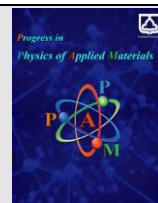




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Electron Gas Hardness of Individual Carbon Nanotubes

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ABSTRACT

Experimental results show that there are uninterpreted physical phenomena in the resistivity behavior of carbon nanotubes (CNT) in terms of their diameter changes. In this paper, a model based on previously published empirical data is created. This model is used later to analysis the effect of repulsion on electron transport throughout CNT. The relationship between the resistivity and the diameter of CNT, with an introduced parameter named 'electron gas hardness' has theoretically investigated. The results show an acceptable theoretical model for the behavior of electrical resistivity to reduce the diameter of nanotubes and is predicted by physico-mathematical calculations. Furthermore, a detailed analysis of the temperature effects on the transport properties in CNT and how compare to electron-phonon interactions that have been shown to affect resistivity and a theoretical model of electrical resistivity to changes of two important parameters of diameter and temperature of carbon nanotubes, physical formulation and modeling is presented. These results are consistent with the experimental results and are generalized.

1. Introduction

Since Iijima's discovery of carbon nanotubes (CNTs) [1-3], nanoscale graphite structures and CNTs have been studied not only for their mechanical and thermal properties [4-9], but also for their electrical properties. There has been considerable interest in the electrical conductivity of most specific nanotubes [10-14]. The electrical conductivity of CNT depends on the structure of the graphite. Nanotubes can be conductor (metal nanotubes) or semiconductors, depending on the diameter and the angle of the nanotube lattice to its axis (chirality vector) [15-18]. Thus, it varies greatly from batch to batch depending on the angle and type of bonds [19-22]. Each atom in a nanotube vibrates in its position, and when electrons (or electric charge) enter a set of atoms, the vibrations of those atoms increase and the electric charge is transmitted from one atom to the other one as they collide with each other. The higher the order of the atoms, the greater the electrical conductivity of those nanotubes [23, 24]. A semiconductor properties depend on its type. For example, the electrical conductivity of the nanotube armchair type is about thousand times more than that of copper [25-27]. CNTs have unique and interesting properties. Among their properties, electrical properties have been vastly studied [28, 221].

In 1994, for the first time, Langer and his coworkers succeeded to measure the electrical resistance of CNTs [32]. In 1998, Frank and his colleagues conducted experiments on the electrical conductivity of nanotubes [33]. Their results showed that the nanotube behaved like a ballistic conductor that is quantized. Later, in 2000, Sanvito et al. used a scattering method to calculate the quantum ballistic conductivity of CNTs [34]. They found that their nanotube samples had unexpected electrical conductivity values, which explained by Frank explanations. Thess et al. calculated the resistivity of the ropes at about $10^{-4} \Omega \cdot \text{cm}$ at 300 K [35]. They did this by directly measuring the resistance using the quadrupole probe. Using the same electrical conductivity, Frank [33] achieved a current density of 10^7 A/cm^2 . Then, Collins et al. [36] stated that the steady-state current density of the nanotube can reach 10^{13} A/cm^2 and it is capable of withstanding such extremely high current density. Dai et al. [37], as well as Ebbesen et al. [38], experimentally measured the dependence of the resistivity on the diameter of the individual nanotubes and showed that the nanotube resistivity decreased smoothly with decreasing its diameter. However, in the above mentioned experimental observations, the resistivity changes as a function of CNT

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diameter but no physical justification has been provided so far. Many different parameters influence on the CNTs resistivity. In this paper, electron gas repulsion phenomenon, is used to justify the variations of the resistivity of CNTs with respect to their diameters.

2. Modelling

Before presenting the model, some phenomena related to the present investigation are necessary to discuss.

2.1. Electron gas repulsion

Electrons can be considered as Two-dimensional electron gas (2DEG) existing across the axial area of the CNT. The graphene comparison we make involves confinement of the electron motion to a single plane (thus 2D), however in the case of CNT, one would expect 1D confinement, i.e. motion along the axis of the nanotube. The idea then is electrons have relaxed behavior in directions perpendicular to the CNT length, which decreases resistivity through repulsion.

