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Article in World Journal of Condensed Matter Physics · January 2015

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High-Frequency Electric Field Induced Nonlinear Electron Transport in Chiral Carbon Nanotubes

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Received 5 November 2014; accepted 21 November 2015; published 24 November 2015

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Abstract

We investigate theoretically the high frequency complex conductivity in carbon nanotubes that are stimulated axially by a strong inhomogeneous electric field of the form $E(t) = E_0 + E_1 \cos(\omega t)$. Using the kinetic approach based on Boltzmann's transport equation with constant relaxation time approximation and the energy spectrum of the electron in the tight-binding approximation, together with Bhatnagar-Gross-Krook collision integral, we predict high-frequency nonlinear effects along the axial and the circumferential directions of the carbon nanotubes that may be useful for the generation of high frequency radiation in the carbon nanotubes.

Keywords

Carbon Nanotubes, High Frequency Electric Field, Electric Current Density, Complex Conductivity

1. Introduction

Carbon nanotube (CNT) is an allotrope of carbon with nanometers size diameter and an aspect ratio as high as $\sim 10^7$. In 1952, images of 50 nanometer diameter tubes of carbon were reported by Radushkevich and Lukyanovich [1].

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How to cite this paper: Abukari, S.S., et al. (2015) High-Frequency Electric Field Induced Nonlinear Electron Transport in Chiral Carbon Nanotubes. *World Journal of Condensed Matter Physics*, 5, 294-300.

<http://dx.doi.org/10.4236/wjcmp.2015.54030>

Using the vapor-growth technique, Orberlin and coworkers [2] reported observation of hollow carbon fibers of nanometer size diameters. Other reports also published similar observation of tubular carbon nanostructures [3]. However, the credit for the discovery of CNTs goes to S. Iijima [4]. This one-atom thick sheet of graphene rolled up into a seamless cylinder has since attracted a great deal of interest, mainly due to their unique thermal, chemical and physical properties [5] [6]. These properties depend on the fundamental indices (n,m) of the CNTs. The indices (n,m) determine the diameter and the chiral angle of the CNTs. As n and m vary, the conduction ranges from metallic to semiconducting [7], with an inverse diameter dependent band gap of ≤ 1 eV [7].

The electron transport properties of the CNTs continue to be the subject of intense research. CNTs have been shown to exhibit ballistic transport [8]-[10], Coulomb-blockade [11]-[13], Luttinger Liquid [14] [15] and superconductivity [16] [17]. The unique architectural, structural and physicochemical properties have made CNT a promising candidate for application in new generation of nanoelectronics [18]-[21], sensors [22] [23], electrochemical capacitors [24]-[26], Li-ion batteries [27]-[29] and terahertz (THz) generation and amplification [30]-[34], just to mention a few. Furthermore, negative differential conductivity properties of CNTs have been reported [6] [35] [36], while phenomena like rectification of electromagnetic waves, domain suppression in negative differential conductivity region, high-frequency (hf) conductivity, high order harmonic generation and many others have also been considered [37]-[40].

In this work, we report on a high frequency (hf) complex conductivity $\sigma(\omega)$ in CNTs using the standard linear electro-dynamical approach [15]-[17]. We use the kinetic approach based on Boltzmann's transport equation with constant relaxation time approximation and the energy spectrum of the electron in the tight-binding approximation, together with Bhatnagar-Gross-Krook (BGK) collision integral. This approach permits adequate allowance for the particle-number conservation law for scattering inhomogeneous field as well as the influence on the electron spectrum by both resonance effects due to Bloch oscillations of the electrons and by the effects connected with the carrier drift and diffusion under conditions of strong spatial dispersions.

2. Theory

Using the simple model of the tight-binding approximation, we describe the energy spectrum of the CNTs as [39]

$$\xi(p) = \xi_o - \Delta_s \cos \frac{p_s d_s}{\hbar} - \Delta_z \cos \frac{p_z d_z}{\hbar}, \quad (1)$$

where the indices s and z correspond to the circumferential and axial directions, respectively. ξ_o is the energy of an outer-shell electron in an isolated carbon atom, Δ_z and Δ_s are the real overlapping integrals for jumps along the respective coordinates, p_s and p_z are the components of momentum tangential to the base helix and along the nanotube axis, respectively. \hbar is Planck's constant, and d_z and d_s are the distances along the CNT axis and helix, respectively.

