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Small polaron hopping transport along DNA molecules

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Abstract

We present a small polaron hopping model for interpreting the strong temperature (T) dependence of the electrical conductivity, σ , observed at high (h) temperatures along DNA molecules. The model takes into account the one-dimensional character of the system and the presence of disorder in the DNA double helix. Percolation-theoretical considerations lead to analytical expressions for the high temperature multiphonon-assisted small polaron hopping conductivity, the hopping distance and their temperature dependence. The experimental data for lambda phage DNA (λ -DNA) and poly(dA)–poly(dT) DNA follow nicely the theoretically predicted behaviour ($\ln \sigma^h \sim T^{-2/3}$). Moreover, our model leads to realistic values of the maximum hopping distances, supporting the idea of multiphonon-assisted hopping of small polarons between next nearest neighbours of the DNA molecular ‘wire’. The low temperature case is also investigated.

1. Introduction

Charge transport in DNA has attracted substantial interest from both biologists and physicists. Early attempts to measure the conductivity of the DNA double helix were performed on pressed pellets [1, 2]. Recent experiments have demonstrated long range charge migration along the double helix, indicating that DNA is a candidate for being a one-dimensional (1D) molecular wire [3–6]. The understanding of the charge transfer mechanism along the DNA double helix is important in the long range chemistry of oxidative DNA damage and repair processes [7, 8], monitoring protein–DNA interactions and for possible applications in nanoelectronic circuit technology [9–15].

However, despite intensive investigation, the nature of the charge transfer mechanism remains a subject of controversy. Several mechanisms have been proposed for the interpretation

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of the strong temperature dependence of the conductivity measured at high temperatures. Unistep superexchange and multistep hopping [16, 17], carrier excitations across single-particle gaps [18], bandlike electronic transport [19], variable range hopping [20] and small polaron transport [21–26] are among the mechanisms suggested. It is plausible that when an electron or a hole is injected into a deformable macromolecule such as DNA, it will induce local distortions of the structure as the latter adjusts to the excess charge and lowers the system energy [27]; in other words a ‘polaronic’ distortion will be formed.

Tran *et al* [18] measured the conductivity and its temperature dependence along the lambda phage DNA (λ -DNA) double helix. Their low temperature data were attributed to ionic conduction due to the counterions. It was also reported that such ionic conduction cannot account for the strong temperature dependence and the large conductivity that they observed at high temperatures. This was attributed to carrier excitations across single-particle gaps or temperature driven hopping transport processes. Alternatively, they noticed that the observed behaviour would occur if phonon-assisted polaron hopping was the actual transport mechanism.

Yoo *et al* [28] reported measurements of electrical transport through poly(dA)–poly(dT) and poly(dG)–poly(dC) DNA molecules containing identical pairs. Their measured I – V characteristics at various temperatures were interpreted using a small polaron hopping model [29]. For the poly(dA)–poly(dT) regime they estimated the hopping distance to be about 16.8 Å (five base pairs), while for poly(dG)–poly(dC) they found it to be 25 Å (seven base pairs). Their effort to interpret the temperature dependence of the conductivity observed by Tran *et al* [18], using the same polaron hopping model over the whole temperature range, was not quite convincing and therefore they did not exclude other possible mechanisms.

Recently Kutnjak *et al* [30] reported measurements of temperature-dependent electrical conductivity obtained in native wet-spun calf thymus Li-DNA. They concluded that their measurements could be rather well described by the activated Arrhenius law but, on the basis of the quality of the fits, they did not exclude hopping.

Tran *et al* [18] emphasized that the models proposed for the interpretation of the electrical conductivity data had not taken into account the inherent disorder associated with the random base sequences, and also the random potentials along the DNA double helix arising from the randomly positioned positively charged counterions. The influence of counterion-induced disorder in DNA conduction has been recently investigated by Adessi and Anantram [31].

Yu and Song [20], to interpret the experimental data reported by Tran *et al* [18], proposed variable range hopping [32], considering DNA as a one-dimensional (1D) disordered system. The possible small polaron character of the carriers was not taken into account; nor was any multiphonon-assisted hopping mechanism which may be present at high temperatures. They concluded that on decreasing temperature, the conduction mechanism changes from nearest neighbour to variable range hopping [32].

