality of ultra-relativistic elementary particles. In this way graphene became an unexpected link between condensed matter physics and quantum electrodynamics. Its existence now enables some quantum-relativistic phenomena, otherwise not observable in high-energy experimental physics, to be simulated and tested in modest experimental environments.^{1–3}

2. GRAPHENE CRYSTAL STRUCTURE AND DISPERSION LAW OF QUASIPARTICLES

Carbon atoms in graphene crystal lattice form hexagone structure. This lattice can be considered as superposition of two triangle lattices A and B, with unit cell out of only two atoms.

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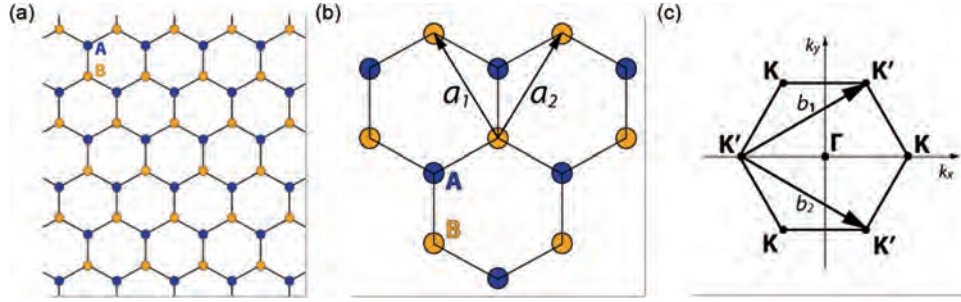


Fig. 1. Graphene crystal lattice consisting of two sublattices A and B (a), unit cell vectors (b), and unit vectors in inverse k space (c) (adopted from Ref. [4]).

The parameters of graphene crystal structure are unit vectors \vec{a}_1 i \vec{a}_2 of hexagone lattice, presented in Figure 1(b). They interconnect two adjacent atoms of the same type and their length is equal to lattice constant a . They can be represented as:

$$\vec{a}_1 = a \left(-\frac{1}{2}, \frac{\sqrt{3}}{2} \right), \quad \vec{a}_2 = a \left(\frac{1}{2}, \frac{\sqrt{3}}{2} \right) \quad (1)$$

The graphene Brillouin zone is presented in Figure 1(c). Unit vectors in inverse k space are:

$$\vec{b}_1 = \frac{2\pi}{a} \left(1, \frac{1}{\sqrt{3}} \right), \quad \vec{b}_2 = \frac{2\pi}{a} \left(1, -\frac{1}{\sqrt{3}} \right) \quad (2)$$

All hexagone nodes of Brillouin zone represent Dirac points. Although there are six of them, only two Dirac points are independent. In Figure 2, two groups of Dirac valleys are depicted by red and blue colours. The valleys belonging to the same group are on the mutual distance of reciprocal vector \vec{b} and represent the same state. It could be imagined that inner space around every hexagone node represents one third of the Dirac valley. By connecting these thirds, two complete Dirac valleys can be obtained, as presented in the right part of Figure 2. These two Dirac points lay on the k_x axes and have coordinates:

$$K = \frac{4\pi}{3a} (1, 0), \quad K' = \frac{4\pi}{3a} (-1, 0) \quad (3)$$

Carbon atoms in graphene hexagone plane are interconnected by strong covalent σ -bonds obtained by sp^2 hybridization of $2s$, $2p_x$ i $2p_y$ valence atomic orbitals, while the fourth $2p_z$ valence orbitals are normal onto graphen plane, they mutually weakly overlap, and are responsible for creating weak π bonds (Fig. 3). The fourth valence electrons represented by $2p_z$ atomic orbitals can hop from one carbon atom to another, making half-filled conducting π -zone responsible for electrical properties of graphene.

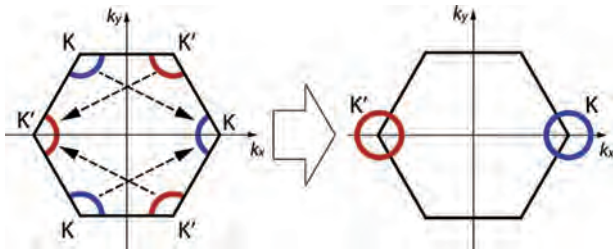


Fig. 2. Position of Dirac points in inverse k space (adopted from Ref. [4]).

