

Influence of temperature variation on the electrical conductivity of zigzag carbon nanotubes under homogeneous axial dc field

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We present theoretical framework investigations of the influence of temperature variation on the electrical conductivity of zigzag carbon nanotubes (CNTs) under the applied homogeneous axial dc field. This study was done semiclassically by solving Boltzmann transport equation to derive the current density of zigzag CNT as a function of homogenous axial dc field and temperature. Plots of the normalized current density versus homogeneous dc field applied along the axis of semiconducting zigzag CNTs as room temperature increases from 293 to 299 K revealed a significant increase in electrical conductivity, whereas in metallic zigzag CNTs, almost constant or a negligible decrease in electrical conductivity is observed. The study predicts semiconducting zigzag CNT as a potential material for temperature sensors since it exhibits a faster response and a substantially higher sensitivity to room temperature changes than the metallic counterpart. The electrical conductivity of metallic zigzag CNTs increases immensely as the temperature is reduced to a very low value which could probably lead to a potential superconductivity property that usually occurs at very low temperatures. These potential temperature sensors and superconductors of nanomaterial have vast applications in current-day science and technology.

Keywords: carbon nanotubes, conductivity, temperature variation, current density, dc field.

Introduction

Carbon nanotubes (CNTs) which are allotropes of carbon can behave like metals or semiconductors depending on the twist of the tubes [1, 2]. Zigzag ($n, 0$) carbon nanotube is metallic or conducting if the integer n is a multiple of 3 or else a semiconductor [3]. Much progress has been made recently showing that carbon nanotubes are advanced quasi-1D materials for future high-performance electronics [4–6]. These quasi-1D nanostructural materials have a wide variety of possible applications [7–13].

There are several reports on negative differential conductivity (NDC) in CNTs where room temperature which is the ambient temperature that is suitable for human occupancy and at which laboratory experiments are usually performed is assumed to be constant throughout the study in each report [14–17]. In reality, room temperature varies

from one climatic region to another, one day to another and even within a day. We are therefore reporting the influence of temperature variation on the electrical conductivity of zigzag CNTs which to the best of our knowledge is limited. Thus, in this paper, we present theoretical framework investigations of the influence of temperature variation on the electrical conductivity of semiconducting and metallic zigzag carbon nanotubes under applied homogeneous axial dc field using the semiclassical Boltzmann's transport equation to derive current density. We probe the behavior of the electric current density of the CNTs as a function of the applied homogeneous axial dc field as room temperature varies from 293 to 299 K especially in semiconducting zigzag carbon nanotubes. We further studied the immense increase of electrical conductivity of metallic zigzag carbon nanotube as the temperature is reduced to a very low value.

Theory

Suppose an undoped single-walled zigzag $(n, 0)$ CNT exposed to a homogeneous axial dc field E_z given by

$$E_z = \frac{V}{L}, \quad (1)$$

where V is the voltage between the CNT ends and L is the length of the nanotube. For a CNT, the energy level spacing $\Delta\varepsilon$ is given by $\Delta\varepsilon = \pi\hbar V_F / L$ [14], $\hbar = h / 2\pi$, h is Planck constant, V_F is Fermi velocity. The investigation is done within the semiclassical approximation in which the motion of the π -electrons are considered as the classical motion of free quasiparticles in the field of the crystalline lattice with dispersion law extracted from the quantum theory [14].

Taking into account the hexagonal crystalline structure of a rolled graphene in a form of CNTs and using the tight binding approximation, the energy for zigzag carbon nanotube (CNT) is expressed as in (2) [18],

$$\varepsilon(s\Delta p_\phi, p_z)_s(p_z) = \pm\gamma_0 \left[1 + 4 \cos(ap_z) \cos\left(\frac{a}{\sqrt{3}}s\Delta p_\phi\right) + 4 \cos^2\left(\frac{a}{\sqrt{3}}s\Delta p_\phi\right) \right]^{\frac{1}{2}}, \quad (2)$$

where a is the lattice constant of the CNT, $\gamma_0 \approx 3.0$ eV is the overlapping integral, p_z is the axial component of quasimomentum, Δp_ϕ is transverse quasimomentum level spacing and s is an integer. The expression for lattice constant a in Eq. (2) is given by

$$a = \frac{3a_{c-c}}{2\hbar}, \quad (3)$$

where $a_{c-c} = 0.142$ nm is the C–C bond length.