We consider the Boltzmann transport equation with constant relaxation time together with Bhatnagar-Gross-Krook (BGK), which account for the spatial effects due to the conservation of the number of scattered particles [41] [42],

$$\frac{\partial f(x, p, t)}{\partial t} + v(p) \frac{\partial f(x, p, t)}{\partial x} + eE(t) \frac{\partial f(x, p, t)}{\partial p} = -\frac{1}{\tau} \left(f(x, p, t) - \frac{n(x)}{n_o} f_o(p) \right). \quad (2)$$

The distribution function $f(x, p, t)$ and the equilibrium distribution function $f_o(p)$ are periodic and can be written in Fourier series as in Equations (3) and (4), respectively.

$$f(x, p, t) = \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} e^{i \left(m \frac{p_s d_s}{\hbar} + n \frac{p_z d_z}{\hbar} \right)} \phi_m(t) \quad (3)$$

and

$$f_o(p) = \frac{n_o d_s d_z}{2I_o \left(\frac{\Delta_s}{\kappa_B T} \right) I_o \left(\frac{\Delta_z}{\kappa_B T} \right)} \sum_{m=-\infty}^{\infty} I_m \left(\frac{\Delta_s}{\kappa_B T} \right) \sum_{n=-\infty}^{\infty} I_n \left(\frac{\Delta_z}{\kappa_B T} \right) e^{i \left(m \frac{p_s d_s}{\hbar} + n \frac{p_z d_z}{\hbar} \right)} \quad (4)$$

where n_o is the equilibrium carrier density, $n(x)$ is the electron density at position x , $v(p)$ is the electron velocity, p

is the electron dynamical momentum, t is elapsed time, τ is the electron relaxation time, e is the electron charge, $E(t)$ is the external electric field, κ_B is the Boltzmann's constant and T is the temperature. The collision integral is taken in the relaxation time τ approximation and further assumed to be constant. $\phi_m(t)$ is the factor by which the Fourier transform of the nonequilibrium distribution function differs from its equilibrium distribution counterpart. $I_m(I_n)$ is the modified Bessel function of the order $m(n)$ and I_o is the modified Bessel function of the zeroth order.

In addition to Equation (2), we employ the Poisson equation that will allow for continuous current, *i.e.*, $\partial E/\partial x = (\varepsilon/\varepsilon_o\varepsilon)[n(x) - n_o]$, where ε is the lattice dielectric constant. For a constant homogeneous electric field E_i , the electron distribution for both the tubular axis z and the base helix s is expressed as:

$$f_{o_{i=s,z}} = \frac{n_o d_i}{2I_o\left(\frac{\Delta_i}{\kappa_B T}\right)} \sum_{m=-\infty}^{\infty} I_m\left(\frac{\Delta_s}{\kappa_B T}\right) \sum_{n=-\infty}^{\infty} I_n\left(\frac{\Delta_z}{\kappa_B T}\right) \frac{v}{(v + im, n\Omega_i)} \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} e^{i\left(\frac{m p_s d_s + n p_z d_z}{\hbar}\right)} \quad (5)$$

where $\Omega_{i=s,z} = ed_i E_i$, $\hbar = 1$ and ν ($\nu = 1/\tau$) is the scattering frequency. We use the conditions of inhomogeneous perturbations with frequency ω and wave-vector k of the following form: $n(x) = n_o + n_{\omega,k} e^{-i(\omega t + kx)}$, $E(x,t) = E_o + E_{\omega,k} e^{-i(\omega t + kx)}$ and $f(x, \varphi_i, t) = f_{o_i} + f_{\omega,k} e^{-i(\omega t + kx)}$, and solving the linearized kinetic equation (Equation (2)), we obtained [41]-[43],

$$\frac{\partial f_{\omega,k}}{\partial \varphi_i} + i[\alpha + \beta \sin \varphi_i] f_{\omega,k} = \frac{\Omega_i}{\Omega_i} \frac{\partial f_{o_i}}{\partial \varphi_i} + \frac{\nu n_{\omega,k}}{n_o} f_o(\varphi_i) \quad (6)$$