These facts enable succesful application of the model of strong coupling for finding graphene electronic structure:⁵

$$H_0 \Psi(\vec{r}) = \varepsilon \Psi(\vec{r}) \quad (4)$$

$$H_0 = -i\hbar v_F \begin{pmatrix} \sigma \nabla & 0 \\ 0 & \sigma^* \nabla \end{pmatrix} \quad (5)$$

$$= -i\hbar v_F \begin{pmatrix} 0 & \nabla_x - i\nabla_y & 0 & 0 \\ \nabla_x + i\nabla_y & 0 & 0 & 0 \\ 0 & 0 & 0 & \nabla_x + i\nabla_y \\ 0 & 0 & \nabla_x - i\nabla_y & 0 \end{pmatrix}$$

$$\Psi(\vec{r}) = \frac{1}{L} \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i\theta_k} \\ s \end{pmatrix} e^{i\vec{k}\cdot\vec{r}}; \quad k_x = k \cos \theta_k; \quad (6)$$

$$k_y = k \sin \theta_k; \quad k = \sqrt{k_x^2 + k_y^2}$$

where L^2 is graphene surface, and $s = +1$ for conducting zone and $s = -1$ for valence zone.⁶ The graphene dispersion low is:^{1,7}

$$E = \pm \hbar v_F \sqrt{1 + 4 \cos^2 \pi k_y a + 4 \cos \pi k_y a \cos \pi k_x a \sqrt{3}} \quad (7)$$

where signs plus and minus correspond to conducting and valence zones, respectively. The both zones are degenerated in Dirac point.

The Fermi level is passing through Dirac point (Fig. 4(a)), so there can be said that graphene is semiconductor without energy gap or semimetal. In case we take into account only area in the vicinity of Dirac points K and K' , dispersion relation can be significantly simplified. It obtains linear dependence:

$$E = \hbar v_F k \quad (8)$$

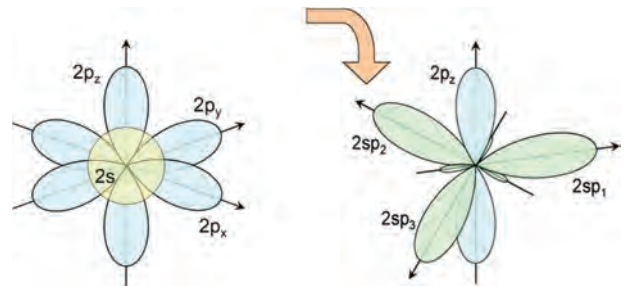


Fig. 3. Atomic orbitals in graphene (adopted from Ref. [5]).

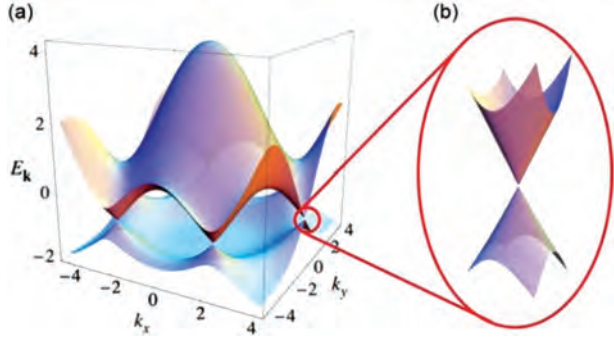


Fig. 4. The graphene dispersion relation (a), and its view in the vicinity of Diracpoint (b) (adopted from Ref. [1]).

where v_F is Fermi velocity (with experimentally obtained value of 10^6 m/s), k is module of wave vector in 2D-space (with components (k_x, k_y)), calculated in K or K' Diracpoints, and \hbar is Dirac constant.^{1,6,8} The zone has the shape of a cone (Fig. 4(b)). Photon also has a similar spectrum and it is therefore said that quasiparticles in graphene (electrons or holes) have zero effective mass. As electrons and holes are fermions, their movement is described by Dirac equation, but with zero mass of particles and antiparticles (similarly to equation for massless neutrino).^{2,5}