In the author's opinion, the electric current on a graphene sheet moves in a state of equilibrium as an electron and plasmonic gas and because the graphene is a flat plate, half of the electron gas lies above the graphene sheet and the other half lies below it [39-41]. This is shown schematically in Fig. 1.

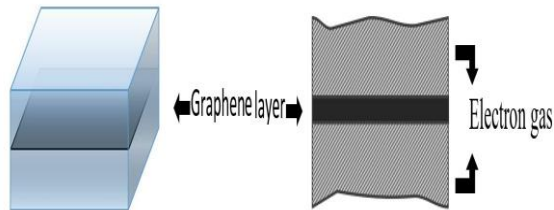


Fig. 1. The schematic shows the presence of electron gas on both sides of the graphene sheet.

The electron gas is formed of weakly bound electrons and it is confined in the crystal. Such electrons are attached to the atoms but not completely bound to them, so they are a bit free in the phonons. Now if this graphene sheet is rolled into a nanotube [42-44] (Fig. 2), the electric repulsion effect of the electron gas appears on the wall of the formed nanotube.

It is evident the large diameter CNTs have constant electrical resistivity values and repulsion effect becomes effectless [45, 46], but this will not be the case for diameters less than 20 nm [37]. Experimental results of electrical resistivity of carbon nanotubes in terms of diameter at room temperature are shown in figure 3 [37]. As shown in the figure, the resistivity behavior of a carbon nanotube varies with its diameter.

As shown in figure 3, the experimental results present two important phenomena. The first one is the decrement of the nanotube diameter from 20nm to 14nm leading to a reduction in the resistivity. As a result, part of the electron gas is forced out of the CNT wall due to electrostatic repulsion that is illustrated in figure 2. The second phenomenon is observed when the diameter of the nanotube is decreased from 14nm to 8nm which causes a fluctuation in the resistivity of CNT. We suggest that this

fluctuation is due to chirality phenomena (Chirality Surfaces Overlap).

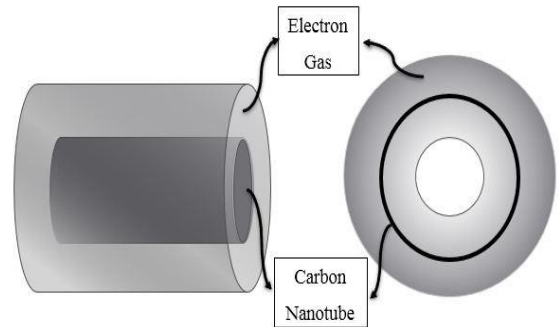


Fig. 2. Scheme showing the uniform distribution of electron gas on inner and outer walls of carbon nanotubes (half of the electron gas inside and half of it outside the nanotube).

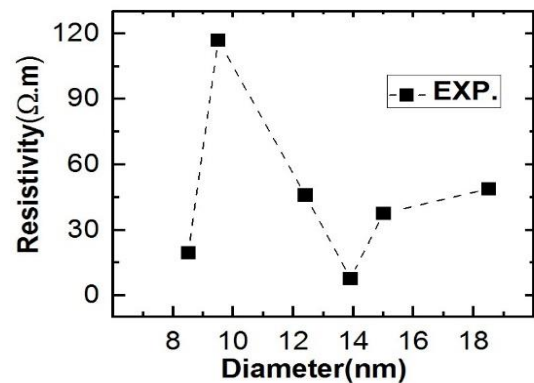


Fig. 3. Experimental results of electrical resistivity of carbon nanotubes in terms of diameter at room temperature [37].

Reduction of carbon nanotube diameter results the increment of electrostatic repulsion effect. This in turn affects the symmetry of electron gas distribution in such a way that the distribution of electron gas on both sides of the CNT wall will no longer be equal. It is rather, it is likely to be distributed more on the outer side of the CNT wall than on its inner one (Fig. 4).

