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Field and temperature dependence of the small polaron hopping electrical conductivity in 1D disordered systems

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Abstract

We investigate the effect of the electric field and the temperature on the electrical conductivity of one-dimensional disordered systems due to phonon assisted hopping of small polarons. The microscopic transport mechanism is treated within the framework of the generalized molecular crystal model and the Kubo formula, while percolation theoretical arguments lead to analytical expressions for the macroscopic behavior of the electrical conductivity at high (multi-phonon assisted hopping) and low (few-phonon assisted hopping) temperatures under the influence of moderate electric fields. The theoretical results are successfully applied to recent experimental findings for a wide temperature range and from low up to moderate electric fields. Comparison is made with other theories.

1. Introduction

Observations during the past three decades have shown strong nonlinearities on the conductivity of one-dimensional (1D) disordered systems, such as amorphous semiconductors [1, 2], amorphous carbon [3], doped polymers [4–6], conjugated polymers [7, 8]. The discovery of the polymeric field effect transistors and light emitting diodes [9, 10] caused an intense research activity devoted to the field dependence of the conductivity in disordered organic materials and conjugated polymers. Carbon nanotubes, nanowires and conductive molecules [11–16], DNA being one of the most representative specimens, have been placed among the most promising materials for nanotechnology [17–19, 11, 20, 21]. Understanding of the carrier transport properties in these materials is important for innovating applications and the refinement of many others.

The absence of long range order in the presence of an electron or a hole in a deformable disordered environment leads to the localization of the carrier and results in the transport of the charge carriers via hopping mechanism. If the interaction energy between the carrier and the phonons is strong compared with the bandwidth, a 'small polaron' may form. Interaction with phonons will normally determine the hopping rate between one localized state to another. The polaronic character of the carriers, the presence of disorder and the effect of the application of a finite electric field along with temperature variations have to be taken into account in a systematic theoretical investigation of the electrical behavior of these systems. The interplay of 'external stimuli' such as the electric field and the temperature is still an open question.

For 3D systems the effect of the electric field on the variable range hopping DC conductivity has been studied in the past by various workers. In the early seventies Apsley and Hughes [22, 23] introduced a method to evaluate the field dependence of the DC conductivity for the case of single-phonon assisted hopping motion in a disordered system. In the late eighties Triberis [24–26], based on the Apsley and Hughes approach, examined the effect of the electric field on the conductivity of the high (and low) temperature multi-phonon (few-phonon) assisted small polaron hopping regime, considering the deformation of the surroundings, induced by the carriers i.e. small polaron formation, as a basic ingredient of the theory.

For 1D disordered systems, in the variable range hopping regime at low temperatures, Fogler and Kelly [27] investigated theoretically the effect of a finite electric field on the resistivity. As they pointed out '... remarkably, the ultimate

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low-T limit where variable range hopping dominates, has long remained untouched by theorists except for one early numerical study [28]. In their analysis [27], they took into account the existence of highly resistive segments ('breaks') on the conducting path of the carriers in 1D systems and found that the role of the breaks diminishes and eventually becomes insignificant as the electric field increases.

More recently, Ma *et al* [29] presented a model to describe hopping transport and the DC conductivity of 1D systems with off-diagonal disorder. Considering the temperature dependence of the DC hopping conductivity, they showed that it increases with the increase of temperature taking much larger values than in the case of the Anderson model with pure diagonal disorder. They also studied the field dependence of the conductivity to find that at low electric fields the hopping conductivity conforms with the ohmic law, but at strong fields it presents non-ohmic characteristics.

A number of experimental reports concern electrical transport properties of 1D disordered systems [30–35]. In these studies several mechanisms have been proposed for the interpretation of the experimental measurements. Multistep hopping [36, 37], carrier excitations across single particle gaps [33], variable range hopping [38] or small polaron transport [13, 14, 39, 40] are among the mechanisms suggested. In some cases phenomenological expressions for the temperature and the field dependence of the electrical conductivity have been employed, such as the activated Arrhenius law or expressions coming from the study of 3D [41] or mesoscopic systems [42], and fitting parameters of obscure physical origin are involved. This is due to the lack of a systematic study of the transport mechanism leading to analytical expressions for these dependencies.

Cumings *et al* [30] measured the electrical resistance R between the ends of a multiwall carbon nanotube during telescopic extension of the nanotube. In order to provide additional support for the carriers' localization in the 1D system they measured the electric field dependence of the system's resistance. They found that at low applied voltages the resistance is approximately constant with some fluctuations due to experimental noise, while at high applied voltages it decreases precipitously with increasing voltage, leading to a nonlinear conductance.