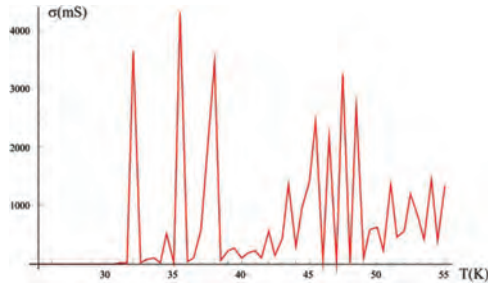
Linear dispersion law is most important, if not unique, characteristic of quantum transport described by Dirac equation. For positive energies (above Dirac point) charge carriers are similar to electrons and are negatively charged. For negative energies, if valence zone is not filled, quasiparticles behave as positively charged holes analogous to positrons. It should be noted that electrons and holes in most solid state materials are usually described by independent Schrödinger equations with corresponding effective masses. On the contrary, electrons and holes in graphene are mutually interconnected, demonstrating property of charge conjugation symmetry.

Quasi particle density of states in graphene is obtained as a number of states per unit energy and per unit surface^{2,9}

$$D(\varepsilon) = g_s g_v \frac{1}{(2\pi)^2} 2\pi k \frac{dk}{d\varepsilon} = g_s g_v \frac{1}{(2\pi)^2} 2\pi k \frac{1}{d\varepsilon/dk} \quad (9)$$

where $g_s = 2$ is degree of spin degeneracy, and $g_v = 2$ degree of valence electrons degeneracy. Finally, quasi particle density of states per unit surface is obtained as follows:

$$D(\varepsilon) = g_s g_v \frac{\varepsilon}{2\pi \hbar^2 v_F^2} = 4 \frac{\varepsilon}{2\pi \hbar^2 v_F^2} \quad (10)$$



3. GRAPHENE ELECTRICAL CONDUCTIVITY

The main limitation of charge carriers mobility in graphene is posed by charged impurities in SiO_2 substrate and by the existing phonon subsystem. The related relaxation mechanisms provide finite lower value for graphene electrical conductivity. Of special interest is finite conductivity in Dirac point. Based on semimetal nature of graphene, the simple single-particle theory predicts either vanishing or infinite electrical conductivity: zero density of state in Dirac point implies vanishing conductivity, while absence of energy gap suggests infinite conductivity. However, it was experimentally obtained that graphene electrical conductivity is finite: $\sigma^{ex} \approx \eta 4e^2/h^2$.¹⁰ On the other hand, more complex theories can predict finite value σ^{th} , which is not in agreement with the experimental one: $\sigma^{th} = \sigma^{ex}/\pi$ (“the mystery of the missing π ”).

We shall analyze electrical conductivity of graphene in the vicinity of Dirac points for linear dispersion law. Our approach hereby applied is semiclassical, via solving Boltzmann transport equation, which is justified when notion of particle trajectory is plausible i.e., when de Broglie wave length of charge carriers is much less than their mean free path, $\lambda \ll \bar{l} = \bar{v}\tau$. When perturbation force originates from electrical field, the estimations show that Boltzmann transport equation is applicable till very strong electrical fields.¹¹ It is also shown in practice that application of Boltzmann transport equation provides results in good agreement with experimental data for high surface densities of charge carriers i.e., $n \geq 10^{12} \text{ cm}^{-2}$.¹² We hereby propose that these conditions are fulfilled.

Further on, for finding graphene electrical conductivity we shall apply semiclassical Boltzmann transport equation in the approximation of relaxation time:¹³

$$\frac{\partial f}{\partial t} + \frac{1}{\hbar} \frac{\partial f}{\partial \vec{k}} \cdot \vec{F} + \frac{\partial f}{\partial \vec{r}} \cdot \vec{v} = -\frac{g_{\vec{k}}}{\tau_{\vec{k}}} \quad (11)$$

where $f(t, \vec{r}, \vec{k})$ is distribution function, $g_{\vec{k}}$ is small discrepancy from equilibrium distribution, and $\tau_{\vec{k}}$ is relaxation time. After linearization of Eq. (11), one obtains

$$\frac{\partial g_{\vec{k}}}{\partial t} + \frac{\partial f_0}{\partial \vec{r}} \cdot \vec{F} + \frac{\partial g_{\vec{k}}}{\partial \vec{r}} \cdot \vec{v} = -\frac{g_{\vec{k}}}{\tau_{\vec{k}}} \quad (12)$$

where $f_0(t, \vec{r}, \vec{k})$ is equilibrium distribution function. By proposing that transport process of charge carriers is stationary and that distribution function does not depend on coordinates, as well as that charge transport is achieved under the influence of

