Length-dependent resistance model for a single-wall Carbon nanotube

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The non-linear length-dependent resistance, \( R(l) \) observed in single-wall Carbon nanotubes (SNTs) is explained through the recently proposed ionization energy (\( E_I \)) based Fermi-Dirac statistics (iFDS). The length here corresponds to the Carbon atoms number (\( N \)) along the SNT. It is also shown that \( R_y(l_y) < R_x(l_x) \) is associated with \( E_I^y < E_I^x \), which can be attributed to different conducting properties in their respective \( y \) and \( x \) directions, or due to chirality.

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I. INTRODUCTION

Enormous amount of research have been poured since the discovery of Carbon (\( C \)) nanotubes (CNTs) by Iijima \textsuperscript{1} in 1991 and consequently, CNTs have been successfully exploited to produce cathode ray tubes \textsuperscript{2} and nano-electronic devices \textsuperscript{3}. Understandably, CNTs are believed to pave the pioneering pace for the nanotechnology boom. Basically, \( C \) can be categorized into graphite, diamond and Fullerenes based on their bonding nature that gives rise to different electronic and structural properties. Unexpectedly, \( C \) in all these three structures with slight manipulations have exposed superconductivity \textsuperscript{4}--\textsuperscript{6}. CNTs’ electronic properties are equivalent to rolled-graphite \textsuperscript{7}--\textsuperscript{10} which also reveal superconductivity in the absence of doping \textsuperscript{11}--\textsuperscript{13} and concentration-dependent non-linear optical properties. The real part of third-order non-linear susceptibility, \( \Re \chi^{(3)} \) was found to be in the order of \( 10^{-11} \) esu for multi-wall CNTs by Elim’s group \textsuperscript{14}. This value is roughly \( 100\times \) larger than that of SNTs, which is due to SNT’s lower \( C \)-atom concentration. The superconducting properties of Boron-doped diamonds \textsuperscript{15} based on resonating-valence-bond mechanism was put forward by Baskaran \textsuperscript{4}--\textsuperscript{5} whereas, the superconducting Fullerenes and its non-linear optical properties have been discussed by Cohen \textit{et al.} \textsuperscript{16} and Elim \textit{et al.} \textsuperscript{17} respectively.

Here, the ionization energy based Fermi-Dirac statistics (iFDS) is employed to derive the length-dependent resistance model, \( R(l) \). The derivation of iFDS and its applications in a wide variety of strongly correlated electronic matter is given in the Refs. \textsuperscript{18}--\textsuperscript{21}. This model is shown to be viable in addressing the recent \( R(l) \) observation reported by de Pablo \textit{et al.} \textsuperscript{22}, Adriotsis \textit{et al.} \textsuperscript{23} and Purewal \textit{et al.} \textsuperscript{24} in CNTs. The length-dependent resistance is an intrinsic property basically because the contact resistance is independent of CNT’s length \textsuperscript{22}. As a consequence, the only questionable result is the magnitude of the resistance, not its length-dependent trend. However, other measurements namely, the temperature(\( T \))-dependent electrical or heat conductance are strongly influenced by the contact resistance due to its own \( T \)-dependence and its large magnitude, usually in the order of the CNTs resistance, which in turn waver the intrinsic experimental \( R(T) \) results. It is interesting to note that the resistance of a SNT is non-linearly proportional to the tube’s length in both metallic and semiconducting SNTs \textsuperscript{24}--\textsuperscript{27} at any given \( T \). However, the calculations carried out by Zhang \textit{et al.} \textsuperscript{24} and Uryu \textit{et al.} \textsuperscript{28}--\textsuperscript{29} for metallic CNTs indicate that the resistance is inversely proportional to the length as a result of resonant tunnelling at interface. In this work, we do not consider heterostructures with resonant tunneling, but rather, on intrinsic metallic and semiconducting SNTs. The resistance model derived here are also suitable in other strongly correlated nanotubes that allow direct-current resistance and/or polarization measurements, or if the \( C \) atoms in CNTs are doped substitutionally with different atoms.