Zhang *et al* [33] used a polaron model to study the effect of large molecular rotations on the usual hopping between neighbouring sites in systems with torsional degrees of freedom such as actual DNA molecules. This, along with the strong anharmonic interactions, may be of relevance for electronic transport in DNA, but these are beyond the scope of the present work.

Most recently Alexandre *et al* [34] reported *ab initio* calculations for poly(dC)–poly(dG) DNA, with up to four C–G pairs. They found a strong hole–lattice coupling and clear evidence for formation of small polarons and estimated the activation energy for polaron hopping and the polaron binding energies. They noticed that the estimate of Yoo *et al* [28] for the mean polaron hopping length, equal to 25 Å, seems surprisingly large for small polarons.

Summarizing the theoretical effort put in so far, it is of importance to underline the following points:

- (i) To our knowledge, none of the models proposed takes explicitly into account the effect of disorder on the small polaron binding energy. This affects drastically the intrinsic transition rate for a hop from a molecular site i to a neighbouring site j , and consequently the temperature dependence of the conductivity [35, 36].
- (ii) The multiphonon-assisted character of the hopping mechanism at high temperatures, which is also reflected in the expression of the intrinsic transition rate, has not been taken into account.
- (iii) There is a lack of a systematic derivation of an analytical expression for the electrical conductivity and the hopping distance as functions of the temperature, appropriate for multiphonon-assisted small polaron transport through a disordered one-dimensional medium, interpreting in a consistent way charge transport along the DNA ‘wire’.

The aim of the present work is to propose a transport model which incorporates all the above characteristics ((i)–(iii)) and interprets consistently the observed conductivity behaviour at high temperatures. In section 2 we give a description of the model used. On the basis of the ‘microscopic’ intrinsic transition rates, percolation arguments appropriate for the one-dimensional case under study lead to macroscopic analytical expressions for the temperature dependence of the low and the high temperature small polaron hopping conductivity. The maximum hopping distance and its temperature dependence are also obtained. Our theoretical results are applied to recent experimental data [18, 28] concerning the temperature dependence of the electrical conductivity of different DNA samples, in section 3.

2. Theory

2.1. The model

The above characteristics ((i)–(iii)) had been taken into account by Triberis and Friedman [35] and Triberis [36] introducing the generalized molecular crystal model (GMCM), as a generalization of Holstein’s MCM [37], for the appropriate study of small polaron hopping conductivity measured in bulk disordered systems.

For the case under study we consider a pair of neighbouring molecular lattice sites of the DNA ‘wire’, \mathbf{r}_i and \mathbf{r}_j , with energy disorder arising from the inherent disorder associated with the random base sequences, and the randomly positioned positively charged counterions [18, 31]. Let us denote by $\epsilon_i(0)$, and $\epsilon_j(0)$ the energies of the electrons on sites at vector positions \mathbf{r}_i , and \mathbf{r}_j , respectively, if the ‘molecular lattice’ sites are constrained not to be displaced in response to the presence of the electron. Due to the disorder, these local electronic energies, $\epsilon_i(0)$ and $\epsilon_j(0)$, are not equal. The energetic non-equivalence of the two sites will affect the small polaron’s binding energy, $E_b(i)$, in the sense that the lower the local electronic energy is, the more localized the electronic wavefunction will tend to be and consequently the larger its binding energy will be. Assuming that the stiffness of the ‘molecular lattice’ is unaltered, the difference in binding energy means a difference in the electron–lattice interaction parameters A_i and A_j , i.e. $E_i(x_i) = \epsilon_i(0) - A_i x_i$ and $E_j(x_j) = \epsilon_j(0) - A_j x_j$ with $A_i \neq A_j$. Here, $E_i(x_i)$ is the electronic energy of the system of the electron and the isolated molecule with configurational coordinate x_i , which represents the deviation of the atoms of the molecule at position \mathbf{r}_i from their equilibrium configuration, i.e. the local vibrational displacement coordinate.

