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Temperature- and thickness-dependent electrical conductivity of few-layer graphene and graphene nanosheets

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ABSTRACT

We established a calculation model of the conductivity of multilayer graphene based on Boltzmann transport equation and 2D electron gas theory. Numerical simulations show that the conductivities of few-layer graphene and graphene nanosheets are reduced when thickness is increased. The reduction rate decreases for micron-range thicknesses and remains constant thereafter. Moreover, the conductivity increases with the increase in temperature, in which the increase rate declines as temperature increases. Higher thickness exhibits a more obvious temperature effect on conductivity. Such effect also increases with the increase in temperature.

1. Introduction

Single-layer graphene (SLG), few-layer graphene (FLG), graphene nanosheets (GNs), graphene nanoribbons (GNRs), and carbon nanotubes have attracted considerable interest because of their unique electrical, thermal, optical, and mechanical properties [1–6]. FLG and GNs are widely applied in nanoelectronics [7–9], supercapacitors [10–12], transparent conducting thin films [13–15], solar energy fuel cells [16–18], biochemical analysis and detection [19,20], and electromagnetic interference shielding and microwave absorption [21,22].

In particular, the conductivities of SLG and multilayer graphene have attracted research interest. Novoselov et al. [23] found that the electrical resistivity of graphene is approximately 1.0 μΩ cm, which is slightly lower than that of silver (1.5 μΩ cm). Experimental data [1,23–25] from 2005 to 2007 show that the electron mobility of graphene ranges from 3×10^3 to 2.7×10^4 $cm^2\,V^{-1}\,s^{-1}$, which is lower than the forecast because of various impurities and defects in the samples. Bolotin et al. [26,27] demonstrated that the highest migration rate among five graphene samples is 2.3×10^5 $cm^2\,V^{-1}\,s^{-1}$. Geim et al. [28] revealed that the mobility of graphene is more than 2.0×10^5 $cm^2\,V^{-1}\,s^{-1}$, which almost remains constant at different temperatures. Romanenko et al. [29] used multilayer graphene with graphite flake-like structures (layer number, 5 to 60; thickness, 2 nm to 18 nm) and found that their electrical conductivity increases with the increase in temperature, implying a negative temperature coefficient of resistance. Boland et al. [30] demonstrated that the electrical conductivity of GNRs decreases with increasing thickness; a similar behavior is observed for graphite. Rouhi et al. [5] found that the electrical resistivity of suspended FLG is approximately 3.35 μΩ/cm. Hwang and Das Sarma [31] theoretically calculated the temperature-dependent electrical conductivity of graphene using the Boltzmann transport equation and the mechanism of phonon scattering. They showed that the temperature coefficient of resistance increases with the increase in temperature from 5 to 500 K. The calculations of Hwang and Das Sarma [32] indicated that graphene exhibits positive and negative temperature coefficients of resistance at temperatures less than and greater than the Fermi temperature, respectively. Perebeinos and Avouris [33] simultaneously considered the scattering of phonons, surface polariton phonons, and charged impurities; they found that the mobility of SLG that is grown on the insulating substrate slightly decreases with increasing temperature. Although high conductivity of SLG can be explained by Boltzmann transport theory, this theory fails to explain the two problems in FLG and GNs. First, the reduction of the mobility of multilayer graphene with increasing temperature; the positive temperature coefficient of resistance results in electronic and optical phonon scattering, which is not observed in [29]. Second, the reduction in graphene

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conductivity with increasing thickness of the graphene cannot be explained by this theory [30].