II. THE LENGTH-DEPENDENT RESISTANCE MODEL

We start with the many-body Hamiltonian \textsuperscript{30}--\textsuperscript{31},

\[-\frac{\hbar^2}{2m}\nabla^2 \varphi = (E + V(r))\varphi, \tag{1}\]

of which,

\[\hat{H}\varphi = (E_0 \pm \xi)\varphi. \tag{2}\]

From Eq. (2), one can notice that the influence of the potential energy on the total energy has been conveniently parameterized as \( \xi \). This energy function, \( \xi \) can be characterized in such a way that \( E_0 \) is the total energy, \( E \) at \( T = 0 \). Add to that, from Eq. (2), it is obvious that the magnitude of \( \xi \) is given by \( \xi = E_{\text{kin}} - E_0 + V(r) \), \( E_{\text{kin}} \) denotes the kinetic energy. Physically, \( \xi \) implies the energy needed to overcome the potential energy that exists in a particular system. That is, \( \xi \) is the energy needed
to excite a particular electron to a finite distance, \( r \), not necessarily \( r \to \infty \). Literally, this is exactly what we need to know in any condensed matter that actually or reasonably defines the properties of the fermions. \( \hat{H} \) is the usual Hamilton operator, \( \varphi \) denotes the many-body eigenstate and \( E_0 \) is the total energy at \( T = 0 \). The + sign of \( \pm \xi \) is for the electron \( (0 \to +\infty) \) while the − sign is for the hole \( (−\infty \to 0) \). In addition, we define the ionization energy in a many-atom system, \( \xi = E_I^{\text{real}} \) is approximately proportional to \( E_I \) of an isolated atom or ion. We can prove the validity of Eq. (2) by means of constructive (existence) and/or direct proofs as given in Ref. [31]. However, for an isolated atom, \( \xi \) is given by

\[
\pm \xi = E_{\text{kin}} - E_0 + V_{\text{pot}} = \pm E_I. \tag{3}
\]

The corresponding total energy is

\[
E_0 \pm \xi = E_{\text{kin}} + V_{\text{pot}} = E_0 \pm E_I. \tag{4}
\]

On the other hand, for an atom in a many-atom system, we can rewrite Eq. (3) as

\[
\pm \xi = E_{\text{kin}} - E_0 + V_{\text{pot}} + V_{\text{many-body}} = E_I + V_{\text{many-body}} = \pm E_I^{\text{real}}. \tag{5}
\]

Note here that \( V_{\text{pot}} \) is the atomic Coulomb potential, while \( V_{\text{many-body}} \) is the many body potential averaged from the periodic potential of the atomic arrangement. The corresponding total energy from Eq. (5) is given by

\[
E_0 \pm \xi = E_{\text{kin}} + V_{\text{pot}} + V_{\text{many-body}} = E_0 \pm E_I + V_{\text{many-body}} = E_0 \pm E_I^{\text{real}}. \tag{6}
\]

In this case, \( E_I^{\text{real}} \) is the ionization energy of an atom in a many-atom system (not isolated). The exact values of \( E_I \) are known for an isolated atom. As a consequence, we can arrive at Eq. (2) from Eq. (6). Appropriately, we cannot use Eq. (2) to isolate the electronic and phonon contributions because we have defined the \( \xi \) as a function of the Coulomb potential \( (V_{\text{pot}}) \), many-body \( (V_{\text{many-body}}) \) and kinetic \( (E_{\text{kin}}) \) energies. Consequently, the total energy can also be rewritten as (from Eq. (6))

\[
E = E_0 \pm \sum_{i} \sum_{j} E_{Ii,j}^{\text{real}}, \tag{7}
\]

where, \( j \) is the sum over the constituent elements in a particular compound. For a C nanotube with only one type of atom, Eq. (7) can be rewritten as

\[
E = E_0 \pm \beta \sum_{i} E_{IIi}. \tag{8}
\]