The “–” and “+” signs correspond to the valence and conduction bands, respectively. Due to the transverse quantization of the quasimomentum p , its transverse component p_ϕ can take n discrete values [14],

$$p_\phi = s\Delta p_\phi = s \frac{\pi\sqrt{3}}{an} (s = 1, \dots, n). \quad (4)$$

Unlike transverse quasimomentum, p_ϕ , the axial quasimomentum p_z is assumed to vary continuously within the range $0 \leq p_z \leq 2\pi/a$, which corresponds to the model of infinitely long CNT ($L = \infty$). This model is applicable to the case under consideration because we are restricted to temperatures and/or voltages well above the level spacing [14, 19], i.e., $k_B \gg \varepsilon_c, \Delta\varepsilon$, where k_B is Boltzmann constant, ε_c is the charging energy and T is the absolute temperature.

Considering the motion of quasiparticles in an external axial electric field is described by the Boltzmann kinetic equation is given by [14, 19]:

$$\frac{\partial f(p, t)}{\partial t} + v_z \frac{\partial f(p, t)}{\partial z} + eE(t) \frac{\partial f(p, t)}{\partial p_z} = -\nu [f(p, t) - f_0(p)], \quad (5)$$

where $f_0(p)$ is equilibrium Fermi distribution function, $f(p, t)$ is the distribution function, v_z is the quasiparticle group velocity along the z axis of carbon nanotube, ν is the frequency, t is time taken, and e is the charge of the propagating electrons. The relaxation term τ given as $\nu = 1/\tau$ of Eq. (5) describes the electron-phonon scattering, electron-electron collisions, etc. Using the method originally developed in the theory of quantum semiconductor superlattices [14], an exact solution of Eq. (5) can be constructed without assuming a weak electric field. Expanding the distribution functions of interest in Fourier series as

$$f(p, t) = \Delta p_\phi \sum_{s=1}^n \delta(p_\phi - s\Delta p_\phi) \sum f_{rs} e^{iarp_z} \psi_\nu(t), \quad (6)$$

$$f_0(p) = \Delta p_\phi \sum_{s=1}^n \delta(p_\phi - s\Delta p_\phi) \sum_{r \neq 0} f_{rs} e^{iarp_z}, \quad (7)$$

where $\delta(p_\phi - s\Delta p_\phi)$ is the Dirac delta function, f_{rs} is the coefficients of the Fourier series and $\psi_\nu(t)$ is the factor by which the Fourier transform of the nonequilibrium distribution function differs from its equilibrium distribution counterpart. Substituting Eqs. (6) and (7) into Eq. (5), the below expression is obtained

$$\frac{\partial \psi_\nu(t)}{\partial t} + (iearE_z + \nu)\psi_\nu(t) = \nu. \quad (8)$$

Solving the homogeneous differential equation corresponding to Eq. (8), we obtain

$$\psi_\nu(t) = \frac{\nu\hbar}{(\nu\hbar + iraeE_z)}. \quad (9)$$

The expression for the coefficients f_{rs} of Eqs. (6) and (7) is found to be

$$f_{rs} = \frac{a}{2\pi\Delta p_\phi} \int_0^{2\pi/a} \frac{\exp(-iarp_z)}{1 + \exp\{\varepsilon_s(p_z)/(k_B T)\}} dp_z \quad (10)$$

The surface current density is defined by [19–21],

$$j_z = \frac{2e}{(2\pi\hbar)^2} \iint f(p, t) v_z(p) d^2 p,$$

or

$$j_z = \frac{2e}{(2\pi\hbar)^2} \sum_{s=1}^n \int_0^{2\pi/a} f(p_z, s\Delta p_\phi, \psi_\nu(t)) v_z(p_z, s\Delta p_\phi) dp_z, \quad (11)$$

where the integration is over the first Brillouin zone, v_z is given by

$$v_z(p_z, s\Delta p_\phi) = \frac{\partial \varepsilon_s(p_z)}{\partial p_z}. \quad (12)$$