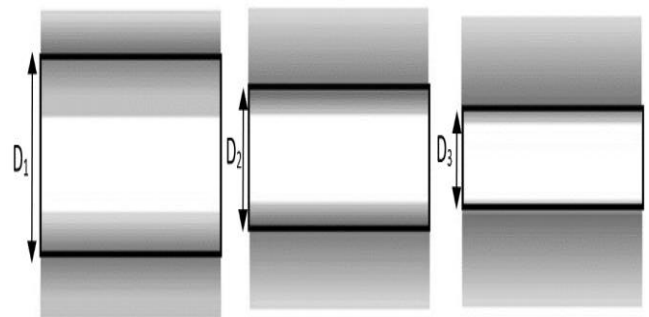


Fig. 4. Movement of electron gas out of the carbon nanotube wall by electrostatic repulsion as the diameter decreases.

As the diameter of the nanotube decreases, the electron gases on the CNT cylindrical wall repel each other, and the mean free path between the collision of electrons and phonons increases. Then the electron gas becomes relaxed in directions perpendicular to the CNT length, and hence this relaxed electron gas has an effect on transport across the CNT. Therefore, by reducing the nanotube diameter, the

effect of electron repulsion will reduce the CNT electrical resistivity. Fig. 4 summarizes the repulsion of electron gas due to diameter reduction. As known the resistivity is given by [47]:

$$\rho = \frac{m^*}{n e^2 \tau} \quad (1)$$

Where m^* is the effective mass of electron, n is the charge density, e is the electric charge, and τ is the mean free time that represents the time interval between two successive collisions. τ is the principal parameter that determines the electrical resistivity ρ . Since the dependence of the electron gas on the phonon (lattice) decreases due to the repulsion phenomenon of the walls, τ increases, and as a result, the electrical resistivity ρ decreases. So when the diameter of individual carbon nanotube decreases, τ increases, which plays a key role in determining the behavior of electrical resistivity.

Although, the repulsion of the electron gas can change the electron density but it is not so significant, hence this change is not taken into account. This effect is very weak in multiwall CNT due to the presence of multiple repulsion instances across concentric electron gases and electron gas seem to be heavy.

2.2. Formulation

As mentioned, according to Eq.1. The main influencing parameter on the resistivity of an individual wall carbon nanotube is the mean free time (τ). Let's suppose that decreasing the CNT diameter by Δx will

increase the mean free time by $\Delta\tau$. In first regard, one can suggest that:

$$\Delta x(\text{diametr variation}) \propto \frac{1}{\Delta\tau(\text{mean free times variation})} \quad (2)$$

Since the above relation did not lead to a satisfactory result, the following form was employed:

$$\Delta x \propto -\Delta\tau \quad (3)$$

In the following, we explain the origin of the negative sign in Eq.3.

Analogical reasoning of Eq.3 is observed in classical mechanic physics about relation between the variation of kinetic energy (K) and potential energy (U):

$$\Delta K = -\Delta U \quad (4)$$

That,

$$K_f - K_i = -(U_f - U_i)$$

$$\text{If } \Delta K > 0 \quad K_f > K_i \rightarrow U_f < U_i$$

f and i, are the indexes of final and initial value of the corresponding parameter respectively.

The significance of negative sign in Eq. 4 lies in the fact that, increment of final kinetic energy is due to decrement of final potential energy.

For clarification, let's consider the following example of Eq. 3:

$$\Delta x \equiv -\Delta\tau \quad \text{example} \quad \left\{ \begin{array}{l} \Delta x > 0 \left\{ \begin{array}{l} (x - x_0) \approx -(\tau - \tau_0) \\ (60 - 54)nm \approx -(12 - 16)\mu s \\ +6nm \approx +4\mu s \\ \Delta x \text{ increment to } 6nm \text{ causes } \Delta\tau \text{ decrement to } 4\mu s. \end{array} \right. \\ \Delta x < 0 \left\{ \begin{array}{l} (x - x_0) \approx -(\tau - \tau_0) \\ (40 - 54)nm \approx -(25 - 16)\mu s \\ -14nm \approx -9\mu s \\ \Delta x \text{ decrement to } 14nm \text{ causes } \Delta\tau \text{ increment to } 9\mu s. \end{array} \right. \end{array} \right. \quad (5)$$