In Eq. (8), we have defined here that \( \beta = 1 + \frac{\langle V(r) \rangle}{E_I} \), where \( \langle V(r) \rangle \) is the averaged many-body potential value. Apart from that, the total energy equation for a free-electron system is given by

\[
E = E_0 \pm \sum_{i} \sum_{j} E_{Ii,j}^{\text{real}} = E_0 \pm [E_{\text{kin}} - E_0 + V_{\text{pot}} + V_{\text{many-body}}] = E_{\text{kin}} + V_{\text{pot}} + V_{\text{many-body}} \iff \text{for electrons } \pm \to + \implies E_{\text{kin}} \iff \text{implies free electrons.} \tag{9}
\]

In Eq. (9) we have substituted Eq. (5) for \( E_I^{\text{real}} \) because the concept of ionization energy is irrelevant for free-electron metals, which do not require excitations from its parent atom to conduct electricity. As such, the carrier density is independent of temperature and the scattering rate is the one that determines the resistivity with respect to temperature, impurities, defects, electron-electron and electron-phonon interactions. In summary, the total energy from Eq. (2) carries the fingerprint of each \( \xi \) atom in a nanotube and it refers to the difference in the energy levels of each atom rather than the absolute values of each energy level (eigenvalues) in each atom. Hence, the kinetic energy of each electron from each atom will be captured by the total energy and preserves the atomic level electronic-fingerprint in the nanotube. Using this newly defined total energy, we can derive the ionization energy based Fermi-Dirac statistics (ifDS) as given below [18]

\[
f_e(E_0, \xi) = \frac{1}{e^{[(E_0+\xi)-E_F^0]/k_BT}+1},
\]

\[
f_h(E_0, \xi) = \frac{1}{e^{[(E_0-\xi)-E_F^0]/k_BT}+1}. \tag{10}
\]

where, \( E_F^0 \) is the Fermi level at \( T = 0 \) and \( k_B \) is the Boltzmann constant. However, substituting the same atom in a nanotube gives rise to the influence of many-body \( V(r) \) and in reality, \( E_I^{\text{real}} \) cannot be evaluated from Eq. (9). Nevertheless, the \( E_I^{\text{real}} \) of an atom or ion in a nanotube is proportional to the isolated atom and/or ion’s \( E_I \) as given in Eq. (8). It is this property that enables one to predict the variation of fermionic excitation probability in C nanotubes. Therefore, one can employ the experimental atomic spectra to estimate, \( \xi = E_I^{\text{real}} \propto E_I \). It is emphasized here that \( E_I \) is zero for Boltzmann particles. As such, one should not assume that the above approximation should give the Boltzmann distribution function (BDF) as a classical limit. One can indeed arrive at BDF by first denying the additional constraint by
substituting \( E_I = 0 \). Importantly, Eq. (10) is the Fermi-Dirac statistics derived specifically for strongly correlated matter, where it is not applicable for free-electron system (or Fermi gas) as shown in Eq. (10).

Now, before we move on, let us re-examine Eq. (5) that seems to say nothing about i) the atomic arrangements and ii) how to isolate the phonon from electronic contribution. Firstly, Eq. (5) is perfectly applicable for any atomic arrangements or crystal structures. The reason is that we can incorporate Eqs. (5) and (10) for both non-bulk system, namely SNTs as well as for bulk system, regardless of its specific crystal structures, since these two equations can be normalized by employing the appropriate density-of-states (DOS). However, for non-bulk system of several atoms, including SNTs, we need to incorporate the atomic arrangement explicitly because the electronic excitation depends on the number of atoms along a certain conducting path (developed here). For bulk system with the number of atoms of the order of \( 10^{23} \), the effect of different crystal structures do not arise because the conducting paths are isotropic and the \( E_I \) here will and can be dressed accordingly to take this structural effect into account \[12 \, 19 \, 20 \, 21 \, 30 \]. For example, pure diamond and graphite will each have different valence states and electronic polarizabilities (the ability of the valence electrons to excite in a particular direction in the presence of electric field), in which, these differences are due to the different excitations of the valence electrons. These different excitations of the valence electrons are the ones that have been captured by Eq. (10) through Eq. (5). Therefore, in our approach, the true DOS and/or atomic arrangements of a particular system are unnecessary. The price we pay for this is that we cannot calculate the many-body eigenstates from Eq. (2), but note here that we can indeed prove Eq. (2) microscopically for real isolated atoms \[31 \]. In other words, our input parameter is the isolated atom-energy-level-difference, or defined here as the ionization energy \( (E_I) \). The theoretical discussion of how \( E_I \) affect the polarizability can be found in Ref. [21].