Now $\varepsilon_s(p_z)/\gamma_0$ is expressed in Fourier series with coefficients ε_{rs} to be determined

$$\varepsilon(p_z, s\Delta p_\phi) = \varepsilon_s(p_z) = \gamma_0 \sum_{r \neq 0} \varepsilon_{rs} \exp(iarp_z). \quad (13)$$

The expression for the coefficients is found to be

$$\varepsilon_{rs} = \frac{a}{2\pi\gamma_0} \int_0^{2\pi/a} \varepsilon_s(p_z) \exp(-iarp_z) dp_z, \quad (14)$$

where $\varepsilon_s(p_z)$ is given by Eq. (2) and from Eqs. (12) and (13), $v_z(p_z, s\Delta p_\phi)$ is obtained as

$$\begin{aligned} v_z(p_z, s\Delta p_\phi) &= \gamma_0 \sum_{r \neq 0} \frac{\partial(\varepsilon_{rs} \exp(iarp_z))}{\partial p_z} \\ &= \gamma_0 \sum_{r \neq 0} iar\varepsilon_{rs} \exp(iarp_z). \end{aligned} \quad (15)$$

From Eqs. (6) and (7)

$$f(p_z, s\Delta p_\phi, \Psi_\nu(t)) = \Delta p_\phi \sum_{r \neq 0} f_{rs} \exp(iarp_z) \Psi_\nu(t). \quad (16)$$

Substituting Eqs. (15) and (16) into Eq. (11), the current density for zigzag CNT as a function of dc field (E_z) and temperature (T) and taking only the real part into consideration we obtained

$$\begin{aligned} j_z &= j_0 \frac{1}{n} \sum_{r=1}^{\infty} \left(\frac{raeE_z \nu \hbar}{\hbar^2 \nu^2 + (raeE_z)^2} \right) \\ &\times \sum_{s=1}^n \left(\frac{a}{2\pi\Delta p_\phi} \int_0^{2\pi/a} \frac{\exp(-iarp_z)}{1 + \exp\{\varepsilon_s(p_z)/k_B T\}} dp_z \right) \varepsilon_{rs}, \end{aligned} \quad (17)$$

where

$$j_0 = \frac{4\sqrt{3}e^2\gamma_0}{\hbar^2}, \quad \varepsilon_{rs} = \frac{a}{2\pi\gamma_0} \int_0^{2\pi/a} \varepsilon_s(p_z) \exp(-iarp_z) dp_z, \text{ and}$$

$$\varepsilon_s(p_z) = \pm\gamma_0 \left[1 + 4 \cos(ap_z) \cos\left(\frac{s\pi}{n}\right) + 4 \cos^2\left(\frac{s\pi}{n}\right) \right]^{1/2}.$$

The Coulomb electron-electron interaction has been neglected in our approach. It has been established that the short-range Coulomb electron-electron interaction, typical for quasi-1D dimensional CNTs, has only weak effects especially at high temperatures [14, 22].

The effect of the space charge injection and accumulation in carbon nanotubes is suppressed to a large extent when the strength of the external electric field is less than 400 kV/cm [23].