The GMCM model [35] is based on a generalized ‘hopping model’ Hamiltonian of the form

$$\langle m|H|n\rangle = \langle m|H_0 + V|n\rangle = E_{i,[n_k]}\delta_{ij}\delta_{[n_k],[n_k']} + \langle m|V|n\rangle. \quad (1)$$

The $\langle m|V|n\rangle$ term [35] is the overlap part of the Hamiltonian, $|n\rangle = |i, [n_{\mathbf{k}}]\rangle$ are the eigenstates of H , H_0 is the zeroth-order (i.e. for the electronic overlap integral of the tight binding theory $J = 0$) Hamiltonian and $[n_{\mathbf{k}}]$ represents the totality of the vibrational quantum numbers $(\dots, n_{\mathbf{k}}, \dots)$ for the occupation of the site with position vector \mathbf{r}_i . The eigenvalues of H_0 are

$$E_{i,[n_{\mathbf{k}}]} = \epsilon_i(0) - E_b(i) + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}}(n_{\mathbf{k}} + \frac{1}{2}). \quad (2)$$

Here, $\omega_{\mathbf{k}}$ is the normal-mode frequency and

$$E_b(i) = \frac{1}{N} \sum_{\mathbf{k}} (A_i^2/2M\omega_{\mathbf{k}}^2), \quad (3)$$

is the small polaron binding energy. N is the number of ‘molecular lattice’ sites and M is the appropriate reduced atomic mass. The relation between $\omega_{\mathbf{k}}$ and its associated wavevector \mathbf{k} , i.e. the dispersion relation, is given by

$$\omega_{\mathbf{k}}^2 = \omega_0^2 + \omega_1^2 \sum_{\mathbf{k}} \cos(\mathbf{k} \cdot \mathbf{h}'), \quad (4)$$

where $k = 2\pi p/N$, the integer p lying in the range $-(N-1)/2 \leq p \leq (N-1)/2$, and \mathbf{h}' indexes the nearest neighbours $(\mathbf{r}_i + \mathbf{h}')$ of an arbitrary site \mathbf{r}_i . ω_0 is the harmonic oscillator frequency associated with the configurational coordinate of the isolated molecule. The relation $\omega_1 \ll \omega_0$ determines the weak dispersion limit. The equations (2) and (3) show the essential features of the GMCM which are:

- (1) Site-dependent local electronic energy $\epsilon_i(0)$.
- (2) Site-dependent electron–lattice interaction parameter, A_i , and concomitant binding energy, $E_b(i)$.

The knowledge of $\langle m|V|n\rangle$ permits the evaluation of the ‘microscopic’ velocity operator [38, 39],

$$u_{ij} = \langle m|u|n\rangle = \left(\frac{i}{\hbar}\right) \langle m|V|n\rangle(\mathbf{r}_j - \mathbf{r}_i), \quad (5)$$

the charge current density operator,

$$j_{ij} = n_c q u_{ij}, \quad (6)$$

where n_c is the charge carrier concentration and q is the carrier’s charge, the conductivity [40],

$$\sigma_{ij} = \int_0^\infty dt \int_0^\beta d\rho \langle j(-i\hbar\rho)j(t)\rangle, \quad (7)$$

where $\beta = 1/k_B T$ and k_B is Boltzmann’s constant. The mobility, μ_{ij} , and consequently the diffusion constant are given by $D_{ij} = \mu_{ij}/e\beta$. Thus, the ‘microscopic’ jump rate reads

$$L_{ij} = \frac{D_{ij}}{|\mathbf{r}_i - \mathbf{r}_j|^2}. \quad (8)$$

2.2. Percolation treatment

Assuming that the dependence on the spatial separation R_{ij} of the two sites is $\exp(-2\alpha R_{ij})$ [41], α^{-1} being the spatial extent of the electronic wavefunction localized at a single site, the ‘microscopic’ intrinsic transition rate γ_{ij} , for a small polaron hopping from a site i to an empty site j is given by

$$\gamma_{ij} = \exp(-2\alpha R_{ij})L_{ij}. \quad (9)$$

Knowing the intrinsic transition rate, we obtain the average equilibrium transition probability [35], W_{ij}^0 :

$$W_{ij}^0 = (W_{ij}^0 W_{ji}^0)^{1/2} = [n_i^0(1 - n_i^0)]^{1/2} [n_j^0(1 - n_j^0)]^{1/2} (\gamma_{ij} \gamma_{ji})^{1/2}, \quad (10)$$

where n_i^0 is the equilibrium occupation probability of the i th site.