\[ \frac{dq}{dl} = n \pi de; \quad \frac{dl}{d\tau} = v_d. \]  \( \text{(12)} \)

As such, one can write the current \( (i) \) as \[32 \]

\[ i = \frac{dq}{d\tau} = \frac{dq}{dl} \times \frac{dl}{d\tau} = n \pi d e v_d. \]  \( \text{(13)} \)

Now, the resistance for a single conducting path or length, of a SNT is

\[ R = \frac{V}{i} = \frac{1}{e} \int_0^{\pi d} E \ dr, \]  \( \text{(14)} \)

\( d \) denotes the tube’s diameter and \( E = \text{electric field} \). We also know that \( m(dv_d/d\tau) = -eE \) that eventually gives \( v_d = -eE/\tau \). Finally, one can arrive at the resistance of a whole nanotube, as given below

\[ R(l) = \frac{(\pi d)^2 E}{i} = \pi d \frac{m}{ne^2 \tau} = \pi d \rho(E_I) \]

\[ = \pi d \frac{\hbar}{e^2} \left( \frac{2\pi m_e^*}{k_B} \right)^{1/2} \frac{T^{3/2}}{\exp \left[ rac{E_I - E_F^0}{k_B T} \right]} \]

\[ = \pi d A (13062) \exp \left[ \frac{E_I - E_F^0}{k_B T} \right]. \]  \( \text{(15)} \)

We have substituted, \( m/\pi e^2 \tau_{e-e} \) for \( \rho(E_I) \) and the electron-electron scattering rate, \( 1/\tau_{e-e} = AT^2 \). The
$\tau_{el,ph}$ has been neglected because SNTs are not free-electron metals, even the metallic ones. However, for heat transport, $\tau_{el,ph}$ is not negligible. $A$ is the $T$-independent scattering rate constant. The numerical value is obtained for $T=300$ K. The 1D resistivity, $\rho(E_I)$ for nanotubes can be written as

$$\rho(E_I) = \frac{\mathcal{A} \hbar}{e^2} \left( \frac{2\pi m}{k_B} \right)^{1/2} T^{3/2} \exp \left[ \frac{E_I - E_F^0}{k_B T} \right].$$ (16)

The calculated curves from Eq. (16) are shown in Fig. 1. Interestingly, one of the curve ($E_I = 150$ K) is comparable with the experimental data in Ref. [24] (see Figure 2b). The calculation of the total average ionization energy (in the respective $y$ and $x$ directions) can be carried out with

$$E_{y,x}^i[C^+] = \sum_{i} \sum_{j} \frac{E_{y,x}^i}{z} N_j.$$ (17)

Unlike ionic bulk systems, CNTs are 1D systems with anisotropic conducting paths, which have covalent bonds. Consequently, the following definitions and descriptions are essential. The $C$ in Eq. (17) represents the Carbon atom while $z$ denotes the number of valence electrons that can be excited, which will eventually contributes to the conductance of CNTs in the presence of applied voltage. Apart from $N$ (the number of $C$ atoms along a conducting path), the number of valence electron that are excited for conduction in the $y$ direction is not equal to the $x$. Meaning, the strength of the resistance or conductance in their respective $y$ and $x$ directions of a SNT originate from the inequality, $E_{y}^i(N) < E_{x}^i(N)$. The subscripts, $i = 1, 2, ..., z$ and $j$ adds the $C$ atoms, 1, 2, and so on continuously along its conducting path or length. In the previous work on superconductors and ferroelectrics [18, 19, 20, 21], Eq. (17) was simply written as the average ionization energy of a single ion as given in

$$E_I = \sum_{i} \frac{E_{y}^i}{z}.$$ (18)

The relative magnitude of $E_I$ was then calculated based on the percentage of dopant to predict the variation of $\rho(T)$ and dielectric constant. On the contrary, SNTs with finite length in nanoscale and with only one type of atoms namely, $C$ requires $E_I$ in the form of Eq. (17).