Results and Discussion

In Fig. 1, we displayed the normalized current density ($J_z = j_z / j_0$) versus electric field (E_z) showing increasing electrical conductivity in both ohmic conductivity region $\partial J_z / \partial E_z > 0$ and NDC region $\partial J_z / \partial E_z < 0$ as the room temperature increases from 293 to 299 K for semiconducting zigzag (a) (2, 0), (b) (4, 0), (c) (5, 0), and (d) (7, 0) CNTs. From each of Fig. 1, it has been observed that the peak normalized current density (J_z) and the electrical conductivity $|\partial J_z / \partial E_z|$ represented by the tangent to the curve at any dc field (E_z) for either Ohmic ($\partial J_z / \partial E_z > 0$) or NDC ($\partial J_z / \partial E_z < 0$) regions increase as the room temperature increases from 293 to 299 K for semiconducting zigzag (a) (2, 0), (b) (4, 0), (c) (5, 0), and (d) (7, 0) CNTs. This could be attributed to the fact that at lower room temperature only small numbers of electrons overcome the energy gap into conduction band for conduction. As the room temperature increases from 293 to 299 K, the number of electrons that acquired enough energy to overcome energy gap into conduction band for conduction increases. Hence the observed increase in electrical conductivity as the room temperature increases from 293 to 299 K which is a measure of the degree of steepness of the curve at any dc field (E_z). In Fig. 2, the normalized current density (J_z) versus electric field (E_z) showing the electrical conductivity as the room temperature increases from 293 to 299 K for (a) metallic zigzag (3, 0) CNT, (b) semiconducting zigzag (8, 0) CNT in each case is displayed. It has been observed that unlike semiconducting zigzag CNTs in which as the room temperature increases from 293 to 299 K the electrical conductivity also increases as shown in Fig. 2(b), the electrical conductivity of metallic zigzag CNTs as the room temperature increases from 293 to 299 K remains almost constant especially in ohmic region as shown in Fig. 2(a). This is due to the fact that in metallic zigzag CNTs, there is no energy gap and the valence band and conduction band touch each other. Hence a relatively small increase in room temperature from 293 to 299 K will not increase the number of conduction electrons in the conduction band and also the vibration of lattice leading to the increase in the scattering rate of electrons as a result of the increase in room temperature from 293 to 299 K is minimal. Therefore, the influence of room temperature changes on the electrical conductivity of semiconducting zigzag CNTs is significant while that of metallic zigzag CNTs is negligible for relatively small changes in room temperature from 293 to 299 K.

In Fig. 3, the normalized current density J_z as a function of electric field E_z showing increasing electrical conductivity as temperature decreases from room temperature of 293 K to a very low temperature of 152 K for metallic zigzag: (a) (3, 0), (b) (6, 0), (c) (9, 0), and (d) (12, 0) CNTs in each case is clearly shown. From each of Fig. 3, it has been observed that the peak normalized current density J_z

