THz-Frequency Conductivity in Monolayer and Bilayer Graphene

Weidong Zhang
Department of Electrical and Computer Engineering
North Carolina State University, Raleigh, NC, 27695
Email: wzhang5@ncsu.edu

Dwight Wooalard
U. S. Army Research Office
RTP, NC, 27709

***Abstract*—The quantum mechanical Boltzmann equation is applied to study the intraband and interband conductivity of monolayer and bilayer graphene. The quantal features for both material systems in response to the field are investigated. The universal feature of conductance of bilayer graphene is verified by the straightforward calculation from the band structure. The intraband conductivity is found important at the THz frequencies.**

I. Introduction

Recent experiments on the transmission spectroscopy within the THz regime have encouraged us to study the THz-frequency conductivity of graphene [1]. At the infrared range, the experimental data is consistent with the universal $e\pi/2h$ value of conductivity ([1], see also, [2] and [3]). At the lower THz range, there is significant conductivity greater than $e\pi/2h$ observed, which could be contributed to the intraband (Drude spectral model). Further, the response of electron transport to the THz-frequency electromagnetic excitations with the time and space variation $\exp(i\omega t - i\mathbf{q}\cdot\mathbf{r})$ is a problem of potential importance to high speed graphene-based applications, such as RF planar circuits, surface plasmon sensor. Additionally the problem is necessary to study for the development of long wavelength plasmon lasers based upon graphene (e. g. [4]).

II. The Models

The approximation q=0 is often used as a simplifying approximation in calculation of conductivity $\sigma(\omega, q)$ [5]. However, the quantum mechanical (QM) Boltzmann equation deviates substantially at large q (by a factor $\hbar^2 q^2/2m/\epsilon_F$, where ϵ_F is the Fermi energy) from its classical limit [6]. This work applies a fully quantum mechanical Boltzman formalism [6], [7] to explore the high frequency conductivity $\sigma(\omega, q)$ of monolayer and bilayer graphene.

This analysis accounts for the density matrix element $\rho_{\mathbf{k}}^{\mathbf{q}} = f^{\omega,\mathbf{q}}(\hbar\mathbf{k})$ that arise between Bloch states, namely with Bloch wavevectors $(\mathbf{k}+\frac{1}{2}\mathbf{q})$, $(\mathbf{k}-\frac{1}{2}\mathbf{q})$ differing by $\mathbf{q}$ under the excitation of $\exp(\pm i\mathbf{q}\cdot\mathbf{r})$. At $q=0$, it is simply the Fermi-Dirac function $\rho_{\mathbf{k}}^{(0)} = f(\epsilon(\mathbf{k}))$, where $\epsilon(\mathbf{k})$ is the electron energy.

The quantum mechanical (QM) Boltzmann equation is

$$[i\omega+\chi+i\omega_{\mathbf{k}}^{\mathbf{q}}+i\gamma_{\mathbf{k}}^{\mathbf{q}}(\omega)]\rho_{\mathbf{k}}^{\mathbf{q}}-\frac{i}{\hbar}C_{\mathbf{k}}^{\mathbf{q}} = \sum_{\mathbf{k}'}\Delta_{\mathbf{k}\mathbf{k}'}^{\mathbf{q}}S_{\mathbf{k}\mathbf{k}'}^{\mathrm{q}}(\rho_{\mathbf{k}'}^{\mathbf{q}}-\rho_{\mathbf{k}}^{\mathbf{q}}) \quad (1)$$

where $\hbar\omega_{\mathbf{k}}^{\mathbf{q}} = \epsilon(\mathbf{k}+\frac{1}{2}\mathbf{q}) - \epsilon(\mathbf{k}-\frac{1}{2}\mathbf{q})$ is the energy difference between the two Bloch states, and the coefficient is