The experimentally measurable lengths (in real space) can be written as (as a function of $L$)

$$L_y = L_x = a_{c-c} \sum_{j=2}^{N} N_j - 1.$$ (19)

The $L_y$ and $L_x$ are the lengths along the $C-C$ atom’s bond. Therefore, the experimentally measurable lengths (in real space) can be written as (as a function of $L$)
\[ l_y = L_y \cos(\theta) = a_{c-e} \cos(\theta) \sum_{j=2}^{N} N_j - 1. \]  \hspace{1cm} (20)

\[ l_x = \frac{1 + 2 \cos(\gamma)}{3} L_x = \frac{a_{c-e}[1 + 2 \cos(\gamma)]}{3} \sum_{j=2}^{N} N_j - 1. \]  \hspace{1cm} (21)

Recall here that the reason \( l \) in Eqs. (20) and (21) are written as functions of \( L \) is to take into account the higher probability of electrons to conduct along the \( L \). The angles, \( \theta \) is the chiral angle, while \( \gamma = 90^\circ - \theta \), which are also defined in Fig. 2. The subscript, \( j = 2 \) indicates the sum starts from the second \( C \) atom and so on. The chiral vector, \( C_h \) is given by

\[ C_h = n a_1 + m a_2, \]

where \( a_1 \) and \( a_2 \) denote the 2D graphene lattice vectors, while \( n \) and \( m \) are integers. \( C_h \) can be related to \( x(\theta = 0^\circ) \) and \( y(\theta = 30^\circ) \) with

\[ C_h(x) = n a_1 + n a_2, \]
\[ C_h(y) = n a_1. \]

Consequently, Eq. (17) in \( y \) and \( x \) directions can be rewritten as

\[ E_i^y[C^x] = \sum_i \frac{E_i^y}{z} \left( \frac{l_y}{a_{c-e} \cos(\theta)} + 1 \right). \]  \hspace{1cm} (22)

\[ E_i^x[C^y] = \sum_i \frac{E_i^x}{z} \left( \frac{3l_x}{a_{c-e} [1 + 2 \cos(\gamma)]} + 1 \right). \]  \hspace{1cm} (23)

Now, one can actually substitute either Eq. (22) or Eq. (23) accordingly into Eq. (15) in order to obtain the length-dependent resistance. In addition, we can see that both Eqs. (22) and (23) are also determined by the chiral vectors.

III. ANALYSIS OF \( R(l) \)

The \( R(l) \) of free-electron metals with isotropic distribution of atoms and electrons can be simply derived as \( R(l) = \frac{\rho}{n \pi} \), \( S \) denotes the cross section area \([32]\). However, CNTs resistance at 300 K, say in the \( x \) direction should be written as

\[ R_x = \pi d \rho(E_l) \]
\[ = \pi d A (13062) \times \exp \left\{ \left[ \frac{3l_x}{a_{c-e} [1 + 2 \cos(\gamma)]} + 1 \right] \frac{E_l - E_F}{k_BT} \right\} \]
\[ \approx \pi d A (13062) \exp \left\{ \frac{B l_x}{T} \right\}. \]  \hspace{1cm} (24)

Equation (24) accommodates the unit for \( \rho(E_l) \), which is \( \Omega \text{ m}^{-1} \) (because the unit for 1D \( n \) from Eq. (13) is \( m^{-1} \)). The length, \( l \) varies exponentially as a result of Eq. (13). Figure 3 a) and b) indicate the influence of length on resistance via Eq. (24). The • in Fig. 3 a) and b) represent the experimental data from de Pablo et al. for the nanotube samples with diameters, \( d = 1.5 \) nm and 1.7 nm respectively. The solid lines are based on Eq. (24).

![Figure 3](image)

FIG. 3: The length-dependent resistance (\( R \)) based on Eq. (24) (solid lines) are plotted to evaluate the experimental data (•) obtained from Ref. [22].