The analytical expressions for W_{ij}^0 at high (h) [35] and low (l) [36] temperatures are different due to the fact that the former is a multiphonon-assisted while the latter is a few-phonon-assisted process. This is reflected in W_{ij}^0 .

At high temperatures W_{ij}^{0h} is given by [35]

$$W_{ij}^{0h} = \gamma_0^h \exp(-2\alpha R_{ij}) \exp[-(|E_i| + |E_j| + 2\epsilon_2)/2k_B T]. \quad (11)$$

Here $\epsilon_2 = [E_b(i) + E_b(j)]/4$ and γ_0^h is given by [35]

$$\gamma_0^h = (J^2/\hbar)(\pi/4\epsilon_2 k_B T)^{1/2}. \quad (12)$$

$E_i = \epsilon_i(0) - E_b(i)$ is the electronic energy when the electron resides on site i . W_{ij}^{0h} depends on both site energies E_i and E_j .

At low temperatures, W_{ij}^{0l} depends solely on E_i or E_j [36]:

$$W_{ij}^{0l} = \gamma_0^l \exp(-2\alpha R_{ij}) \exp[-(|E_i| + |E_j| + |E_i - E_j|)/2k_B T], \quad (13)$$

with γ_0^l given by

$$\gamma_0^l = (\omega_0/\pi) [\pi J \exp(-2\epsilon_2/\hbar\omega_0)/\hbar\omega_0]^2 [(4\epsilon_2/\hbar\omega_0)^{\Delta_{ij}/\hbar\omega_0} / (\Delta_{ij}/\hbar\omega_0)!]. \quad (14)$$

This results in different percolation conditions and consequently in different behaviours of the conductivity for the two regimes, taking no account of correlations between bonds due to the energy of the common site [42].

The transport problem is transformed to an equivalent network of impedances Z_{ij} [41], given by

$$Z_{ij} = [(q^2/k_B T)(W_{ij}^0)]^{-1}. \quad (15)$$

At high temperatures, the impedances Z_{ij}^h are given by

$$Z_{ij}^h = Z_0^h \exp(\xi_{ij}^h), \quad (16)$$

where

$$Z_0^h = k_B T/q^2 \gamma_0^h. \quad (17)$$

$$\xi_{ij}^h = 2\alpha R_{ij} + E_{ij}/k_B T, \quad (18)$$

and

$$E_{ij} = [|E_i| + |E_j| + 2\epsilon_2]/2. \quad (19)$$

Assuming, as shown in [35], that the electronic energy, is mainly polaronic, i.e. $|E_i| \simeq E_b(i)$, $|E_j| \simeq E_b(j)$, hereafter we shall refer to the binding energy as the site energy, and we shall use the symbol E_i instead of $E_b(i)$ for convenience.

Percolation takes place when

$$Z_{ij}^h \leq Z_c^h. \quad (20)$$

The inverse quantity Z_c^{h-1} characterizes the macroscopically observed conductivity of the material.

Due to equation (16) the condition for percolation, given by equation (20), reads

$$\xi_{ij}^h \leq \xi_c^h \quad (21)$$

or equivalently

$$\frac{R_{ij}}{r_m^h} + \frac{E_i}{E_m^h} + \frac{E_j}{E_m^h} \leq 1, \quad (22)$$

where $r_m^h = \xi_c^h/2\alpha$ is the maximum hopping distance and $E_m^h = k_B' T \xi_c^h$ is the maximum site energy. $k_B' = \frac{4}{3}k_B$.

The conductivity of the specimen is expressed as

$$\sigma^h = \sigma_0^h \exp(-\xi_c^h), \quad (23)$$

where $\sigma_0^h = (Z_0^h)^{-1}$.

We apply the arguments given in [31] to our one-dimensional case. We define $P(Z_c)$ as the average number of impedances of magnitude Z_c or less connected to a given site, where Z_c is the impedance of the specimen.