$$C_{\mathbf{k}}^{\mathbf{q}} = \left\langle \mathbf{k}+\frac{1}{2}\mathbf{q}|H'|\mathbf{k}-\frac{1}{2}\mathbf{q}\right\rangle [f(\hbar(\mathbf{k}+\frac{1}{2}\mathbf{q})) - f(\hbar(\mathbf{k}-\frac{1}{2}\mathbf{q}))] \quad (2)$$

where the first term is the Hamiltonian of optical excitation between initial and final states. Another important term is $\gamma_{\mathbf{k}}^{\mathbf{q}}(\omega)$ is the impurity's scattering, which is often proportional to the amplitude of q. On the right are the collisions terms. By imitating the Vlasov equation for plasma, the collisionless QM Boltzmann equation becomes

$$[i\omega+\chi+i\omega_{\mathbf{k}}^{\mathbf{q}}+i\gamma_{\mathbf{k}}^{\mathbf{q}}(\omega)]\rho_{\mathbf{k}}^{\mathbf{q}} - i/\hbar C_{\mathbf{k}}^{\mathbf{q}} = 0 \quad (3)$$

A. Monolayer graphene

The Hamiltonian of the monolayer graphene is

$$H = \begin{pmatrix} 0 & \hbar v_F(k_x - ik_y) \\ \hbar v_F(k_x + ik_y) & 0 \end{pmatrix} \quad (4)$$

where v_F is the Fermi velocity. k_x, k_y are the x,y projections of $\mathbf{k}$. The wavefunctions are

$$|\mathbf{K}, \Psi_{c,v}\rangle = \frac{1}{\sqrt{2}}\exp(i\mathbf{k}\cdot\mathbf{r})\begin{pmatrix} \exp(-i\theta(\mathbf{k})/2) \\ \pm\exp(i\theta(\mathbf{k})/2) \end{pmatrix} \quad (5)$$

where $\theta(\mathbf{k})$ is the angle constructed by the k_x, k_y. The $+$ is for the conduction band, and $-$ is for the valence band.

The optical excitation is $H' = ev_F\sigma\cdot\mathbf{A}$, where A is the vector potential and σ is the Pauli matrix. The velocity of electron is $\mathrm{v}_{\mathbf{k}} = \left\langle \mathbf{k}\pm\frac{1}{2}\mathbf{q}|\frac{\partial H}{\hbar\partial k_x}|\mathbf{k}\pm\frac{1}{2}\mathbf{q}\right\rangle$. The energy dispersions are $\epsilon(\mathbf{k}) = \pm\hbar v_F|\mathbf{k}|$.

The current is related to the density of matrix

$$J_q = e\int d\mathbf{k}\rho_{\mathbf{k}}^{\mathbf{q}}\mathbf{v}_{\mathbf{k}} \quad (6)$$

If we use the wavefunctions above, evaluate the optical perturbation matrix element of $C_{\mathbf{k}}^{\mathbf{q}}$ and perform the integration over the momentum space, we obtain the intraband conductivity

$$\sigma_{\mathtt{intra}} = -i\frac{e^2v_F^2\hbar}{\pi^2}\int_0^\infty kdk\int_0^{2\pi}d\theta\cos^2\Theta \left\{\frac{1}{(\hbar\omega+\epsilon_{c-}-\epsilon_{c+}+i\Gamma(q))}\frac{f_{c-}-f_{c+}}{\epsilon_{c-}-\epsilon_{c+}} - \frac{1}{(-\hbar\omega+\epsilon_{c+}-\epsilon_{c-}+i\Gamma(q))}\frac{f_{c+}-f_{c-}}{\epsilon_{c+}-\epsilon_{c-}}\right\} \quad (7)$$