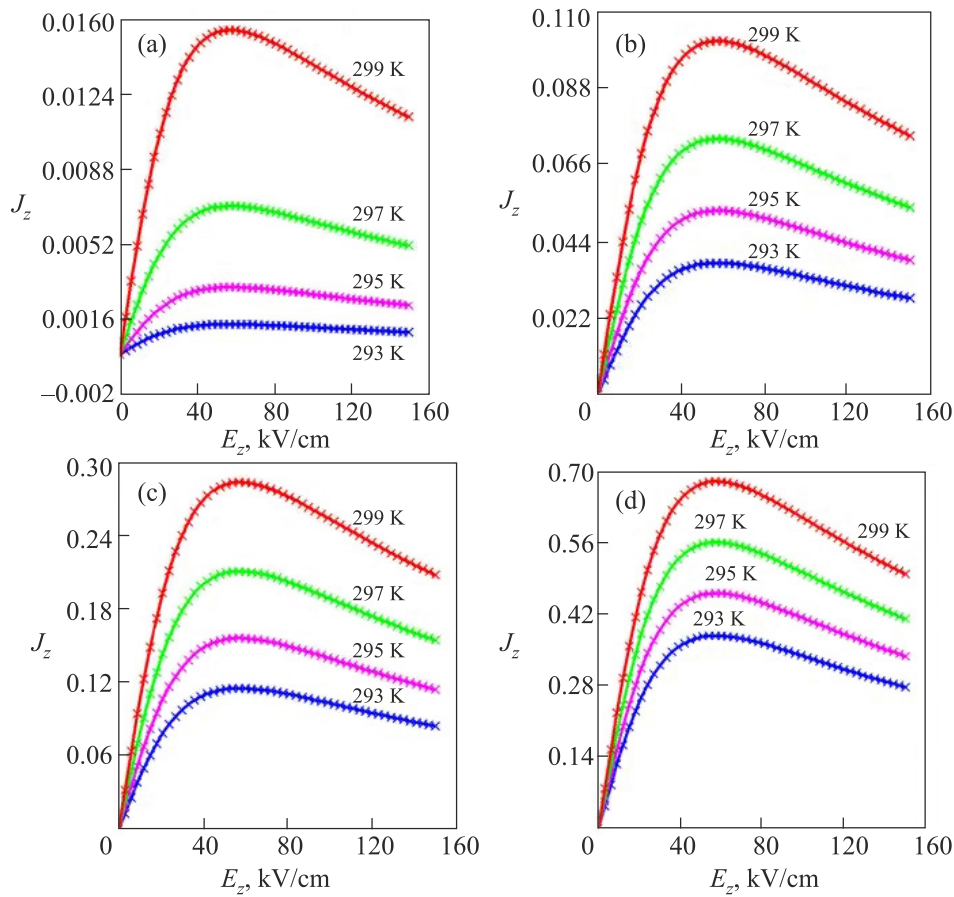


Fig. 1. (Color online) Plots of normalized current density J_z versus electric field E_z showing increasing electrical conductivity $|\partial J_z / \partial E_z|$ as the room temperature increases from 293 to 299 K for semiconducting zigzag: (a) (2, 0), (b) (4, 0), (c) (5, 0), and (d) (7, 0) CNTs, $\nu = 1$ THz.

and the electrical conductivity at any dc field E_z represented by the tangent to the curve at that particular dc field E_z for either Ohmic or NDC regions increases as the temperature decreases from 293 to 153 K for metallic zigzag CNTs. This could be attributed to the fact that at a lower temperature the rate of vibration of the lattice decreases leading to decrease in the scattering rate of electrons which results in increase of

the electrical conductivity in metallic zigzag CNTs as temperature decreases. Hence for metallic zigzag CNTs, as the temperature is reduced to a very low value, the conductivity increases and this behavior could probably lead to the superconductivity property of metallic which occurs at very low temperatures.

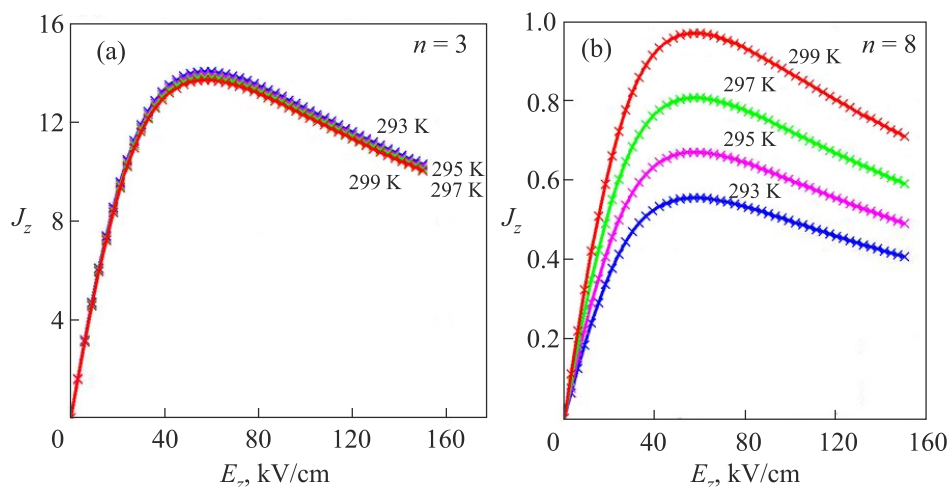


Fig. 2. (Color online) A plot of normalized current density J_z versus electric field E_z showing increasing electrical conductivity as the room temperature increases from 293 to 299 K for (a) metallic zigzag (3, 0) CNT, (b) semiconducting zigzag (8, 0) CNT, $\nu = 1$ THz.