In this study, two types of directional transmission channels of carriers were considered using Boltzmann transport theory and 2D electron gas model. The channels were analyzed based on the differences in the electric field distribution of carbon ion layers in SLG, FLG (2 to 9 layers) and GNs (>10 layers; ~3 mm to 100 nm thickness). The surface channels were only observed during electron–electron scattering, and the expression of relaxation time was provided. The interlayer channels were observed during electron–electron and electron–phonon scattering; the former was stronger than the latter. The electrical conductivities of SLG, FLG, GNs, and graphite pieces were numerically computed using Boltzmann transport theory. Results show that the conductivities of FLG and GNs decrease with increasing thickness, and the conductivities remained constant for micron-range thickness. The negative temperature coefficient of resistance increases with temperature for graphite sheet.

2. Electronic transport in 2D plane

2.1. Electrical conductivity of 2D electron gas

Graphene is a 2D material that comprises a single layer of carbon atoms and exhibits a lattice structure that resembles a hexagonal honeycomb. In a flat hexagonal lattice, the sp^2 hybridization of three valence electrons of each carbon and three nearest neighbor carbon atoms forms a stable σ bond (bond length, 0.142 nm). The p orbital electrons that remain in each carbon form π bonds; π electrons freely move to yield a 2D electron gas system. At time t, the electronic number within $\mathrm{d}x\mathrm{d}y$ and $\mathrm{d}k_x\mathrm{d}k_y$ is given as follows:

$$(x, y; k_x, k_y)\mathrm{d}x\mathrm{d}y\mathrm{d}k_x\mathrm{d}k_y \tag{1}$$

After time $\mathrm{d}t$, the incremental number of electrons in the same cell area and momentum interval is $\mathrm{d}f\mathrm{d}x\mathrm{d}y\mathrm{d}k_x\mathrm{d}k_y$ [Eq. (2)]:

$$\begin{aligned}\frac{\mathrm{d}f}{\mathrm{d}t} &= \frac{\partial f}{\partial t} + \frac{\partial f}{\partial x}\frac{\partial x}{\partial t} + \frac{\partial f}{\partial y}\frac{\partial y}{\partial t} + \frac{\partial f}{\partial k_x}\frac{\partial k_x}{\partial t} + \frac{\partial f}{\partial k_y}\frac{\partial k_y}{\partial t} \\ &= \frac{\partial f}{\partial t} + v_x\frac{\partial x}{\partial t} + v_y\frac{\partial y}{\partial t} + F_x\frac{\partial f}{\hbar\partial k_x} + F_y\frac{\partial f}{\hbar\partial k_y}\end{aligned} \tag{2}$$

where F_x and F_y are the x- and y-components of the external field, respectively, and v_x and v_y are the corresponding x- and y-components of the electronic velocity. The distribution state of the electrons changes from f to the original f_0 after relaxation time τ. Thus, from Eq. (2), Boltzmann transport equation can be written as follows:

$$\frac{\partial f}{\partial t} + v_x\frac{\partial x}{\partial t} + v_y\frac{\partial y}{\partial t} + F_x\frac{\partial f}{\hbar\partial k_x} + F_y\frac{\partial f}{\hbar\partial k_y} = -\frac{f - f_0}{\tau} \tag{3}$$

where f_0 is the Fermi function. The distribution function is applied by a stable and uniform external electric field $\boldsymbol{E}_x$ along the x-axis; Eq. (3) is simplified as follows:

$$-e\boldsymbol{E}_x \cdot (\boldsymbol{v}_x + \boldsymbol{v}_y)\frac{\partial f}{\partial \varepsilon} = -\frac{f - f_0}{\tau} \tag{4}$$

Given that the external electric field is far less than the atomic internal counterpart, f and f_0 are nearly similar [Eq. (4)].

$$f = f_0 + \tau e\boldsymbol{E}_x \cdot (\boldsymbol{v}_x + \boldsymbol{v}_y)\frac{\partial f}{\partial \varepsilon} \tag{5}$$