Aleshin *et al* [31] studied the charge transport in polydiacetylene quasi-1D single crystals (PDA-PTS) as a function of the temperature, for a wide temperature range, 25 up to 300 K, and moderate electric fields up to 10^5 Vm^{-1}. Their measurements showed that as the electric field increases, the deviations from the ohmic behavior become apparent and the temperature dependence of the current becomes weaker. Different transport mechanisms and corresponding expressions for the electrical conductivity, based on rather different microscopical approaches were used for the interpretation of the experimental findings, such as strongly thermally activated hopping or variable range hopping conduction. At $T > 50$ K, they reported thermally activated nearest-neighbor hopping by small polarons as a possible transport mechanism.

Tang *et al* [32] reported the electric transport properties of mono-sized carbon nanotubes fabricated in the channels of $AlPO_4$-5 zeolite crystals. Polarized Raman spectra measurement showed the 1D behavior of the carbon nanotubes. Varying temperature within the range of 25 up to 160 K they observed a conductance monotonically decreasing with decreasing temperature. The hopping mechanism was phenomenologically described as a combination of thermal activation and tunneling. Different fits of the temperature dependence of the conductance measured at zero bias voltage and at low temperatures, of the form $\ln\sigma_{1D} \sim T^{-1/(d+1)}$ with $d = 1, 2$ and 3, showed that their measured results were in good-agreement with the $\ln\sigma_{1D} \sim T^{-1/2}$ behavior, implying 1D hopping of electrons in their carbon nanotubes.

Recently, Triberis *et al* [43, 44], based on the generalized molecular crystal model (GMCM), introduced by Triberis and Friedman [45], and theoretical percolation arguments, investigated small polaron hopping transport in 1D disordered systems at high temperatures (h), ignoring the effect of correlations. An analytical expression for the temperature dependence of the electrical conductivity, $\ln\sigma^{h} \sim T^{-2/3}$, was obtained. This result reproduced satisfactorily the experimental data reported for λ-DNA and for poly(dA)-poly(dT) DNA [33, 34], considering DNA as a one-dimensional disordered molecular wire in which small polarons are the charge carriers. $\ln\sigma^{h}$-versus-$T^{-2/3}$ plots permitted the evaluation of the maximum hopping distance.

Most recently Triberis and Dimakogianni [46] showed that the inclusion of correlations (cr) leads to a $\ln\sigma^{h,cr} \sim T^{-1/2}$ law, a result which is consistent with the corresponding 3D case.

An analytical expression predicting the electric field and temperature dependence of the DC conductivity taking into account the 1D character of the system, the presence of disorder and the polaronic character of the carriers remains to be theoretically established. It is the purpose of the present work to contribute in this direction for the high (multi-phonon assisted) and the low (few-phonon assisted) temperature small polaron hopping regime when low and moderate electric fields are applied and correlations are ignored.

The present paper attempts to extend the percolation theory of hopping conduction of small polarons beyond the limit of linear response (i.e. the 'ohmic regime') which manifests itself applying low electric fields. The main attention is paid to the regime of moderate fields. We use this term to describe the region of transport where the logarithm of the conductivity can be expanded in powers of the field F, and well approximated by the lowest non-zero power. It will turn out that the natural expansion parameter is $e\alpha^{-1}F/2k_BT$, hence in moderate fields one requires $e\alpha^{-1}F/2k_BT < 1$, where α^{-1} is the spatial extend of the localized small polaron wavefunction and e the electronic charge.

The model used is based on the picture of dissipative motion of charged classical particles in a one-dimensional lattice potential comprised of 'molecular lattice sites' along which small polarons are transported, in the presence of disorder, under the influence of an electric field. In a low or moderate electric field, the electron–lattice site interaction leads to the localization of the particles on the lattice sites. Their transport mechanism has hopping-like character assisted

by the phonons and the electric field. Applying large electric fields the motion of the particle becomes unbounded [47] i.e. the application of large electric fields could result in the dissociation of small polarons.

Strong electric fields could also favor hops mainly oriented in the direction opposite to the electric field, making vanishingly small the probability of hops in the direction of the field. This imposes the use of the concept of the directed percolation [48, 49].

Implications of this sort, any effects of the electric field on the electronic wavefunctions or any processes such as Poole–Frenkel, are not considered in the present work.

The theoretical results are applied to the full set of experimental data of Aleshin *et al* [31] and comparison is made with other theories.

2. The model

The model we use for the study of the small polaron hopping motion in a disordered system is the generalized molecular crystal model (GMCM), introduced by Triberis and Friedman [45]. It is based on a generalized 'hopping model' Hamiltonian of the form

$$\langle m|H|n\rangle = \langle m|H_0 + V|n\rangle = E_{i,\{n_{\mathbf{k}}\}}\delta_{ij}\delta_{\{n_{\mathbf{k}}\},\{n_{\mathbf{k}'}\}} + \langle m|V|n\rangle, \tag{1}$$

where $|n\rangle = |i, \{n_{\mathbf{k}}\}\rangle$, are the eigenstates of H, and H_0 is the zeroth-order (i.e. the electronic transfer integral $J = 0$) Hamiltonian used by Holstein [50] with corresponding eigenvalues,

$$E_{i,\{n_{\mathbf{k}}\}} = \epsilon_i(0) - E_{\mathrm{b}}(i) + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}}(n_{\mathbf{k}} + 1/2). \tag{2}$$