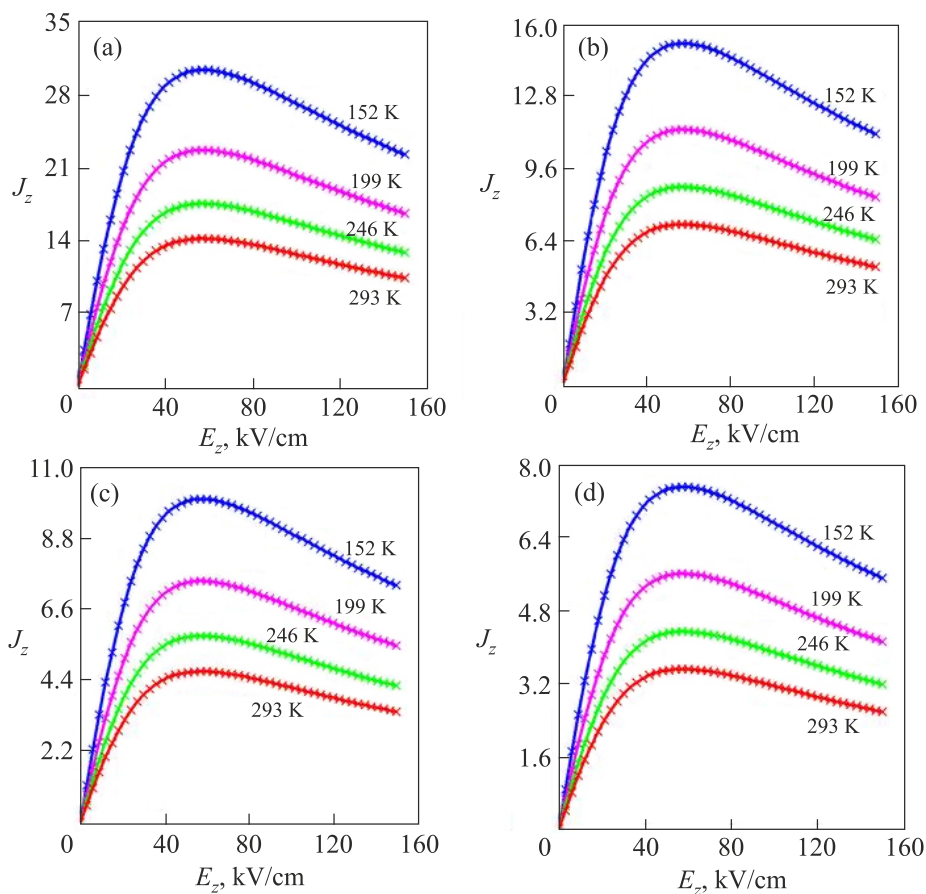


Fig. 3. (Color online) A plot of normalized current density J_z versus electric field E_z showing increasing electrical conductivity as temperature decreases from room temperature of 293 K to a very low of 153 K for metallic (a) (3, 0), (b) (6, 0), (c) (9, 0), and (d) (12, 0) zigzag CNTs, $\nu=1$ THz.