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Here $\epsilon_{\pm} = \epsilon(\mathbf{k} \pm \frac{1}{2}\mathbf{q})$ and $f_{c\pm} = f_c(\epsilon(\mathbf{k} \pm \frac{1}{2}\mathbf{q}))$. $\gamma_{\mathrm{k}}^{\pm \mathrm{q}}(\mp\omega)$ has been replaced with its imaginary component $\Gamma(q)$, which is the inverse of the relaxation time of the states under frequent impurity's scattering events [6]. The interband conductivity is

$$\begin{aligned}\sigma_{\mathtt{inter}} = i\frac{e^2 v_F^2 \hbar}{\pi^2}\int_0^{\infty} kdk \int_0^{2\pi} d\theta \sin^2\Theta \\ \left\{\frac{1}{(\hbar\omega + \epsilon_{c-} - \epsilon_{v+} - i\chi)}\frac{(f_{c-} - f_{v+})}{(\epsilon_{c-} - \epsilon_{v+})}\right. \\ + \frac{1}{(\hbar\omega + \epsilon_{v-} - \epsilon_{c+} - i\chi)}\frac{(f_{v-} - f_{c+})}{(\epsilon_{v-} - \epsilon_{c+})} \\ - \frac{1}{(-\hbar\omega + \epsilon_{c+} - \epsilon_{v-} - i\chi)}\frac{(f_{c+} - f_{v-})}{(\epsilon_{c+} - \epsilon_{v-})} \\ \left. - \frac{1}{(-\hbar\omega + \epsilon_{v+} - \epsilon_{c-} - i\chi)}\frac{(f_{v+} - f_{c-})}{(\epsilon_{v+} - \epsilon_{c-})}\right\} \quad (8)\end{aligned}$$

Here $\epsilon_{c,v\pm} = \epsilon_{c,v}(\mathbf{k} \pm \frac{1}{2}\mathbf{q})$ and $f_{c,v\pm} = f_{c,v}(\epsilon(\mathbf{k} \pm \frac{1}{2}\mathbf{q}))$. The variable Θ in both cases is $\Theta = \frac{1}{2}[\theta(\mathbf{k} + \frac{\mathbf{q}}{2}) + \theta(\mathbf{k} - \frac{\mathbf{q}}{2})]$. $\chi > 0$ is real and eventually $\chi \to 0$.

B. Bilayer graphene

The Hamiltonian for the bilayer graphene in a manner of approximation is [8],

$$H = -\frac{\hbar^2}{2m}\begin{pmatrix} 0 & (k_x - ik_y)^2 \\ (k_x + ik_y)^2 & 0 \end{pmatrix} \quad (9)$$

where $m = 0.054$ is the effective mass. Here we take the lowest order and assume there is no gap open [8]. The basis functions of H_0 can be found for the conduction band,

$$|\mathbf{K}, \Psi_{c,v}\rangle = \frac{1}{\sqrt{2}}\exp(i\mathbf{k}\cdot\mathbf{r})\begin{pmatrix} \pm e^{-2i\theta(\mathbf{k})} \\ 1 \end{pmatrix} \quad (10)$$

where + sign is for the conduction band and - sign is for the valence band. The energy dispersions are $\epsilon(\mathbf{k}) = \pm\hbar^2 k^2/2m$. The intraband conductivity is

$$\begin{aligned}\sigma_{\mathtt{intra}} = i\frac{\hbar^3 e^2}{4m^2\pi^2}\int_0^{\infty} k^3 dk \int_0^{2\pi}\cos\theta(\mathbf{k})d\theta \\ \left[\frac{\Theta(\mathbf{k} - \frac{\mathbf{q}}{2}, \mathbf{k} + \frac{\mathbf{q}}{2})}{(\hbar\omega + \epsilon_{c-} - \epsilon_{c+} + i\Gamma(q))}\frac{f_{c-} - f_{c+}}{\epsilon_{c-} - \epsilon_{c+}} + \right. \\ \left. \frac{\Theta(\mathbf{k} + \frac{\mathbf{q}}{2}, \mathbf{k} - \frac{\mathbf{q}}{2})}{(\hbar\omega - \epsilon_{c+} + \epsilon_{c-} - i\Gamma(q))}\frac{f_{c+} - f_{c-}}{\epsilon_{c+} - \epsilon_{c-}}\right] \quad (11)\end{aligned}$$