According to Pollak [43], the condition for percolation reads

$$P(Z_c) = \theta. \quad (24)$$

At high temperatures the function $P(Z_c^h)$ can be evaluated from the distribution of the random variables E_i , E_j and R_{ij} , which enter into the expression for Z :

$$\theta = \int_{z=\text{const}} p(E_i, E_j, R_{ij}) dR_{ij} dE_i dE_j. \quad (25)$$

We assume a lack of correlation between successive impedances in a chain, and between the distributions of R_{ij} and site energies. The integration extends over surfaces defined by

$$\frac{R_{ij}}{r_m^h} + \frac{2E_j - \Delta_{ij}}{E_m^h} = 1. \quad (26)$$

We obtain

$$P(Z_c^h) = \theta = \int_0^{E_m^h} \left[\int_{\Delta}^{\Delta'} \frac{1}{N_s} N(E - \Delta) N(E) \left(\int_0^{R'} dR \right) dE \right] d\Delta, \quad (27)$$

where N_s is the concentration of sites, $N(E)$ is the dos, $\Delta' \equiv (E_m^h + \Delta)/2$ and $R' = r_m^h [1 - (2E - \Delta)/E_m^h]$. The indices of E_i , Δ_{ij} and R_{ij} have been dropped. For the one-dimensional case, for θ we use the value 2 [44]. We have chosen a constant density of states N_0 [35, 45–47], over an energy range E_m^h . Performing the integrations in equation (27), we obtain

$$\xi_c^h = (T_0^h/T)^{2/3}, \quad (28)$$

where $T_0^h = 27^{1/2} N_s^{1/2} \alpha^{1/2} / N_0 k_B$.

Thus, the conductivity of the high temperature multiphonon-assisted small polaron hopping one-dimensional regime is given by

$$\sigma^h = \sigma_0^h \exp[-(T_0^h)^{2/3} T^{-2/3}]. \quad (29)$$

For a given temperature, equation (29) is expressed as

$$\sigma^h = \sigma_0^h \exp(-2r_m^h/\alpha^{-1}). \quad (30)$$

Here,

$$r_m^h = \alpha^{-1} (T_0^h)^{2/3} T^{-2/3} / 2, \quad (31)$$

i.e. the maximum hopping distance follows a $T^{-2/3}$ law.

Equation (29) allows the evaluation of T_0^h from the slope of the fits of the experimental data reported for DNA samples, plotted as $\ln \sigma^h$ versus $T^{-2/3}$. Using equation (31) we evaluate the maximum small polaron hopping distance at the temperature of interest.

At low temperatures, where only few phonons assist the small polaron hopping, the low temperature analogue of equation (22) reads

$$\frac{R_{ij}}{r_m^l} + \frac{E}{E_m^l} \leq 1, \quad (32)$$

where E equals E_j if $E_j > E_i$ or E_i if $E_i > E_j$, $E_m^l = k_B T \xi_c^l$ and $r_m^l = \xi_c^l / 2\alpha$. Integrating over surfaces of constant Z defined by

$$\frac{R_{ij}}{r_m^l} + \frac{E_i(\text{or } E_j)}{E_m^l} = 1 \quad (33)$$

we obtain

$$\theta = \int_{z=\text{const}} p(E, R_{ij}) dR_{ij} dE \quad (34)$$

or

$$P(Z_c^l) = \theta = \int_0^{E_m^l} N(E) \left(\int_0^{R_m^l} dR \right) dE. \quad (35)$$

Here $R_m^l = (r_m^l / E_m^l)(E_m^l - E)$.

Consequently, at low temperatures, $\xi_c^l = (T_0^l / T)^{1/2}$, with $T_0^l = (4\alpha / N_0 k_B)$, and the conductivity is given by

$$\sigma^l = \sigma_0^l \exp[-(T_0^l)^{1/2} T^{-1/2}], \quad (36)$$

where $\sigma_0^l = (Z_0^l)^{-1}$. $Z_0^l = k_B T / q^2 \gamma_0^l$.