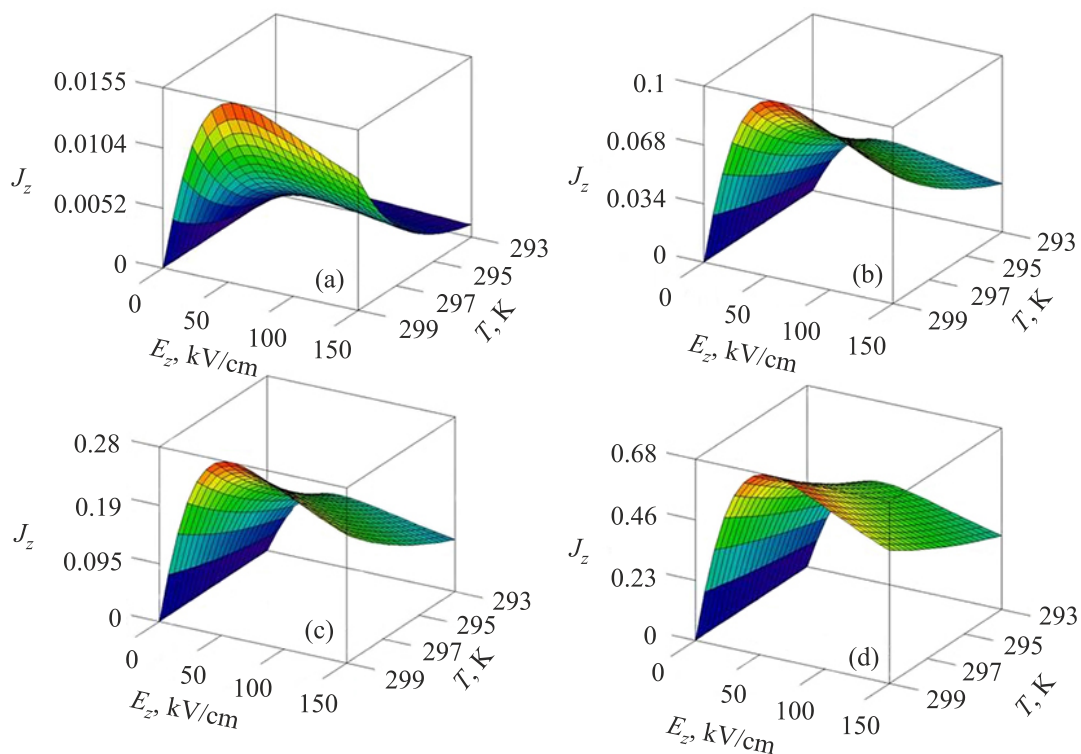


Fig. 4. (Color online) 3D plots of normalized current density J_z versus dc field E_z and temperature T for semiconducting zigzag: (a) (2, 0), (b) (4, 0), (c) (5, 0), and (d) (7, 0) CNTs, $\nu=1$ THz.

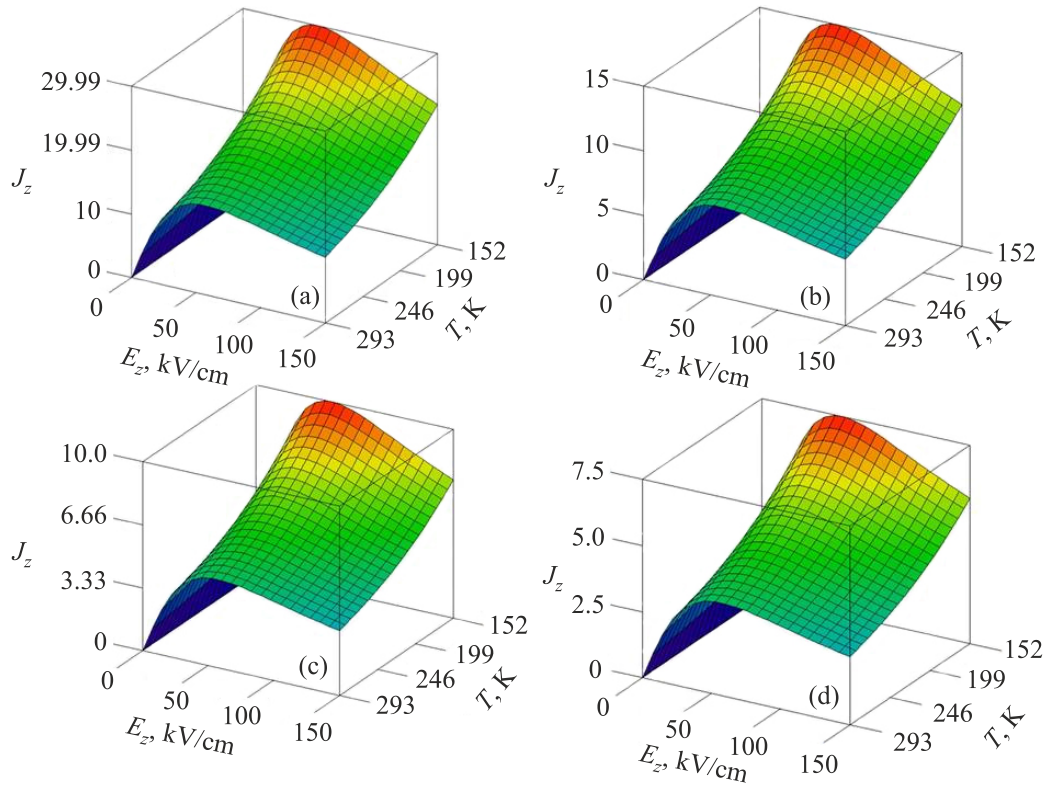


Fig. 5. (Color online) 3D plots of normalized current density J_z versus dc field E_z and temperature T for metallic zigzag: (a) (3, 0), (b) (6, 0), (c) (9, 0), and (d) (12, 0) CNTs, $\nu = 1$ THz.