Here, $\{n_{\mathbf{k}}\}$ represents the totality of vibrational quantum numbers $(\ldots, n_{\mathbf{k}}, \ldots)$ for the occupation of the site with position vector $\mathbf{r}_i$,

$$E_{\mathrm{b}}(i) = \left(\frac{1}{N}\right)\sum_{\mathbf{k}} \frac{A_i^2}{2M\omega_{\mathbf{k}}^2}, \tag{3}$$

is the small polaron binding energy, and $\epsilon_i(0)$ is the local electronic energy. N is the number of lattice sites and A_i is the electron–lattice interaction parameter.

Equations (2) and (3) show the essential extends of the MCM which are:

(1) a site-dependent local electronic energy, $\epsilon_i(0)$,
(2) a site-dependent interaction parameter, A_i, and concomitant binding energy, $E_{\mathrm{b}}(i) = N^{-1}\sum_{\mathbf{k}}(A_i^2/2M\omega_{\mathbf{k}}^2)$.

The treatment refers to the non-adiabatic limit, i.e. in the physical situation where the electron is no longer able to follow rapid fluctuations of the lattice and, hence, it does not respond quickly enough to the occurrence of a coincident event in order to overcome the energy barrier. In this case, J can be treated as a small perturbation in the lowest order [51, 52].

3. Field dependence of the conductivity for the high temperature small polaron hopping regime

When a carrier hops from a site i of energy E_i to a site j of energy E_j, at distance R_{ij}, the average equilibrium transition probability at *high* (h) *temperatures* ($\hbar\omega_0 \ll k_{\mathrm{B}}T$ [45, 53]), $W_{ij}^{0\mathrm{h}}$, is given by [45]

$$W_{ij}^{0\mathrm{h}} = \gamma_0^{\mathrm{h}} \exp(-2\alpha R_{ij}) \exp[-(|E_i| + |E_j| + 2\epsilon_2)/2k_{\mathrm{B}}T]. \tag{4}$$

Here, $\epsilon_2 = (E_i + E_j)/4$ and $\gamma_0^{\mathrm{h}} = (J^2/\hbar)(\pi/4\epsilon_2 k_{\mathrm{B}}T)^{1/2}$. The energy of the carrier is taken mainly polaronic [45].

Using equation (4) and defining for convenience the reduced variables $R'_{ij} = 2\alpha R_{ij}$, $E'_i = E_i/k'_{\mathrm{B}}T$, $E'_j = E_j/k'_{\mathrm{B}}T$, where $k'_{\mathrm{B}} = 4k_{\mathrm{B}}/3$, the average equilibrium transition probability can be written as

$$W_{ij}^{0\mathrm{h}} = \gamma_0^{\mathrm{h}} \exp[-(R'_{ij} + E'_i + E'_j)]. \tag{5}$$

Following [22, 24], as the hopping probability, $W_{ij}^{0\mathrm{h}}$, depends on R'_{ij}, E'_i and E'_j it is logical to combine R'_{ij} with E'_i and E'_j into a single parameter. It is this single parameter, the 'range' $\Re^{0\mathrm{h}}$ between two sites which determines the probability of hopping between them. The states between which the small polaron hops may be regarded as points in a three-dimensional random array (one spatial coordinate and two energy coordinates, cf equation (5)), where the 'distance' between two states is given by the range $\Re^{0\mathrm{h}}$. The shorter this 'distance' is the greater the hopping probability. Conduction is the result of many series of hops through this three-dimensional array, and as short range hops are favored, it is the average nearest-neighbor 'distance' between states in this three-dimensional space which determines the overall conductivity.

Thus,

$$W_{ij}^{0\mathrm{h}} = \gamma_0^{\mathrm{h}} \exp(-\Re^{0\mathrm{h}}), \tag{6}$$

where

$$\Re^{0\mathrm{h}} = R'_{ij} + E'_i + E'_j. \tag{7}$$

In the presence of an electric field, F, the average transition probability, $W_{ij}^{\mathrm{h}}(F)$, assuming that the occupation probabilities do not change in the presence of the electric field, will be given by

$$W_{ij}^{\mathrm{h}}(F) = W_{ij}^{0\mathrm{h}} \exp(-eFR_{ij}\cos\theta/k_{\mathrm{B}}T), \tag{8}$$

or

$$W_{ij}^{\mathrm{h}}(F) = \gamma_0^{\mathrm{h}} \exp(-[R'_{ij}(1 + \lambda\cos\theta) + E'_i + E'_j]), \tag{9}$$

where

$$\lambda = eF/2\alpha k_{\mathrm{B}}T. \tag{10}$$

For the 1D case θ takes the values 0 or π.