where $\alpha = -(\omega + i\nu)/\Omega_i$, $\beta = k\Delta_i(d_i/\hbar)$ is the normalized wave number of the perturbations, and $\varphi_i = p_i d_i/\hbar$ ($i = s, z$) is the dimensionless quasimomentum. The electron flux along the tubular axis (Φ_z) and the base helix (Φ_s) can be expressed as [39] [40],

$$\Phi_z = \frac{2e}{(2\pi\hbar)^2} \iint v(p_z) f(p, t) dp_s dp_z \quad (7a)$$

$$\Phi_s = \frac{2e}{(2\pi\hbar)^2} \iint v(p_s) f(p, t) dp_s dp_z \quad (7b)$$

where the integration is done over the first Brillouin zone. Using Equation (7) together with the solution of Equation (6) we obtain the following expressions:

$$\Phi_z(\omega, k) = -\frac{\varepsilon' \varepsilon_o v_z(\Omega_z)}{I\left(\frac{\Delta_z}{\kappa_B T}\right)} \sum_{m,i=-\infty}^{\infty} I_i\left(\frac{\Delta_z}{\kappa_B T}\right) \left[1 - \frac{l\omega_p^2}{k\Omega_z(\nu + il\Omega_z)}\right] \frac{i^l m j_m(\beta_z) \Omega_z j_{m-1}(\beta_z)}{\omega + i\nu - m\Omega_z} \quad (8a)$$

$$\Phi_s(\omega, k) = -\frac{\varepsilon' \varepsilon_o v_s(\Omega_s)}{I\left(\frac{\Delta_s}{\kappa_B T}\right)} \sum_{m,i=-\infty}^{\infty} I_i\left(\frac{\Delta_s}{\kappa_B T}\right) \left[1 - \frac{l\omega_p^2}{k\Omega_s(\nu + il\Omega_s)}\right] \frac{i^l m j_m(\beta_s) \Omega_s j_{m-1}(\beta_s)}{\omega + i\nu - m\Omega_s} \quad (8b)$$

Defining the axial j_z and the circumferential j_s components of the current density as $j_z = \Phi_z + \Phi_s \sin \theta_h$ and $j_s = \Phi_s \cos \theta_h$, respectively [39] [40], where θ_h is the chiral angle of the CNTs, the current densities are expressed as [43] [44],

$$j_z(\omega, k) = -\frac{\varepsilon' \varepsilon_o v(\Omega_z)}{I_o\left(\frac{\Delta_z}{\kappa_B T}\right)} \sum_{m,i=-\infty}^{\infty} I_i\left(\frac{\Delta_z}{\kappa_B T}\right) \left[1 - \frac{l\omega_p^2}{k\Omega_z(\nu + il\Omega_z)}\right] \frac{i^l m J_m(\beta_z) \Omega_z J_{m-1}(\beta_z)}{\omega + i\nu - m\Omega_z} \\ - \frac{\varepsilon' \varepsilon_o v(\Omega_s)}{I_o\left(\frac{\Delta_s}{\kappa_B T}\right)} \sum_{m,i=-\infty}^{\infty} I_i\left(\frac{\Delta_s}{\kappa_B T}\right) \left[1 - \frac{l\omega_p^2}{k\Omega_s(\nu + il\Omega_s)}\right] \frac{i^l m J_m(\beta_s) \Omega_s J_{m-1}(\beta_s)}{\omega + i\nu - m\Omega_s} \sin \theta_h \quad (9a)$$

and

$$j_s(\omega, k) = -\frac{\varepsilon' \varepsilon_o v(\Omega_s)}{I_o \left(\frac{\Delta_s}{\kappa_B T} \right)} \sum_{m,i=-\infty}^{\infty} I_i \left(\frac{\Delta_s}{\kappa_B T} \right) \left[1 - \frac{l \omega_p^2}{k \Omega_s (v + i l \Omega_s)} \right] \frac{i^l m J_m(\beta_s) \Omega_z J_{m-l}(\beta_s)}{\omega + i v - m \Omega_s} \cos \theta_h. \quad (9b)$$