Considering Eq.3, one may rewrite Eq.1 in the following form:

$$\Delta x \approx -\left(\frac{m^*}{ne^2 \rho} - \frac{m^*}{ne^2 \rho_0}\right) \approx -\frac{m^*}{ne^2} \left(\frac{1}{\rho} - \frac{1}{\rho_0}\right) \approx \frac{m^*}{ne^2 \rho_0} \frac{\Delta\rho}{\rho} \approx \tau_0 \frac{\Delta\rho}{\rho} \quad (6)$$

ρ_0 is the electrical resistivity of the normal state or the saturated state, i.e. when there is no repulsion of electronic gas. In Eq. 6, the ratio of changes in electrical resistivity to resistivity is proportional to the variation of carbon nanotube diameter.

Here, ρ_0 is the electrical resistivity of the normal state or the saturated state, i.e. when there is no repulsion of electronic gas.

Eq. 5 can be extended to the following relations:

$$\frac{\Delta\rho}{\rho} \tau_0 \approx \Delta x \rightarrow \frac{\Delta\rho}{\rho} \approx \frac{1}{\tau_0} \Delta x \rightarrow \frac{\Delta\rho}{\rho} = \gamma \Delta x \quad (7)$$

The γ parameter is considered as a proportionality constant assuming $\frac{1}{\tau_0} \approx \gamma$.

where γ is the proportionality constant that will be explained physically later in this section. Considering infinitesimal change in CNT diameter, the above relation can be written in the following form:

$$\int_{\rho_{tv}=\rho_0-\rho}^{\rho_0} \frac{d\rho}{\rho} = \gamma \int_{x_0}^x dx \tag{8}$$

Here, at large CNT diameter (x), the resistivity has its normal value (ρ_0), i.e., electron gas repulsion phenomenon does not occurred. For $x = x_0$ (x_0 is the diameter at which the electrical resistivity of a carbon nanotube is normally close to zero) the resistivity has a trifling value ρ_{tv} .

Now one can simply solve Eq. 8 as following:

$$\ln \rho \Big|_{\rho_0-\rho}^{\rho_0} = \gamma(x-x_0) \rightarrow \frac{\rho_0}{\rho_0-\rho} = e^{\gamma(x-x_0)} \tag{9}$$

The above relation may be written in the following form:

$$\rho = \rho_0(1 - e^{-\gamma(x-x_0)}) \tag{10}$$

Eq.10 shows the carbon nanotube resistivity as a function of its diameter. It is the basic equation presented by the proposed theoretical model. Fitting this equation to the experimental results gives the value of γ parameter that depends on the lattice phonon.

After fitting using the experimental data available in Ref. [37], one finds:

$$\rho = 49.1(1 - e^{-1.16(x-13.75)}) \tag{11}$$

The values ρ_0 and x_0 in equation 9 are replaced with experimental values from Dai et al. (Ref 37). It is worth mentioning that the values, 49.1 $\Omega.m$ and 13.75 nm for ρ_0 and x_0 respectively, have been calculated by the model based on data presented in Ref. [37].

Each one of the ρ_0 , x_0 , and γ parameters expresses a specific physical meaning. For example, for $x_0= 13.75$ nm, the electrical resistivity of carbon nanotube is close to zero. This means that (at the critical diameter, the contribution of the electron gas repulsion to reducing the resistivity is negligible), and there is almost no electron-phonon interaction.

Therefore, $x_0=13.75$ nm is defined as the critical diameter (D_c). ρ_0 , when the effect of electron gas repulsion is not yet present, is found to be 49.1 $\Omega.m$. The value of γ is found to be 1.16 nm^{-1} and varies from substance to substance.

While the experimental results [37] provides no data concerning the CNT electrical resistivity beyond a diameter of 18.5nm, the present proposed theoretical model states that at large diameters, greater than 20nm, the electrical resistivity reaches a saturation value (49.1 $\Omega.m$), and the repulsion effect of the electron gas is eliminated. In addition, the model introduces another important parameter called the critical diameter (D_c), less than which the electrical resistivity is zero. The experimental results [37] show a certain value of electrical resistivity for a diameter of 13.9 nm and nothing else is said if the diameter is reduced below this value, while the present model clearly

shows that if it is 13.75nm or less, the electrical resistivity becomes zero.