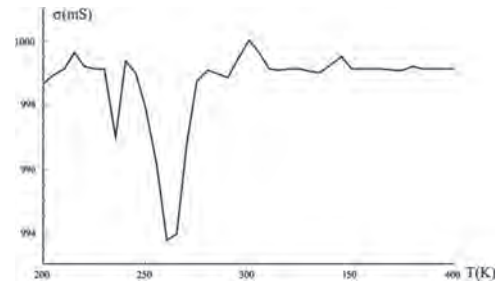


Fig. 5. Temperature dependance of graphene electrical conductivity in case of relaxation on charged impurities, for temperature ranges $T \ll T_{BG}$ (left) and $T > T_{BG}$ (right).

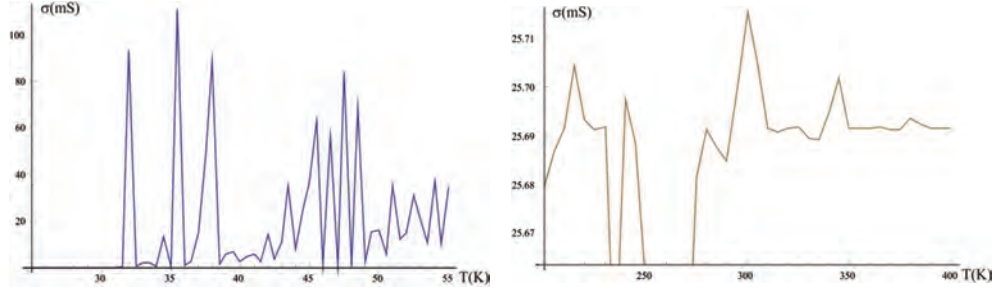


Fig. 6. Temperature dependence of graphene electrical conductivity in case of relaxation on neutral impurities, for temperature ranges $T \ll T_{BG}$ (left) and $T > T_{BG}$ (right).

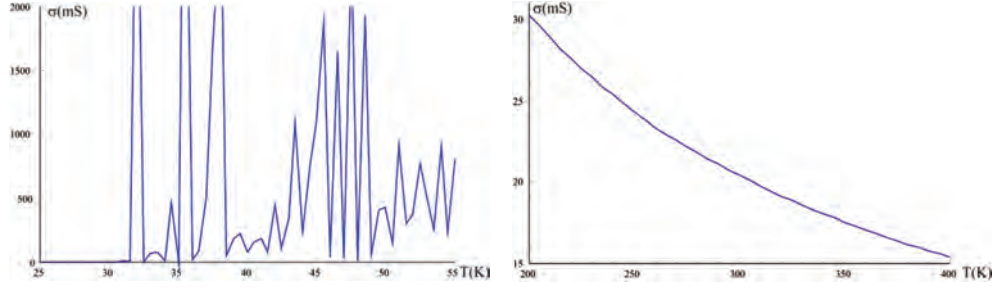


Fig. 7. Temperature dependence of graphene electrical conductivity in case of relaxation on both charged impurities and phonons, for temperature ranges $T \ll T_{BG}$ (left) and $T > T_{BG}$ (right).

homogenous electrical field directed along X -axes, $\vec{F} = -e\vec{E}$, Eq. (12) is transformed into:

$$\vec{F} \cdot \frac{1}{\hbar} \frac{\partial f_0}{\partial \vec{k}} = -\frac{g_{\vec{k}}}{\tau_{\vec{k}}} \quad (13)$$

By convenient transformations relation for small perturbation is obtained:

$$\frac{1}{\hbar} \frac{\partial f_0}{\partial \vec{k}} = \frac{1}{\hbar} \frac{\partial f_0}{\partial \varepsilon} \frac{\partial \varepsilon}{\partial \vec{k}} = \frac{\partial f_0}{\partial \varepsilon} \vec{v} \quad (14)$$

$$g_{\vec{k}} = e\tau_{\vec{k}} \vec{v} \cdot \vec{E} \left(-\frac{\partial f_0}{\partial \varepsilon} \right) \quad (15)$$