The maximum hopping distance at low temperatures is given by

$$r_m^l = \alpha^{-1} (T_0^l)^{1/2} T^{-1/2} / 2. \quad (37)$$

Triberis and Friedman [35] reported that the $T^{-2/5}$ law they derived for small polaron transport in bulk disordered semiconductors, at high temperatures, follows a general law which is based on the GMCM and percolation arguments, namely a $T^{-\epsilon/(\epsilon+r)}$ law. Here, ϵ is the energy's dimensions (number) involved in the percolation condition and r the spatial dimensions involved. This law has been successfully applied to interpret the temperature dependence of the conductivity of a variety of amorphous materials. A brief review is presented in [48]. For the low temperature (bulk) case ($\epsilon = 1$ and $r = 3$) the $T^{-1/4}$ law (Mott's law) was obtained, which is also a widely accepted behaviour. For the case of longitudinal conduction at low temperatures in thin films ($\epsilon = 1$ and $r = 2$) a $T^{-1/3}$ law was obtained [43]. For our 1D case at high temperatures ($\epsilon = 2$ and $r = 1$) a $T^{-2/3}$ law was expected, in accordance with our result, while at low temperatures ($\epsilon = 1$ and $r = 1$) we obtain a $T^{-1/2}$ law, which is in accordance with the variable range hopping results for localized states [43, 49, 50].

3. Results and discussion

We apply our theoretical results to recently reported experimental data. The Tran *et al* [18] data at 12 GHz show pronounced strong temperature-dependent conductivity from 227 to 342 K, i.e. in a 115 K wide temperature region (I). The corresponding values of the conductivity vary from 0.28 up to 4.29 $\Omega^{-1} \text{ cm}^{-1}$, respectively. At 100 GHz strong temperature dependence is observed in a 90 K wide temperature region. Yoo *et al* [28], for poly(dA)–poly(dT) DNA molecules, observed a similar behaviour from 178 to 306 K, i.e. in a 128 K wide temperature region (II). The corresponding values of the conductance (G) vary from $3.31 \times 10^{-11} \Omega^{-1}$ up to $1.5 \times 10^{-8} \Omega^{-1}$, respectively. Their measurements on poly(dG)–poly(dC) samples relate to a much narrower temperature region about 89 K wide. The Kutnjak *et al* [30] data relate to a

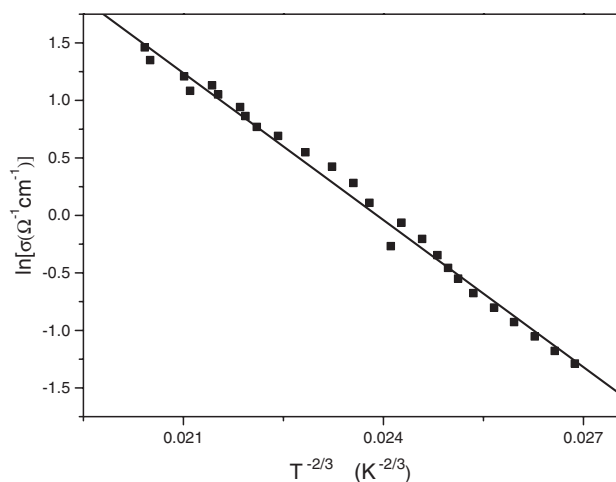


Figure 1. $\ln \sigma$ versus $T^{-2/3}$ plot of the Tran *et al* [18] (12 GHz) (λ -DNA) data for the high temperature (227–342 K) region.

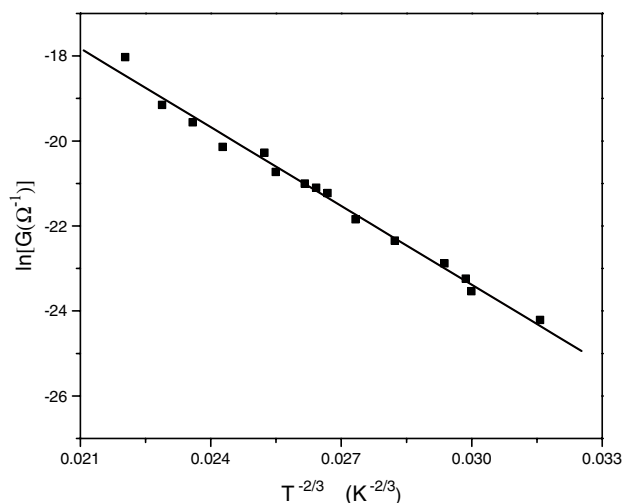


Figure 2. $\ln G$ versus $T^{-2/3}$ plot of the Yoo *et al* [24] poly(dA)–poly(dT) DNA data for the high temperature (178–306 K) region.

narrow temperature region of about 40 K up to 45 K. The experimental values of the measured conductivity vary by orders of magnitude within different temperature ranges, more or less wide. We have applied our theoretical approach to the data which relate to the wider ‘high temperature’ regions (I and II) to maximize the credibility of the fits. As ‘high temperature’ regions we characterize those in which the measured conductivity shows a pronounced strong dependence, although this is strictly determined by the condition $\hbar\omega_0 \leq k_B T$ [39]. The low temperature region is characterized by a very weak temperature dependence of the conductivity.