Thus, we may write

$$W_{ij}^{\mathrm{h}}(F) = \gamma_0^{\mathrm{h}} \exp(-\Re_{ij}^{\mathrm{h}}), \tag{11}$$

where

$$\Re_{ij}^{\mathrm{h}} = R'_{ij}(1 + \lambda\cos\theta) + E'_i + E'_j. \tag{12}$$

Then, the conductivity $\sigma^{\rm h}(F)$ varies as $\exp(-\bar{\Re}^{\rm h}_{\rm nn})$ or

$$\ln\sigma^{\rm h}(F) = \text{const} - \bar{\Re}^{\rm h}_{\rm nn}, \tag{13}$$

where, $\bar{\Re}^{\rm h}_{\rm nn}$ is the average nearest-neighbor range. The problem is therefore to calculate this quantity.

Firstly, we have to obtain $N^{\rm h}(\Re^{\rm h})$, the total number of states within a range $\Re^{\rm h}_{ij}$. For our case we can write the range $\Re^{\rm h}_{ij}$ as

$$\Re^{\rm h} = R'(1+\lambda\cos\theta) + 2E' - \Delta', \tag{14}$$

where $E'_i = E'_j - \Delta'_{ij}$, Δ'_{ij} is the disorder energy between the two sites i and j, and the indices from E'_j, Δ'_{ij} and R'_{ij} have been dropped.

In reduced coordinates

$$N^{\rm h}(\Re^{\rm h}) = \sum_{\theta=0,\pi}\left\{C^{\rm h}_1\int_0^{\Re^{\rm h}}\left[\int_{\Delta'}^{\Delta^*}\left(\int_0^{R^{\rm h*}} {\rm d}R^{\rm h'}\right){\rm d}E'\right]{\rm d}\Delta'\right\}, \tag{15}$$

where $C^{\rm h}_1 = (k'_{\rm B}T)^2N_0^2/2\alpha N_{\rm S}$, $\Delta^{\rm h*} = (\Re^{\rm h}+\Delta')/2$ and $R^{\rm h*} = [\Re^{\rm h} - (2E'-\Delta')]/(1+\lambda\cos\vartheta)$.

Here, $N_{\rm S}$ is the concentration of sites, and N_0 is the density of states (D.O.S) assumed to be constant, i.e. $N(E_j) = N(E_i) = N_0$. We have also taken $E_{\rm F} = 0$.

Integrating we obtain

$$N^{\rm h}(\Re^{\rm h}) = C^{\rm h}_2(\Re^{\rm h})^3, \tag{16}$$

where $C^{\rm h}_2 = C^{\rm h}_1/6(1-\lambda^2)$.

Defining

$$\Delta N(\Re) = \frac{\partial N(\Re)}{\partial\Re}, \tag{17}$$

the number of states with ranges between $\Re$ and $\Re + {\rm d}\Re$ is $\Delta N(\Re)\,{\rm d}\Re$. Then the probability that a state with range $\Re$ is the nearest neighbor in the three-dimensional space is given by

$$P_{\rm nn}(\Re) = S(\Delta N(\Re)/S)[1 - N(\Re)/S]^{S-1}. \tag{18}$$

Here, S is the total number of states in the system which is clearly very large. Thus, we may write

$$P_{\rm nn}(\Re) = \Delta N(\Re)\exp(-N(\Re)), \tag{19}$$

as the nearest-neighbor distribution.

Having evaluated $N^{\rm h}(\Re^{\rm h})$ we obtain

$$\Delta N^{\rm h}(\Re^{\rm h}) = C^{\rm h}_3(\Re^{\rm h})^2, \tag{20}$$

where $C^{\rm h}_3 = 3C^{\rm h}_2$. Then the nearest-neighbor distribution becomes

$$P_{\rm nn}(\Re^{\rm h}) = 3C^{\rm h}_2(\Re^{\rm h})^2\exp[-C^{\rm h}_2(\Re^{\rm h})^3]. \tag{21}$$

The average nearest-neighbor range $\bar{\Re}_{\rm nn}$ is given by

$$\bar{\Re}_{\rm nn} = \frac{\int_0^\infty \Re P_{\rm nn}(\Re)\,{\rm d}\Re}{\int_0^\infty P_{\rm nn}(\Re)\,{\rm d}\Re}. \tag{22}$$

Substituting $P^{\rm h}_{\rm nn}(\Re^{\rm h})$ from (21) into (22) and integrating, we obtain

$$\bar{\Re}^{\rm h}_{\rm nn} = \frac{\Gamma(\frac{4}{3})}{(C^{\rm h}_2)^{1/3}}, \tag{23}$$

where $\Gamma(n)$ is the gamma function. Thus,

$$\bar{\Re}^{\rm h}_{\rm nn} = \left(\frac{T^{\rm h}_0}{T}\right)^{2/3}\left(1 - \frac{F^2}{g(T)}\right)^{1/3}, \tag{24}$$