where the function Θ is defined as

$$\begin{aligned}\Theta(-\mathbf{q}/2, +\mathbf{q}/2) \\ = \left(e^{2i\theta(\mathbf{k}+\frac{\mathbf{q}}{2}) - 2i\theta(\mathbf{k})} + e^{2i\theta(\mathbf{k}) - 2i\theta(\mathbf{k}-\frac{\mathbf{q}}{2})}\right) \\ \left(e^{2i\theta(\mathbf{k}-\mathbf{q}/2) - i\theta(\mathbf{k})} + e^{i\theta(\mathbf{k}) - 2i\theta(\mathbf{k}+\mathbf{q}/2)}\right) \quad (12)\end{aligned}$$

, and $\epsilon_{c\pm}$ and $f_{c\pm}$ are denoted the same as above. The interband conductivity is found

$$\begin{aligned}\sigma_{\mathtt{inter}} = -i\frac{e^2\hbar^3}{4m^2\pi^2}\int_0^{\infty} k^3 dk \int_0^{2\pi}\cos\theta(\mathbf{k})d\theta \\ \left\{\frac{\Lambda(\mathbf{q}/2, -\mathbf{q}/2)}{(\hbar\omega + \epsilon_{c-} - \epsilon_{v+} - i\chi)}\frac{(f_{c-} - f_{v+})}{(\epsilon_{c-} - \epsilon_{v+})}\right. \\ + \frac{\Lambda(\mathbf{q}/2, -\mathbf{q}/2)}{(\hbar\omega + \epsilon_{v-} - \epsilon_{c+} - i\chi)}\frac{(f_{v-} - f_{c+})}{(\epsilon_{v-} - \epsilon_{c+})} \\ - \frac{\Lambda(-\mathbf{q}/2, +\mathbf{q}/2)}{(-\hbar\omega + \epsilon_{c+} - \epsilon_{v-} - i\chi)}\frac{(f_{c+} - f_{v-})}{(\epsilon_{c+} - \epsilon_{v-})} \\ \left. - \frac{\Lambda(-\mathbf{q}/2, +\mathbf{q}/2)}{(-\hbar\omega + \epsilon_{v+} - \epsilon_{c-} - i\chi)}\frac{(f_{v+} - f_{c-})}{(\epsilon_{v+} - \epsilon_{c-})}\right\} \quad (13)\end{aligned}$$

where the function of Λ is defined as

$$\begin{aligned}\Lambda(\mathbf{q}/2, -\mathbf{q}/2) \\ = \left(-e^{2i\theta(\mathbf{k}+\frac{\mathbf{q}}{2}) - i\theta(\mathbf{k})} + e^{i\theta(\mathbf{k}) - 2i\theta(\mathbf{k}-\frac{\mathbf{q}}{2})}\right) \\ \left(e^{2i\theta(\mathbf{k}-\frac{\mathbf{q}}{2}) - 2i\theta(\mathbf{k})} - e^{2i\theta(\mathbf{k}) - 2i\theta(\mathbf{k}+\frac{\mathbf{q}}{2})}\right) \quad (14)\end{aligned}$$

and the $\Lambda(-\mathbf{q}/2, \mathbf{q}/2)$ term is to switch the sign of $\mathbf{q}$ above.

When $q = 0$, the conductivity equations are the same as obtained from other methods (e. g. [5]).

III. Results

We discuss the intraband and interband conductivity separately.

A. Intraband

The analysis equations (7) and (11) reveal that the Drude peak if the photon energy $\hbar\omega$ is equal to $\Gamma(q)$. Therefore, the intraband frequency-dependent conductivity $\sigma_{\mathtt{intra}}$ becomes important at low optical frequencies. Also, the $\Gamma(q)$ is proportional to the spatial wavevector q [6], implying the space variations of electromagnetic fields need be considered at low optical frequencies.