Figures 1 and 2 show $\ln \sigma$ and $\ln G$ as a function of $T^{-2/3}$ for the Tran *et al* (12 GHz) (λ -DNA) [18] (I) and Yoo *et al* (poly(dA)–poly(dT)) [28] (II) high temperature data. For α^{-1} we use the value 2 Å which is within the range of values usually used for DNA and similar structures [20, 51]. The experimental data follow nicely the $T^{-2/3}$ law.

Given that the strong temperature dependence of the measured conductivity can be equally nicely fitted using different exponential laws, as has been attempted in the past [18, 20, 28, 30], the quality of the fits does not necessarily guarantee the validity of our theoretical approach. However, our theoretical analysis allows the evaluation of the maximum hopping distance. This, along with the inclusion of the appropriate characteristics ((i)–(iii)) of the system under study, is important for testing the reliability of our theory.

The value of T_0^h is determined from the slopes (equation (29)) of the curves given in figures 1 and 2. Using equation (31) we evaluate the maximum hopping distance $r_m^h(T)$ at the lower and higher temperatures of the ‘high temperature’ region. For the Tran *et al* [18] data(I), we obtain $T_0^h(\text{I}) = 8.79 \times 10^3$ K, $r_m^h(\text{I})(227 \text{ K}) = 11.4 \text{ \AA}$ (three base pairs) and $r_m^h(\text{I})(342 \text{ K}) = 8.7 \text{ \AA}$ (two base pairs). Correspondingly, for the Yoo *et al* [28] data(II), we obtain $T_0^h(\text{II}) = 1.45 \times 10^4$ K, $r_m^h(\text{II})(178 \text{ K}) = 18.8 \text{ \AA}$ (five base pairs) and $r_m^h(\text{II})(306 \text{ K}) = 13 \text{ \AA}$ (four base pairs).

The value of $r_m^h(\text{II})$ obtained at 306 K (13 Å) for the poly(dA)–poly(dT) sample is comparable with the value 16.8 Å reported by Yoo *et al* [28]. Here we must note that the lack of an analytical expression based on a transport mechanism, appropriate for the specific system, forced Yoo *et al* [28] to use the (fitting) parameter b to fit their experimental data as $I \sim bV$. According to the authors, the parameter b has a temperature dependence whose physical origin is not clear. The above probably resulted from an unconvincing attempt, as the authors [28] noticed, at the interpretation of Tran *et al* data. The approach presented in the present work does not need any fitting parameter because the transport mechanism responsible is described in detail; it is based on first principles, leading to analytical expressions for the conductivity as a function of the temperature, and the maximum hopping distance as well.

Our results support small polaron hopping to next neighbour sites, at high temperatures, as responsible for the strong temperature dependence of the conductivity measured. They imply that as the temperature increases, in the high temperature regime, shorter hops (or equivalently smaller impedances; cf equations (29) and (30)) contribute to the transport process, resulting in an increase of the conductivity, in accordance with experiment.

We also investigated the possibility of an interpretation of the weak temperature dependence of the conductivity at low temperatures, using the low temperature $T^{-1/2}$ results, given by equations (36) and (37). We obtained unacceptably low values for the maximum hopping distances, much lower than those obtained for the high temperature regime, in contrast with the much lower conductivity measured. Thus, small polaron hopping does not seem to be responsible for the charge transport, at low temperatures, in DNA, in accordance with other reports [18, 28].

4. Conclusion

In conclusion, on the basis of a multiphonon-assisted small polaron hopping model we have obtained analytical expressions for the high temperature hopping conductivity in the presence of disorder in one dimension. Comparison of the theoretical results with recent experimental data for charge transport along the DNA double helix leads to realistic maximum hopping distances and consistent interpretation of their temperature dependence. The above results support the idea of multiphonon-assisted hopping of small polarons between next nearest neighbours as the transport mechanism responsible for the strong high temperature dependence of the electrical conductivity.

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