The current density is found via expression^{9,14}

$$J = \frac{g_s g_v}{A} \sum_k e v g_k \quad (16)$$

By inserting expression for perturbation (15) into expression for current density (16), on the basis of $J = \sigma E$ one obtains expression for electrical conductivity:

$$J = \frac{g_s g_v}{A} \sum_k e v g_k = \left\{ \frac{1}{A} \frac{A}{(2\pi)^2} g_s g_v \int_0^{2\pi} d\theta \int_0^\infty k dk e^2 v_x^2 \tau \left(-\frac{\partial f_0}{\partial \varepsilon} \right) E \right\};$$

$$v_x = v \cos \theta \quad (17)$$

$$\sigma = \frac{1}{(2\pi)^2} g_s g_v \int_0^{2\pi} \cos^2 \theta d\theta \int_0^\infty k dk e^2 v^2 \tau_k \left(-\frac{\partial f_0}{\partial \varepsilon} \right) \quad (18)$$

By applying transformation

$$k dk \rightarrow \frac{\varepsilon d\varepsilon}{(\hbar v_F)^2}$$

we get convenient expression for electrical conductivity of graphene:

$$\sigma = \frac{e^2}{2} \int_0^\infty \frac{g_s g_v}{2\pi (\hbar v_F)^2} v^2 \tau(\varepsilon) \left(-\frac{\partial f_0}{\partial \varepsilon} \right) d\varepsilon = \frac{e^2}{2} \int_0^\infty D(\varepsilon) v^2 \tau(\varepsilon) \left(-\frac{\partial f_0}{\partial \varepsilon} \right) d\varepsilon \quad (19)$$

To calculate total relaxation time we apply Matthiessen rule:

$$\frac{1}{\tau} = \sum_i \frac{1}{\tau_i} \quad (20)$$

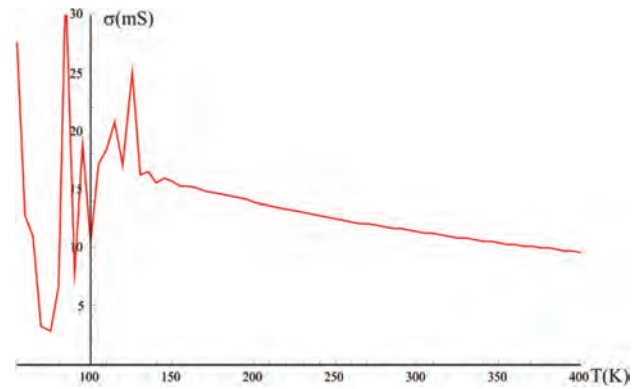


Fig. 8. Dependency of electrical conductivity on temperature in a wider temperature interval in cases when all observed relaxation mechanisms are at work.

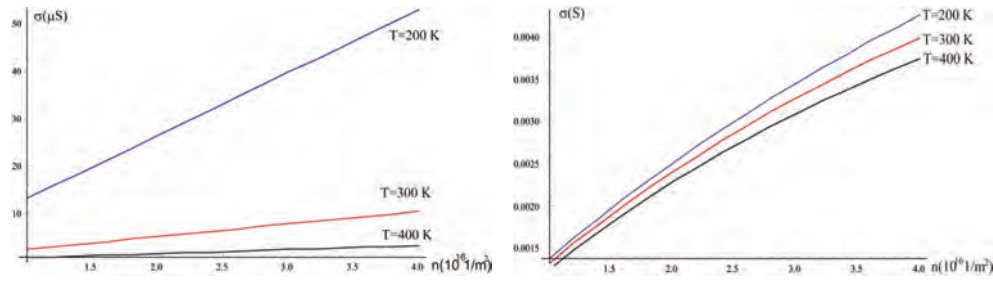


Fig. 9. Dependency of electrical conductivity on the concentration of charge carriers for temperatures lower than T_{BG} (left) and temperatures higher than T_{BG} (right).