where

$$g(T) = \left(\frac{2\alpha k_{\rm B}T}{e}\right)^2, \tag{25}$$

and

$$T^{\rm h}_0 = \frac{2.18\alpha^{1/2}N_{\rm S}^{1/2}}{k_{\rm B}N_0}. \tag{26}$$

Consequently, the conductivity, $\sigma^{\rm h}(F,T)$, varies as,

$$\sigma^{\rm h}(F,T) \propto \exp\left[-\left(\frac{T^{\rm h}_0}{T}\right)^{2/3}\left(1-\frac{F^2}{g(T)}\right)^{1/3}\right], \tag{27}$$

or

$$\ln\sigma^{\rm h}(F,T) \propto -\left(\frac{T^{\rm h}_0}{T}\right)^{2/3}\left(1-\frac{F^2}{g(T)}\right)^{1/3}. \tag{28}$$

When $F^2/g(T) \ll 1$, i.e. $e\alpha^{-1}F \ll 2k_{\rm B}T$, the expression for the conductivity can be written as

$$\ln\sigma^{\rm h}(F,T) \propto \ln\sigma^{\rm h}(0,T) + h(F)/f^{\rm h}(T), \tag{29}$$

where

$$\ln\sigma^{\rm h}(0,T) = -\left(\frac{T^{\rm h}_0}{T}\right)^{2/3}, \tag{30}$$

$$f^{\rm h}(T) = \left[\frac{1}{3}\left(\frac{T^{\rm h}_0}{T}\right)^{2/3}\frac{1}{g(T)}\right]^{-1}, \tag{31}$$

and

$$h(F) = F^2. \tag{32}$$

We notice that for $F = 0$ we regain the $\exp[-(T^{\rm h}_0/T)^{2/3}]$-behavior for the conductivity, $\sigma^{\rm h}(0,T)$, which was predicted by Triberis *et al* [43] using a different percolation approach, while $T^{\rm h}_0$ has the same dependence on the parameters involved as the corresponding quantity, $T^{\rm h}_0$, used there.

4. Field dependence of the conductivity for the low temperature small polaron hopping regime

At low temperatures (l), the average equilibrium transition probability, $W^{0{\rm l}}_{ij}$, reads [54]

$$W^{0{\rm l}}_{ij} = \gamma^{\rm l}_0\exp(-2\alpha R_{ij})\exp[-(|E_i|+|E_j|+|E_i-E_j|)/2k_{\rm B}T]. \tag{33}$$

Here, $\gamma^{\rm l}_0 = (\omega_0/\pi)[\pi J\exp(-2\varepsilon_2/\hbar\omega_0)/\hbar\omega_0]^2[(4\varepsilon_2/\hbar\omega_0)^{\Delta_{ij}/\hbar\omega_0}/(\Delta_{ij}/\hbar\omega_0)!]$.

In the presence of an electric field, F, and taking $E_i > E_j$, the average transition probability, $W^{\rm l}_{ij}(F)$ is given by

$$W^{\rm l}_{ij}(F) = \gamma^{\rm l}_0\exp(-\Re^{\rm l}), \tag{34}$$

where

$$\Re^{\rm l} = R'_{ij}(1+\lambda\cos\theta) + E^*_i, \tag{35}$$

where $E^*_i = E_i/k_{\rm B}T$. Here, $\Re^{\rm l}$ is the 'distance' between two states in a two-dimensional random array (one spatial

coordinate and one energy coordinate) (cf equation (35)) where the small polaron may be regarded to hop.

In order to evaluate the average nearest-neighbor range $\bar{\Re}^{\mathrm{l}}_{\mathrm{nn}}$ that determines $\sigma^{\mathrm{l}}(F,T)$, we have to obtain $N^{\mathrm{l}}(\Re^{\mathrm{l}})$. Using the following equation where the indices from E^*_i, R'_{ij} have been dropped. In reduced coordinates,

$$N^{\mathrm{l}}(\Re^{\mathrm{l}}) = \sum_{\theta=0,\pi}\left\{C^{\mathrm{l}}_1\left[\int_0^{\Re^{\mathrm{l}}}\left(\int_0^{R^{\mathrm{l}*}} \mathrm{d}R^{\mathrm{l}\prime}\right)\mathrm{d}E^*\right]\right\}, \tag{36}$$

where $C^{\mathrm{l}}_1 = k_{\mathrm{B}}TN_0/2\alpha$ and $R^{\mathrm{l}*} = (\Re^{\mathrm{l}} - E^*)/(1+\lambda\cos\vartheta)$, we obtain

$$N^{\mathrm{l}}(\Re^{\mathrm{l}}) = C^{\mathrm{l}}_2(\Re^{\mathrm{l}})^2, \tag{37}$$

where $C^{\mathrm{l}}_2 = C^{\mathrm{l}}_1/(1-\lambda^2)$.