Finally, in this model, gamma is introduced as an important physical parameter that indicates the interaction of the electron gas or the connection of the electron gas to the phonon lattice in an individual carbon nanotube material, and its value is different from one material to another. In other words, it shows the effects of electron gas dependence on the phonon lattice or *the Electron Gas Hardness* (EGH) in an individual carbon nanotube material.

In other words, *the electron gas hardness* indicates to what extent the electron gas is bound to the phonon lattice in the carbon nanotube material. In fact, it can be said that if the γ value of a nanotube material is high, its electron gas is more dependent on the phonon lattice, and in such a condition, the electron gas may be called, "hard". Therefore, electron gas hardness is a measure implying to what extent the electron gas is dependent on the phonon lattice. In the present study, the γ value in an individual carbon nanotube is found to be 1.16 nm^{-1} .

Fig. 5 demonstrates the model of Eq. 9 with the experimental results [37]. As the figure clearly shows, the resistivity of carbon nanotubes reduces with diameter.

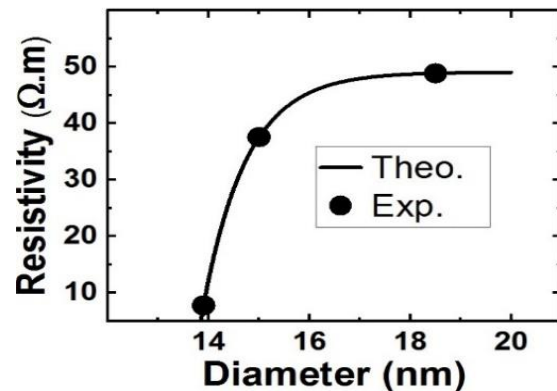


Fig. 5. CNT resistivity vs diameter. Solid curve is the theoretical result and the solid points are experimental findings [37].

It is worth noting that at high diameters of the CNT resistivity is not changing with diameter, indicating that the electron repulsion phenomenon becomes almost ineffective. Therefore, it may be deduce that any change in the electron gas density would affect the electrical resistivity of carbon nanotubes. Theoretically, there seems no phenomenon other than the electron gas repulsion one causing change in the CNT electrical resistivity.

3. Temperature Effect on the Critical Diameter value an individual Carbon Nanotube

As mentioned before, the critical diameter of an individual CNT calculated at room temperature using the proposed theoretical model and found to be 13.75 nm. It is well understood that the resistivity of all materials depends on temperature, as the resistivity of a conductor increases with temperature.

It is obvious that increasing temperature causes increment in phonon vibration amplitude and the electrons in the metal experience more collisions leading to increment of its resistivity.

Even though many studies have been performed at cryogenic and room temperatures [48], the following equation has been used for changes in CNT's temperature ($\leq 100K$) [49]:

$$\rho = \rho_0(1 + \alpha\Delta T) \tag{12}$$

Where ρ_0 is the resistivity at the reference temperature (for example $T_R = 300$ K) and α is the temperature coefficient of the resistivity.

Table 1. Resistivity values of some common metals [49].

| Material | $\rho_0(\mu\Omega.cm)$ | $\rho(\mu\Omega.cm)$ | $\alpha (K^{-1})$ |
|--------------------|------------------------|----------------------|-------------------|
| | at 298 K | at 373 K | |
| Silver | 1.59 | 2.35 | 0.00637 |
| Copper, Annealed | 1.72 | 2.51 | 0.00608 |
| Copper, Hard Drawn | 1.77 | 2.54 | 0.00575 |
| Aluminum | 2.28 | 3.06 | 0.00455 |
| Gold | 2.44 | 3.12 | 0.00372 |
| Tungsten | 5.60 | 6.50 | 0.00214 |
| Iron | 9.71 | 11.01 | 0.00179 |
| Nickel | 11 | 12.20 | 0.00145 |
| Steel | 10.4 | 11.40 | 0.00128 |
| Platinum | 10 | 10.60 | 0.00080 |
| Brass | 7 | 7.40 | 0.00076 |
| Nichrome | 100 | 100.08 | 0.00001 |

In the present study, the temperature coefficient of carbon nanotube material is assumed to be around $0.005 K^{-1}$, near to that of Ag, Cu, and Al as given in literature [49].