In particular, for relaxation on impurities and phonons,¹³

$$\frac{1}{\tau} = \frac{1}{\tau_{im}} + \frac{1}{\tau_{ph}} \quad (21)$$

In case of relaxation on charged impurities (long-range Coulomb interaction), relaxation time is:^{7,14}

$$\frac{1}{\tau_{im}} = \frac{u_0^2}{\hbar \varepsilon_F}; \quad u_0 = \frac{\sqrt{n_i^c} Z e^2}{4 \tilde{\varepsilon}_0 \tilde{\varepsilon}_r (1 + \tilde{\gamma})}; \quad \gamma = \tilde{\gamma} k_F \quad (22)$$

where ε_F is graphene Fermi energy, $\tilde{\varepsilon}_0 \tilde{\varepsilon}_r$ is product of dielectric constant of vacuum and relative dielectric constant of substrate, n_i^c is concentration of charged impurities, and γ is wave vector.

In case of relaxation on neutral impurities (short-range Coulomb interaction), relaxation time is:^{1,6,8}

$$\frac{1}{\tau_{im}} = \frac{n_d V_0^2}{\hbar} \frac{\varepsilon_F}{4(\hbar v_F)^2} \quad (23)$$

where n_d is concentration of neutral impurities, and V_0 is applied contact voltage. The collision potential in this case has a form $V(\vec{r}) = V_0 \delta(\vec{r})$, where $\delta(\vec{r})$ is Dirac function.

In case of relaxation on phonons, we analyze the temperature ranges when $T \geq T_{BG}$ and when $T < T_{BG}$, where T_{BG} is Bloch-Grüneisen temperature, being the temperature limit below which phonon gas is degenerate while above which is non-degenerate.¹⁵

In case of $T \geq T_{BG}$ inverse relaxation time is^{14,15}

$$\frac{1}{\tau_{ph}(\varepsilon)} = \frac{1}{\hbar^3} \frac{\varepsilon}{4v_F^2} \frac{D^2}{\rho_m v_{ph}^2} k_B T \quad (24)$$

while in case of $T < T_{BG}$ ^{14,15}

$$\frac{1}{\tau_{ph}} = \frac{1}{\pi} \frac{1}{\varepsilon_F} \frac{1}{k_F} \frac{1}{2\rho_m v_{ph}} \frac{D^2}{(\hbar v_{ph})^4} \frac{4! \zeta(4)}{(k_B T)^4} \quad (25)$$

In expressions (24–25) D is deformation potential of impurity, ρ_m is graphene surface density, ε_F is graphene Fermi energy, k_F is graphene Fermi wave vector, v_{ph} is graphene phonon velocity, and $\zeta(4)$ is Zeta function.

For numerical calculation of electrical conductivity from Eqs. (19) and (20)–(25), we shall adopt the following values of parameters:¹⁴

$$\begin{aligned} D &= 30.4 \cdot 10^{-19} \text{ J}; & \rho_m &= 7.6 \cdot 10^{-7} \text{ kg/m}^2; & v_{ph} &= 2 \cdot 10^4 \text{ m/s}; \\ \varepsilon_F &= 4.168 \cdot 10^{-19} \text{ J}; & v_F &= 10^6 \text{ m/s}; & k_F &= 4.8 \cdot 10^8 \text{ m}^{-1}; \\ n_i^c &= 4 \cdot 10^{15} \text{ m}^{-2}; & Z &= 1; & \tilde{\varepsilon}_r &= 2.4; & \tilde{\gamma} &= 4.2; \\ n_d &= 0.4 \cdot 10^{14} \text{ m}^{-2}; & V_0 &= 16 \cdot 10^{-37} \text{ Jm}^2; & T_{BG} &= 54 \text{ K} \end{aligned} \quad (26)$$

For lower temperatures, Eq. (19) is transformed into^{14,16,17}

$$\begin{aligned} \sigma &= \frac{4\pi e^2}{h^2} \int_0^\infty dx \left\{ k_B T x + \varepsilon_F \left[1 - \frac{\pi^2}{6} \left(\frac{k_B T}{\varepsilon_F} \right)^2 \right] \right\} \tau(x) \frac{e^x}{(e^x + 1)^2}; \\ x &= \frac{\varepsilon - \mu}{k_B T}; \quad \mu = \varepsilon_F - \frac{\pi^2 (k_B T)^2}{6 \varepsilon_F} \end{aligned} \quad (27)$$

while for higher temperatures, it becomes

$$\begin{aligned} \sigma &= \frac{4\pi e^2}{h^2} \int_0^\infty dx \left\{ k_B T x + \frac{1}{4 \ln 2} \frac{\varepsilon_F^2}{k_B T} \right\} \tau(x) \frac{e^x}{(e^x + 1)^2}; \\ x &= \frac{\varepsilon - \mu}{k_B T}; \quad \mu = \frac{1}{4 \ln 2} \frac{\varepsilon_F^2}{k_B T} \end{aligned} \quad (28)$$

When taking into account only relaxation on charged impurities for temperatures $T \ll T_{BG}$, numerical calculation of Eq. (27) results in the graph of Figure 5 (left), while for temperatures $T > T_{BG}$ numerical calculation of Eq. (28) results in the graph of Figure 5 (right).