The electronic state number upon considering the relativistic effects 2D plane and energy range $\varepsilon - \varepsilon + \mathrm{d}\varepsilon$ is presented by Eq. (6):

$$D(\varepsilon)\mathrm{d}\varepsilon = \frac{4\pi A}{v^2h^2}\varepsilon\mathrm{d}\varepsilon \tag{6}$$

where A is the area of the 2D electronic plane. The current density is given as follows:

$$\boldsymbol{J}_x = -\frac{e}{Ad}\int \boldsymbol{v}_x f D(\varepsilon)\mathrm{d}\varepsilon = \left[\frac{4\pi e^2}{2dh^2}\int_0^\infty \varepsilon\tau(\varepsilon)\frac{\partial f_0}{\partial \varepsilon}\mathrm{d}\varepsilon\right]\boldsymbol{E}_x \tag{7}$$

where d is the thickness of electronic plane. Based on Ohm's law and polynomial approximation of Fermi integral, the conductivity of a 2D electron gas is obtained using Eq. (7).

$$\sigma = \frac{2\pi e^2}{dh^2}\left\{\varepsilon_F\tau(\varepsilon_F) + \frac{(\pi k_B T)^2}{6}\left[2\tau'(\varepsilon_F) + \varepsilon_F\tau''(\varepsilon_F)\right] + \cdots\right\} \tag{8}$$

where τ is determined by the mechanism of electron scattering.

2.2. Mechanism of electron scattering in 2D plane

Under electric field $\boldsymbol{E}$, the free electrons (π electrons) move directionally in graphene [Fig. 1(a)]. Simultaneously, these electrons are scattered by lattices (phonons) and other free electrons [Figs. 1(b) and 1(c)].

Suppose that d_a and d_e are diameters of C atoms (C ions) and electrons, respectively, v_e and v_a are the respective electronic and atomic speeds, and 2θ is the intersection angle between the directions of electronic movement and scattering. Furthermore, $d_{12} = (d_a + d_e)/2$ is the approximate diameter of the carbon atom. This parameter should be centered in the lattice. The electron collision must be centered on the circumference. At time $\mathrm{d}t$, the electrons scattered along the $\boldsymbol{n}$ direction and confined in the parallelogram at the bottom and top portions are $d_{12}\mathrm{d}\theta$ and $(v_e - v_a)\cos\theta\mathrm{d}t$, respectively; thus, the area is $d_{12}(v_e - v_a)\cos\theta\mathrm{d}\theta\mathrm{d}t$. Therefore, the probability of electrons scattered by the phonon is given as follows:

$$\frac{1}{\tau_a} = \frac{1}{n_{es}}\iint f_a f_e d_{12}(v_e - v_a)\cos\theta\mathrm{d}\theta\mathrm{d}v_a\mathrm{d}v_e \tag{9}$$

where f_a and f_e respectively represent the speed distributions of the phonons and electrons in the 2D plane. Given a phonon at the origin of the geocentric coordinate system, Eq. (9) can be expressed as follows:

$$\frac{1}{\tau_a} = \left(1 + \frac{m_0}{m_a}\right)^{1/2} n_{as}d_a\overline{v}_e \approx n_{as}d_a\overline{v}_e \tag{10}$$

where m_a and m_0 are the respective mass of atoms and electrons, $\overline{v_e}$ is the average of electronic speeds, and n_{as} is the atomic concentration. The scattering probability between the electrons is obtained from Eq. (10). Considering the inter-electron Coulomb force [34,35], the scattering radius for two electrons that approach at a relative velocity [Fig. 1(c)] is presented by Eq. (11):

$$\frac{e^2}{4\pi\varepsilon_0\varepsilon_r d_{12}} = \frac{m_e^*}{4}v_{er}^2 \tag{11}$$

where v_{er} is the relative velocity of the unified electrons and m_e^* is the effective mass of electron. Based on Eqs. (9) and (11), the inter-carrier scattering is determined as follows:

$$\frac{1}{\tau_e} = \frac{\sqrt{2}n_{es}e^2}{4\varepsilon_0\varepsilon_r m_e^*\overline{v}_e} \tag{12}$$

where n_{es} is the electron concentration.