To put the observed influence of temperature variation on electrical conductivity of zigzag carbon nanotubes under dc field in perspective, the 3-dimensional behavior of the normalized current density J_z as a function of the dc field E_z and the temperature T for semiconducting and metallic zigzag CNTs are displayed in Figs. 4 and 5, respectively.

In each of Fig. 4, the differential conductivity $|\partial J_z / \partial E_z|$ and the peak normalized current density are at the lowest values when the room temperature is relative low (i.e., $T = 293$ K). For each semiconducting zigzag (2, 0), (4, 0), (5, 0), and (7, 0) CNTs, as the temperature gradually increases, the differential conductivity and the peak current density increase until the highest values are obtained at relatively high room temperature of 299 K as shown in Fig. 4.

Unlike semiconducting zigzag CNTs, the electrical conductivity for metallic zigzag CNTs as the temperature changes is quite different in that the differential conductivity and peak normalized current density increase with decreasing temperature from 293 to 152 K. In each of Fig. 5, the differential conductivity $|\partial J_z / \partial E_z|$ and the peak normalized current density are at the lowest values when the temperature is relatively high (i.e., $T = 293$ K). For each metallic zigzag (3, 0), (6, 0), (9, 0), and (12, 0) CNTs, as the temperature gradually decreases, the electrical conductivity and the peak normalized current density increase until the highest values are obtained at the relatively low temperature of 152 K as shown in Fig. 5.

Conclusion

In conclusion, the influence of temperature variation on the electrical conductivity of zigzag carbon nanotubes (CNTs) has been studied theoretically using a semiclassical approach. A higher significant increase of electrical conductivity in semiconducting zigzag CNTs has been observed whereas a negligible decrease in metallic zigzag CNTs has been also noticed as the room temperature increases from 293 to 299 K. Also, the electrical conductivity of each metallic zigzag CNT increases immensely as the temperature is reduced to a very low value which could lead to potential superconductivity property which occurs at very low temperatures. Therefore, the study predicts semiconducting zigzag CNT as potential material for temperature sensors since they exhibit a faster response and a substantially higher sensitivity to room temperature changes than metallic CNT. Furthermore, metallic zigzag CNTs could probably exhibit superconductivity behavior since their conductivity increases immensely at very low temperatures.

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Вплив зміни температури на електропровідність
зигзагоподібних вуглецевих нанотрубок
в однорідному осьовому постійному
електричному полі

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Представлено теоретичні дослідження впливу температурних змін на електропровідність зигзагоподібних вуглецевих нанотрубок (ВНТ) під дією однорідного осьового електричного постійного поля. Рівняння Больцмана використано для визначення щільності струму зигзагоподібних ВНТ як функції однорідного осьового постійного поля та температури. Графіки нормованої щільності струму в залежності від однорідного постійного поля, яке прикладене вздовж осі напівпровідникових зигзагоподібних ВНТ, при підвищенні кімнатної температури з 293 до 299 К показали значне збільшення електропровідності, тоді як в металевих зигзагоподібних ВНТ вона практично постійна або незначно зменшується. Передбачається, що напівпровідникові зигзагоподібні ВНТ можуть стати потенційним матеріалом для датчиків температури, оскільки демонструють більш швидкий відгук та значно вищу чутливість до змін кімнатної температури, ніж їх металеві аналоги. Електрична провідність металевих зигзагоподібних ВНТ значно збільшується при зниженні температури до дуже низького значення, що, ймовірно, може привести до надпровідності, яка зазвичай виникає при дуже низьких температурах. Ці потенційні датчики температури та надпровідники з наноматеріалів знаходять широке застосування в сучасній науці та техніці.

Ключові слова: вуглецеві нанотрубки, провідність, зміна температури, щільність струму, постійне поле.