Table 1, presents resistivity values of some common metals obtained experimentally [49].

In the following, by placing the values for parameters from Eq. 10 in Eq. 11, as well as using the data given in Table 1, the value of temperature coefficient of the resistivity for individual carbon nanotubes at $T=300K$ is assumed: $\alpha = +0.005K^{-1}$.

Then, carbon nanotubes resistivity as a function of temperature can be written as:

$$\rho(T) = 49.1(1 + 0.005(T - T_{R=300K})) \tag{13}$$

α is positive for metals, meaning that their resistivity increases with increasing the temperature, while the temperature coefficient of resistivity for semiconductors is negative, which means that their resistivity decreases with increasing temperature leading to better conductivity at high temperatures. Because with increasing thermal thermal fluctuations, the number of free charge carriers to establish an electric current increases and for larger temperature changes, α may change in a nonlinear fashion and hence a nonlinear equation may be needed to find it [50,51].

Although α can behave nonlinearly but it is assumed in the present study as a linear function of temperature. That is, ρ is supposed to vary linearly with T. It is also assumed that the governing relations follow Eq. 13.

Therefore, if the CNT temperature is slightly higher than the reference temperature ($T=300K$), the phonons and their vibrations should have a greater effect, hence one

expects the previously calculated D_c , the nanotube resistivity will not be to achieve zero resistivity for CNT, the nanotube diameter has to be further decreased. This means that at higher temperatures, the curve showing the variation of CNT resistivity as a function of its diameter when compared with that shown in Fig.4 experiences a shift to the smaller diameters with higher values. The use of $49.1 \mu\Omega.cm$ as ρ_0 limits the relationship to only CNT of a certain size as it was determined to be the resistivity of CNT above a critical diameter.

On the other hand, it has been reported that at temperatures lower than 300K, CNTs with large diameters show phonon lattice resistivity close to zero [52, 53].

It seems that the first effect of CNT temperature is on the value of its critical diameter (D_c) where, there is an inverse ratio between CNT temperature and its critical diameter, as stated before. That is, with increasing temperature above room temperature $T=300K$, the critical diameter of a CNT should be less than 13.75nm.

$$D_c = x_0 = \frac{\beta}{T} \rightarrow \beta = 13.75nm \times T_R \tag{14}$$

Here β is the proportionality constant showing the dependence of the electron gas on the phonon lattice and its unit is nmK . It is clear from the above relation that at room temperature, it reduces to the value of 13.75 nm as expected. In this condition, the electron gas is not bound to the atomic lattice and causes the nanotube resistivity to be zero. It is natural that the increase in CNT temperature affects electron-phonon collisions, which results increasing the resistivity. Hence for CNT resistivity to meet zero, the critical diameter of carbon nanotube must be further reduced. Therefore, Eq. 8 is modified in other to include the temperature as a factor affecting CNT resistivity.

$$\rho = 49.1(1 - e^{-1.16(x-13.75nm \times \frac{300K}{T})}) = \rho_0(1 - e^{-\gamma(x-\frac{\beta}{T})}) \tag{15}$$

Here, at $T=300K$, Eq. 15 reduces to Eq. 10 using which the critical diameter was found to be 13.75 nm and the electrical resistivity of carbon nanotube was found close to zero. However, if the CNT temperature is increased, its electrical resistivity will no more be zero at $D_c=13.75$ nm, and the phonon vibrations disturb the Electron gas.

Therefore, one again come across the relation expressing the temperature dependence of CNT critical diameter, i.e., say that $D_c = x_0 = \frac{\beta}{T}$. This means that, the higher the temperature, the lower should be the critical diameter in order to increase the freedom of action for electrons. In other words, at temperatures higher than room temperature, the repulsion of electron gas is more effective.