Using $j_i(\omega, k) = \sigma(\omega, k) E(\omega, k)$ and linearizing the result with $J_o(\beta) = 1$ for $\beta \rightarrow 0$, the expressions for the complex conductivities along the axial and the circumferential directions are expressed, respectively, as:

$$\sigma_z(\omega) = -\varepsilon' \varepsilon_o \omega_p^2 \frac{I_1}{I_o} \left\{ \frac{v^2 (\Omega_z - v^2 + i v \omega)}{(v^2 + \Omega_z^2) [(\omega + i v)^2 - \Omega_z^2]} \right\} - \varepsilon' \varepsilon_o \omega_p^2 \frac{I_1}{I_o} \left\{ \frac{v^2 (\Omega_s - v^2 + i v \omega)}{(v^2 + \Omega_s^2) [(\omega + i v)^2 - \Omega_s^2]} \right\} \sin \theta_h \quad (10a)$$

and

$$\sigma_s(\omega) = -\varepsilon' \varepsilon_o \omega_p^2 \frac{I_1}{I_o} \left\{ \frac{v^2 (\Omega_s - v^2 + i v \omega)}{(v^2 + \Omega_s^2) [(\omega + i v)^2 - \Omega_s^2]} \right\} \cos \theta_h. \quad (10b)$$

3. Results and Discussions

Using the solution to the Boltzmann's transport equation with constant relaxation time together with Bhatnagar-Gross-Krook collision integral, we obtained the expressions for the complex conductivities along the axial and circumferential directions of the CNTs. We analyzed the effects of the high frequency electric field by considering the dependence of the complex conductivity $\sigma(\omega)/\sigma_o$ on the dimensionless frequency ω/Ω (Equation (10)). Shown in **Figure 1** are the plots of the complex conductivities along the axial [**Figure 1(a)**] and the

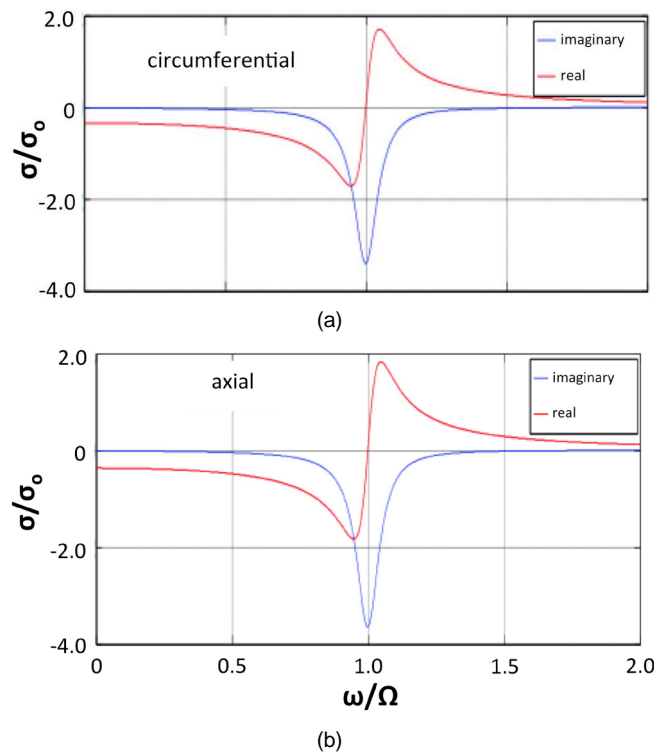


Figure 1. The real (red solid curve) and the imaginary (blue solid curve) parts of the normalized complex conductivity σ/σ_o as a function of normalized frequency ω/Ω along the circumferential (top) and axial (bottom) directions of CNT.

circumferential [Figure 1(b)] directions. The red and the blue curves represent the real and the imaginary parts of the conductivity, respectively. The complex conductivity along both directions exhibits similar characteristics. The dimensionless complex conductivity $\sigma(\omega)/\sigma_0$ depends strongly and nonlinearly on the normalized frequency ω/Ω . It can be seen from the plots that the real part of the complex conductivity becomes more negative with increasing the frequency until a resonance minimum occurs just before the Bloch frequency Ω . This negative-conductivity resonance near the Bloch frequency makes the CNT a potential active medium for Bloch oscillations without domain instabilities induced by negative dc conductivity.

4. Conclusion

In summary, we have obtained the nonlinear high frequency conductivity in CNTs that are stimulated axial with a strong inhomogeneous electric field by using the Boltzmann's equation with constant relaxation time approximation together with the Bhatnagar-Gross-Krook collision integral. We predict this high-frequency nonlinear effect along the axial and the circumferential directions of the carbon nanotubes may be useful for the generation of a high frequency radiation in the carbon nanotubes.

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