Importantly, the fittings in Fig. 3 a) and b) clearly demonstrate that Eq. (24) gives a reasonable approximation. With this model at our disposal, one can utilize the fitting parameters namely, \( \pi d A (13062) = 37 \) k\( \Omega \) for \( d = 1.5 \) nm whereas \( \pi d A (13062) = 300 \) k\( \Omega \) for \( d = 1.7 \) nm. Therefore, \( A_{1.5} = 6.01 \times 10^8 \text{ s}^{-1} \text{K}^{-2} \) and \( A_{1.7} = 4.30 \times 10^8 \text{ s}^{-1} \text{K}^{-2} \). As a result of this, the e-e scattering rate
for 1.5 nm and 1.7 nm nanotubes are respectively given by $\tau_{e-e} = 1.85 \times 10^{-14}$ s and $\tau_{e-e} = 2.58 \times 10^{-15}$ s. Eventually, the mean free path, $l_e = v_F \times \tau_{e-e} = (8.1 \times 10^4)(1.85 \times 10^{-14}) = 15$ nm for $d = 1.5$ nm, and for $d = 1.7$ nm, $l_e = 2$ nm. Here, the Fermi velocity, $v_F$ is obtained from Ref. [12]. The other fitting parameter, $B$ for $d = 1.5$ nm and 1.7 nm are found to be 0.47 and 0.18 respectively. Throughout this resistance calculations, $e-ph$ scattering has been neglected in the usual sense, because iFDS have had the electrons dressed with $e$-ph interactions in any way. In fact, the existence of polaronic effect via $e_I$ has been discussed using iFDS. Parallel to this, Perebeinos et al. [34] and Ando [35]. Contrary to iFDS, the latter methods utilize the free-electron theory and subsequently the $e-ph$ interaction was determined in order to couple it with those free-electrons so as to describe the excitation of electrons and holes with different types of atoms. Consequently, one can notice that Eq. (23) does not ignore $e-ph$ interactions in any way. In fact, the existence of polaronic effect via $e_I$ has been discussed using iFDS. Parallel to this, Perebeinos et al. [34] have also found strong polaronic effect in SNTs as inevitable.

The properties of phonons and its influence in CNTs specifically and other nanostructures generally have been discussed extensively in the Refs. [36, 37, 38, 39, 40, 41, 42, 43]. Apart from that, Chen et al. [44] pointed out the possibility of superconductivity and ferromagnetism in SNTs doped by a chain of $C$ atoms. Whereas, Ichida et al. [45] have carried out the necessary analysis on the relaxation dynamics of photoexcited states in SNTs using femtosecond spectroscopy. They found an interesting relationship of which, the $e-ph$ interaction increases with decreasing tube diameter. Qualitatively, their result explains why for small $d$ (1.5 nm), the $B$ (0.47) determined earlier is $2.6 \times$ larger than the magnitude of $B$ (0.18), which is for large $d$ (1.7 nm). Recall here that $B$ corresponds to $E_I$, which is associated to the heavier effective mass (polaronic effect). In other words, this polaronic effect is due to the interaction between non-free-electrons and phonons, which enhances the effective mass of the charge carriers. On the contrary, for the well known $e-ph$ interaction in metals, free-electrons and phonons interact, that eventually gives rise to $e-ph$ scattering. Having said that, we can now compare our predicted values for $l_e$ (2 to 15 nm) with the values obtained by considering the short optical phonon mean-free-path (10 to 20 nm, for low bias-voltage and $d = 1.5$ to 2 nm) that limits the electrons mean-free-path.

### IV. CONCLUSIONS

In conclusion, the ionization energy based Fermi-Dirac statistics has been employed to derive the length-dependent resistance in a single-wall Carbon nanotube. It has been shown that such dependence is inevitable in a low dimensional and non-free-electron systems at nanoscales by using the the recent experimental findings. In this paper, it is also highlighted that simple equations derived using iFDS are able to capture the transport properties of single-wall Carbon nanotubes with reasonable accuracy.

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