From equation (27)

$$\Delta N^{\mathrm{l}}(\Re^{\mathrm{l}}) = C^{\mathrm{l}}_3\Re^{\mathrm{l}}, \tag{38}$$

where $C^{\mathrm{l}}_3 = 2C^{\mathrm{l}}_2$ and the average nearest-neighbor distribution becomes

$$P^{\mathrm{l}}_{\mathrm{nn}}(\Re^{\mathrm{l}}) = 2C^{\mathrm{l}}_2\Re^{\mathrm{l}}\exp[-C^{\mathrm{l}}_2(\Re^{\mathrm{l}})^2]. \tag{39}$$

Substituting $P^{\mathrm{l}}_{\mathrm{nn}}(\Re^{\mathrm{l}})$ into equation (22) we obtain

$$\bar{\Re}^{\mathrm{l}}_{\mathrm{nn}} = \frac{\Gamma(\frac{3}{2})}{(C^{\mathrm{l}}_2)^{1/2}}. \tag{40}$$

Thus,

$$\bar{\Re}^{\mathrm{l}}_{\mathrm{nn}} = \left(\frac{T^{\mathrm{l}}_0}{T}\right)^{1/2}\left(1-\frac{F^2}{g(T)}\right)^{1/2}, \tag{41}$$

where

$$g(T) = \left(\frac{2\alpha k_{\mathrm{B}}T}{e}\right)^2, \tag{42}$$

and

$$T^{\mathrm{l}}_0 = \frac{1.57\alpha}{k_{\mathrm{B}}N_0}. \tag{43}$$

The conductivity, $\sigma^{\mathrm{l}}(F,T)$, varies now as,

$$\sigma^{\mathrm{l}}(F,T) \propto \exp\left[-\left(\frac{T^{\mathrm{l}}_0}{T}\right)^{1/2}\left(1-\frac{F^2}{g(T)}\right)^{1/2}\right]. \tag{44}$$

Thus,

$$\ln\sigma^{\mathrm{l}}(F,T) \propto -\left(\frac{T^{\mathrm{l}}_0}{T}\right)^{1/2}\left(1-\frac{F^2}{g(T)}\right)^{1/2}. \tag{45}$$

When $F^2/g(T) \ll 1$, i.e. $e\alpha^{-1}F \ll 2k_{\mathrm{B}}T$, the expression for the conductivity can be written as

$$\ln\sigma^{\mathrm{l}}(F,T) \propto \ln\sigma^{\mathrm{l}}(0,T) + h(F)/f^{\mathrm{l}}(T), \tag{46}$$

where

$$\ln\sigma^{\mathrm{l}}(0,T) = -\left(\frac{T^{\mathrm{l}}_0}{T}\right)^{1/2}, \tag{47}$$

and

$$f^{\mathrm{l}}(T) = \left[\frac{1}{2}\left(\frac{T^{\mathrm{l}}_0}{T}\right)^{1/2}\frac{1}{g(T)}\right]^{-1}, \tag{48}$$

and

$$h(F) = F^2. \tag{49}$$

For $F = 0$ we regain the $\exp[-(T^{\mathrm{l}}_0/T)^{1/2}]$-behavior for the conductivity, $\sigma^{\mathrm{l}}(0,T)$, which has been predicted by Triberis *et al* [43].

5. Results and discussion

As we pointed out in section 1 several transport mechanisms and corresponding expressions for the temperature and electrical field dependence of the electrical conductivity have been proposed for the interpretation of the experimental findings. Aleshin *et al* [31] measured the electrical resistivity as a function of temperature, for a wide temperature range (25–300 K), and moderate electric fields up to 10^5 V m^{-1}, in polydiacetylene quasi-1D single crystals (PDA-PTS). Polydiacetylene is a unique, essentially one-dimensional, fully conjugated polymer. They reported that at $T > 50$ K nearest-neighbor hopping of small polarons is probable, consequently, these data are appropriate for the application of our theoretical results.

Before proceeding to the analysis of Aleshin *et al* data we have to discuss certain issues concerning our theoretical approach.

Since the early days of small polaron theory Emin [55] pointed out that: 'While the site-to-site small polaron jump rate provides sufficient information to determine the carrier's (trap-free) d.c. mobility in a crystal, in a disordered material a major statistical problem (at least, in principle) remains. Namely there are an overwhelmingly large number of inequivalent paths via which the carrier can move through the sample .... To determine the d.c. conductivity in such a situation is one of the tasks of the so-called percolation theory'. This was for Triberis and Friedman [45] and Triberis [45, 54, 56] the motivation and the challenge for a number of publications in the past on small polaron hopping transport i.e. to start from the microscopic expression for the jump rates of a small polaron hopping between two successive sites, and applying percolation theoretical considerations similar to those used in the case of variable range hopping, to obtain the macroscopic conductivity of the material as a function of the temperature and the electric field in 3D disordered systems.

In the present investigation we follow the same direction applying a different percolation approach [24].