Replacing Eq. 10 in Eq. 15, the following equation is obtained:

$$\rho(D,T) = \rho_0(1 + \alpha(T - T_R))[1 - e^{-\gamma(D-\frac{\beta}{T})}] \tag{16}$$

Inserting the already found data, Eq. 16 may be given in the following form:

$$\rho(D,T) = 49.1(1 + .005(T - 300)) \left[1 - e^{-1.16(D - \frac{4125}{T})} \right] \quad (17)$$

Fig. 6 is a 3D sketch of the electrical resistivity of a CNT as a function of its temperature and diameter. In the present theoretical investigation, the temperature of CNT is changed from 200K to 400K and its diameter is changed from 0 to 100 nm.

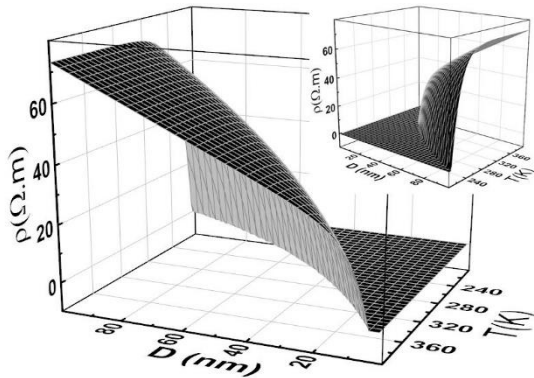


Fig. 6. Electrical resistivity of individual CNT as a function of temperature and diameter.

It is clear from Fig. 6 that at a certain temperature, as the diameter of the CNT increases, its electrical resistivity also increases (from zero to 65.73 Ω.m). The reason for an upper limit of ρ value may be attributed to the condition at which the repulsion effect of electron gas no longer occurs and hence the resistivity is said to be saturated. At lower temperatures, this effect weakens the repulsion of electron gas. The bottom of the diagram also shows the diameters and temperatures at which the resistivity of an individual carbon nanotube is zero. For example, whenever the temperature rises at a certain diameter, an electron-phonon interaction occurs in the atomic lattice and the electrical resistivity of the CNT increases. On the other hand, when the temperature and diameter of carbon nanotube increase, the electrical resistivity also increases and saturates at large diameters and high temperatures.

4. Conclusions

The electrical properties of CNTs depend on the structure of graphite. Nanotubes can be conductor (metal nanotubes) or semiconductor, depending on the diameter and chirality vector. In this paper, a theoretical model based on the electrical resistivity behavior of individual carbon nanotubes as a function of diameter is established and presented. For this purpose, the idea of electrical repulsion of electron gas in the inner wall of carbon nanotubes and its effect on the electrical resistivity behavior of CNT due to the reduction in its diameter was used. In this way, a formula describing the impact of CNT diameter on their electrical resistivity was introduced, using which the electrical resistivity behavior of an individual CNT was modeled theoretically and found in good agreement with the experimental results. Fitting presented model to the experimental results led to introducing electron gas hardness, a parameter indicating the extent the electron gas is bound to the phonon lattice in the carbon nanotube material. In addition, the critical diameter $D_c = 13.75$ nm

was defined, at which the electrical resistivity (ρ) of the nanotube becomes zero. Further, it was shown that ρ as a function of CNT diameter decreases with decreasing the diameter due to the electron gas repulsion phenomenon. It was also shown that at temperatures other than room temperature, the resistivity of carbon nanotube with diameter of 13.75nm would be no longer zero as the phonon vibrations disrupt the movement of electrons. Therefore, it was concluded that the most effective parameters affecting on the CNT electrical resistivity are its diameter and the temperature. It was shown that one may expect that the critical diameter at which the electrical resistivity approaches zero would be smaller at room temperature than at higher temperatures. The model can be used for nanotubes as long as these nanotubes are so far apart that they do not affect each other (Individual Carbon Nanotubes). Hence the model cannot be used for multi-wall nanotubes because the connection between the walls is weak and of the Van der Waals type (sliding of the walls on each other). Finally, the theoretical model presented in this investigation was shown to be consistent with the existing empirical reports.

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