In case of relaxation on neutral impurities, we obtain the graphs in Figure 6.

When taking into account both relaxation on charged impurities and on phonons, using relaxation times given by Matthiessen rule, numerical calculation of Equations (27) and (28) results in the graphs of Figure 7.

Figure 8 shows that at lower temperatures, electrical conductivity oscillates substantially, while at the higher (above 150 K) monotonously decreases with temperature.

Electrical conductivity depends on the concentration of charge carriers via Fermi energy $\varepsilon_F = \hbar v_F \sqrt{\pi n}$. For practical reasons, it is necessary to analyze the dependence of electrical conductivity on concentration of charge carriers at different temperatures. Results are shown in Figure 9.

Figure 9 shows that with the increase in temperature, the electrical conductivity of graphenes, in both temperature areas, rises more slowly.

If dependency of chemical potential on temperature is used for the calculation of the value of electrical conductivity, the results differ slightly from the cases when the chemical potential is constant.

4. CONCLUSION

Graphene research is the most propulsive field in solid state physics and materials science.^{19–29} Graphene is continuing to surprise us with its properties, like extraordinary mechanical

strength and stretching, exceptional electrical and thermal conductivities, optical transparency etc. What is most attractive in graphene is its unusual system of charge carriers and unique transport properties. By new methods of obtaining more perfect graphene structures (encapsulation of graphene between boron-nitride crystallites)³ mobility of charge carriers and hence transport properties can be improved.

Graphene is the first and most prominent representative of the new class of materials—2D crystals. It reveals the new paradigm—relativistic solid state physics. The uniqueness of its electronic properties are charge carriers described not by Schrödinger equation but by Dirac-Weil equation similarly to massless fermions. Graphene represents serious candidate to replace silicon in microelectronics. The first realized prototypes of future devices based on graphene should be mentioned: field effect transistors and ballistic transistors at room temperature, extremely sensitive gas sensors, single-electron transistors, liquid-crystal displays and solar batteries with one graphene electrode, spin transistors etc.² Therefore, exploration of graphene properties, and especially transport ones, is fully justified and most important.

In this paper electrical conductivity of graphene in the vicinity of Dirac points for linear dispersion law is analyzed. Our approach is semiclassical, by solving Boltzmann transport equation in approximation of relaxation time. The various relaxation mechanisms of charge carriers in graphene are analyzed: on charged impurities, on neutral impurities, and on phonons.

For relaxation times adopted from the referent sources, the dependence of electrical conductivity of graphene was found numerically depending on the temperature and concentration of charge carriers.

Finally, it should be noted that impurities in graphene originate from its substrate (mostly SiO₂ or SiC). For graphene samples abundant in impurities there is a minimal electrical conductivity, while in pristine samples electrical conductivity is much larger, which is the case when graphene layer is realized not on substrate but in some other way.³ The main relaxation centers in graphene are charged impurities which create long-range Coulomb potentials. Local defects (vacancies, dislocations) contribute to the same dependence of relaxation time versus wave vector \vec{k} as charged impurities do. Even more, these local defects provide final densities of states of charge carriers in Dirac points, which must be taken into account in analysis of lower bound of graphene electrical conductivity. Namely, the simplified single-particle theory predicts that graphene electrical conductivity linearly decreases with decrease of charge carriers concentration i.e., when $n < n_i$ (where n_i is concentration of impurities), so in Dirac points graphene electrical conductivity should vanish. However, the experiment and more complex theories demonstrate

that graphene electrical conductivity has some minimal value of the order e^2/h ,¹⁸ showing that for low concentrations of charge carriers semiclassical Boltzmann approach hereby applied is not valid.

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