The ohmic behavior of the system is characterized by the free-field temperature-dependent conductivity $\sigma(0,T)$. According to Triberis and Friedman [45] a $T^{-\epsilon/(\epsilon+r)}$ law governs the T-dependence of the small polaron hopping conductivity. Here, ϵ is the energy's dimensions (number), involved in the percolation condition, which is different at 'high' and 'low' temperatures, and r is 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 [26]. For the high-T and 3D case ($\epsilon = 2$ and $r = 3$) a $T^{-2/5}$ law was obtained [45]. For the low temperature 3D case ($\epsilon = 1$ and $r = 3$), the $T^{-1/4}$ law (Mott's law) was obtained [54, 57], which is also a widely accepted behavior. For the case of longitudinal conduction, at low-T, in thin films (2D case), ($\epsilon = 1$ and $r = 2$) a $T^{-1/3}$ law was obtained [58]. For the 1D case (the DNA case) at high temperatures ($\epsilon = 2$ and $r = 1$) a $T^{-2/3}$ law was obtained [43], while at low temperatures and 1D ($\epsilon = 1$ and $r = 1$), Triberis *et al* [46] obtained, consistently, a $T^{-1/2}$ law. The results presented in the present work, for

$F = 0$, are consistent with the $T^{-\epsilon/(\epsilon+r)}$ law, although the percolation treatment we use here is different.

In our analysis the charge carriers involved in the transport are small polarons and these carriers are responsible for the observed conductivity either at high or at low temperatures.

Due to the fact that small polaron hopping at low temperatures is due to few-phonon assisted hopping, instead of many-phonon assisted hopping (which is relevant at high temperatures), the intrinsic transition rate, for small polarons, at low-T and the corresponding percolation condition are similar to those which characterize variable range hopping of 'bare' (not small polarons) electrons localized in a disordered environment [59, 60]. Consequently, the qualitative behavior of the conductivity due to small polaron hopping at low temperatures is similar to the one of localized 'bare' electrons hopping via variable range hopping in a disordered material. Certain parameters involved in the analytical expressions in both cases, which are refereed to either small polarons or bare localized electrons, do not affect the qualitative behavior of the conductivity.

The study of the effect of the electric field along with the temperature variation, for the 3D case, for the high temperature regime reads [24], $\ln\sigma^{\mathrm{h}}(F,T) \propto -T^{-2/5}(1 - F^2/g(T))^{2/5}$, and for the low temperature case [25], $\ln\sigma^{\mathrm{l}}(F,T) \propto -T^{-1/4}(1 - F^2/g(T))^{1/2}$.

The present work, for the 1D case, predicts: for the high temperature regime, (equation (28)), $\ln\sigma^{\mathrm{h}}(F,T) \propto -T^{-2/3}(1 - F^2/g(T))^{1/3}$, and for the low temperature case, (equation (45)), $\ln\sigma^{\mathrm{l}}(F,T) \propto -T^{-1/2}(1 - F^2/g(T))^{1/2}$.

It is clear that dimensionality affects the behavior of the electrical conductivity.

Moreover, in our treatment we have taken the density of states $N(E_i) = N(E_j) = N_0$ to be constant [45, 61–63] over the energy range E_m, which is a frequently used assumption.

The effect of the form of the DOS on the DC conductivity of the small polaron hopping regime, in disordered systems, has been analytically investigated, by one of the authors [64] considering various energy-dependent DOS models: $N(E) = N_0 E^n$, $N(E) = N_0 + \lambda E (\lambda > 0)$, for the low-$T$ and high-T regime. In the present work we consider a constant DOS because the inclusion of the energy dependence of the DOS would give very complicated dependencies and shadow the interplay of the temperature and the electric field, which is the scope of the present work. Furthermore, comparison can be made with the $F = 0$ results [43] where the same form of the DOS has been used.

Another point that needs further clarification is the choice of the 'high' or the 'low' temperature region, where the corresponding equations (27), (29) and (44), (46) apply.

According to the mathematical analysis of the generalized molecular crystal model it is the condition $\hbar\omega_0 \ll k_{\mathrm{B}}T$ [53] that determines the 'high' or the corresponding 'low' temperature regime [53, 65]. This mathematical analysis leads to the evaluation of the intrinsic transition rate, which differs at high temperatures (multi-phonon assisted hopping), compared with that at low temperatures (few-phonon assisted hopping), and consequently, results to different percolation conditions. Thus, it is the condition $\hbar\omega_0 \ll k_{\mathrm{B}}T$ which distinguishes the