The effect of space variations is further examined by assuming the $\Gamma(q)$ is a constant without any dependence of q. The intraband frequency-dependent component of the conduction-band conductivity for bilayer graphene is plotted in Fig. 1, where the value of Γ was taken as $\Gamma = 0.005$eV (this value may be larger, see Ref. [3]) and an upper Fermi energy of $k_F = 51,0001$/cm was assumed. Here, the maximum conductivity corresponds close to the photonic energy value used for Γ , i.e., $f = \Gamma/2\pi\hbar$=1.2 THz. Hence, the conductivity in the THz regime is strongly influenced by impurity scattering and potentially may be applied to characterize the level of doping and/or defects in bilayer graphene. Also, if q (or $\hbar^2 q^2/2m$) becomes comparable with k_F (or E_F), the intraband conductivity varies appreciably which is an indicator of charge accumulation or depletion effects. This behaviour is consistent with the prediction given in [6]. Therefore at large q, the QM Boltzmann equation is more appropriate than the semi-classical one.

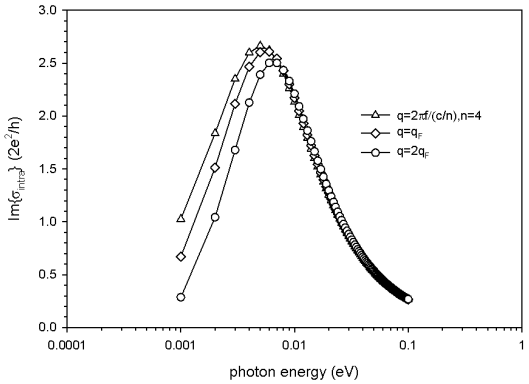

Fig. 1. The imaginary part of the intraband conductivity in the conduction band of bilayer graphene. $k_F = 51,000$/cm, and $\Gamma \approx$0.005eV. The temperature is 300K.

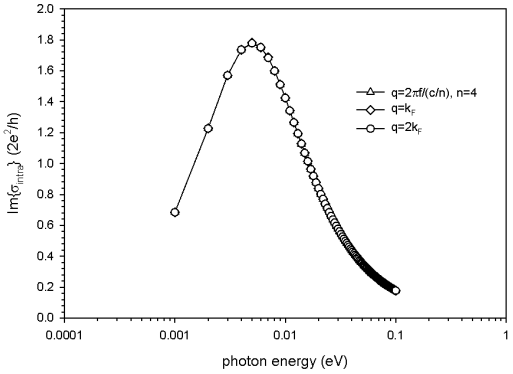

Fig. 2. The imaginary part of the intraband conductivity in the conduction band of monomlayer graphene. $k_F = 1,100$/cm, and $\Gamma \approx$0.005eV. The temperature is 300K.

The calculations for monlayer (see Fig. 2) indicate there is no deviation in $\sigma_{\mathtt{intra}}$ even at large $q \sim k_F$ as was observed for the bilayer case. In this case, the semi-classical Boltzmann equation is still good.

B. Interband

The temperature dependence of the dynamical conductivity for the monolayer graphene is plotted in Fig. 3. The conductivity switches to $e^2/4\hbar$ when the photon energy $\hbar\omega$ is around the twice of the Fermi energy ϵ_F. The intraband conductivity for the bilayer graphene is plotted in Fig. 4. At the high optical frequencies, the sheet conductivity is very close to $e^2/4\hbar$, almost the same as in the case of monolayer graphene. This is consistent with what was observed in the experiments of graphite [9].

When the value of q is compared to the Fermi surfaces (in a hypothetical case of surface plamson), the conductivity deviates from their semi-classical limit in both monolayer and

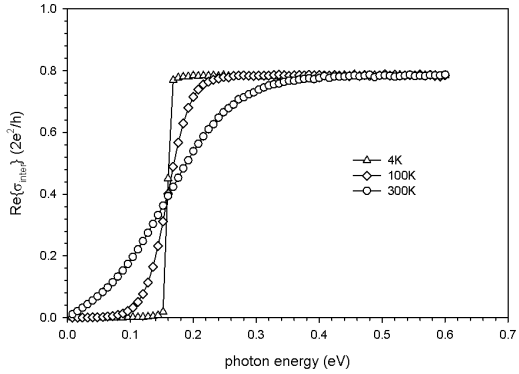

Fig. 3. The real part of the interband conductivity in the conduction band of momolayer graphene at different temperatures. $k_F = 1.1 \times 10^6$/cm, $\epsilon_F =$ 79.6meV, and q=0.

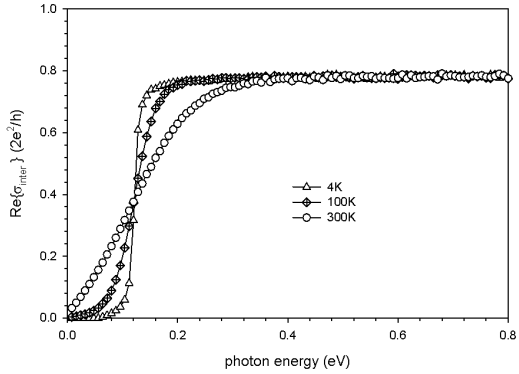

Fig. 4. The real part of the interband conductivity in the conduction band of bilayer graphene at different temperatures. $k_F = 3.1 \times 10^6$/cm, $\epsilon_F =$ 60.5meV, and q=0.

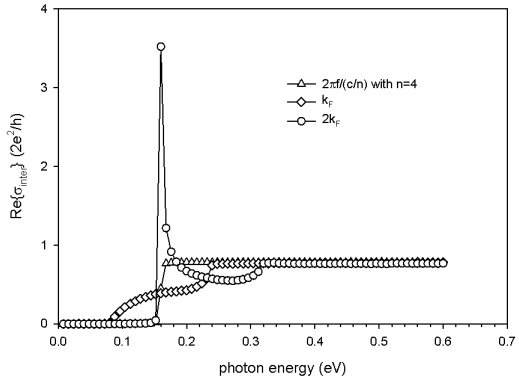

Fig. 5. The real part of the interband conductivity in the conduction band of momolayer graphene on the space variation. $k_F = 1.1 \times 10^6$/cm, $\epsilon_F =$ 79.6meV. The temperature is 4K.

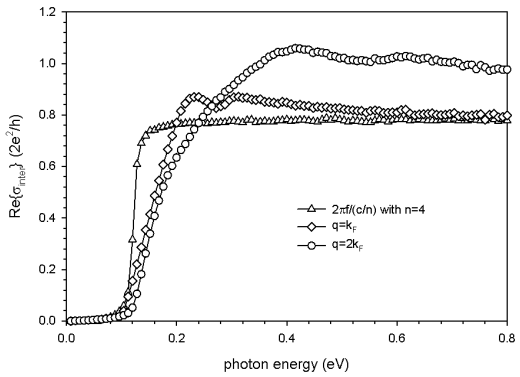

Fig. 6. The real part of the interband conductivity in the conduction band of bilayer graphene on the space variation. $k_F = 3.1 \times 10^6$/cm, $\epsilon_F = 60.5$meV. The temperature is 4K.

bilayer graphene. In the examples plotted Fig.5 and Fig. 6, the Fermi energy levels are in the valence bands. Particularly, the deviation mostly occurs near the twice of the Fermi energy.

Thus, in some cases of larger frequencies and space gradients, the semi-classical Boltzmann equation need be transformed into the quantum mechanical domain.

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