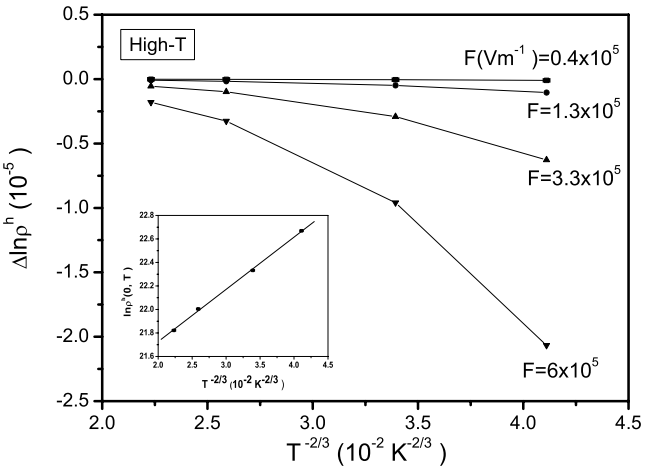

Figure 1. $\Delta\ln\rho^{\mathrm{h}}$-versus-$T^{-2/3}$ for the high temperature regime ($T = 120, 160, 240, 300$ K) and different electric field values: $F = 0.4 \times 10^5$, 1.3×10^5, 3.3×10^5 and 6×10^5 V m^{-1}. The inset presents the $\ln\rho(0,T)$-versus-$T^{-2/3}$ high temperature ohmic behavior.

'high' from the 'low' temperature regime. In both temperature regimes small polarons hop across the 1D medium via hops of variable ranges. Which temperature in real systems is indeed 'high' or 'low' depends on the system under study. This has been manifested (at high temperatures) by Triberis *et al* [43], for the case of DNA.

Proceeding to the analysis of Aleshin *et al* [31], we observe that their experimental data reported for temperatures in the range of 50 up to 300 K and electric field values up to 10^5 V m^{-1}, show a distinctively different behavior. For temperatures in the range of 50 up to 90 K we observe much stronger variations of the resistivity in terms of F/T compared with those in the temperature range of 120 up to 300 K, for the same electric field values. This observation allows us to consider the first region as the 'low' temperature region while the second as the 'high' temperature region.

For these temperature regions we apply, equation (29) and (46), respectively, for a representative value of the extend of the small polaron wavefunction, i.e. $\alpha^{-1} = 2$ Å [38, 66]. The condition $e\alpha^{-1}F \ll 2k_{\mathrm{B}}T$, holds for all the field and temperature values considered. In fact it holds for much higher electric field values.

In figure 1 we show the deviation of the electrical conductivity from the ohmic behavior i.e. $\Delta\ln\rho^{\mathrm{h}} = \ln\rho^{\mathrm{h}}(F,T) - \ln\rho^{\mathrm{h}}(0,T)$ as a function of $T^{-2/3}$, for different electric field values, for the high-T regime. The $\rho(0,T)$ experimental values have been obtained extrapolating, at $F = 0$ V m^{-1}, the experimental data presented in [31] as ρ-versus-F/T. Specifically, we show the $\Delta\ln\rho^{\mathrm{h}}$-versus-$T^{-2/3}$ for $T = 120, 160, 240, 300$ K and $F = 0.4 \times 10^5$ V m^{-1} (full squares), $F = 1.3 \times 10^5$ V m^{-1} (full circles), $F = 3.3 \times 10^5$ V m^{-1} (full triangles) and $F = 6 \times 10^5$ V m^{-1} (overturned full triangles). In the inset we show the $\ln\rho^{\mathrm{h}}(0,T)$-versus-$T^{-2/3}$ ohmic behavior. T_0^{h} is approximately equal to 296 K.

In figure 2 we show the deviation of the electrical conductivity from the ohmic behavior i.e. $\Delta\ln\rho^{\mathrm{l}} = \ln\rho^{\mathrm{l}}(F,T) - \ln\rho^{\mathrm{l}}(0,T)$ as a function of $T^{-1/2}$, for different

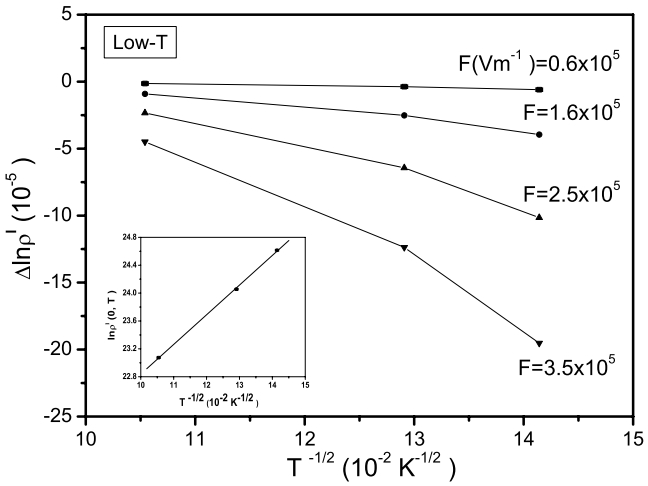

Figure 2. $\Delta \ln \rho^{\mathrm{l}}$-versus-$T^{-1/2}$ for the low temperature regime ($T = 50, 60, 90$ K) and different electric field values: $F = 0.6 \times 10^5$, 1.6×10^5, 2.5×10^5 and 3.5×10^5 V m^{-1}. The inset presents the $\ln \rho(0, T)$-versus-$T^{-1/2}$ low temperature ohmic behavior.

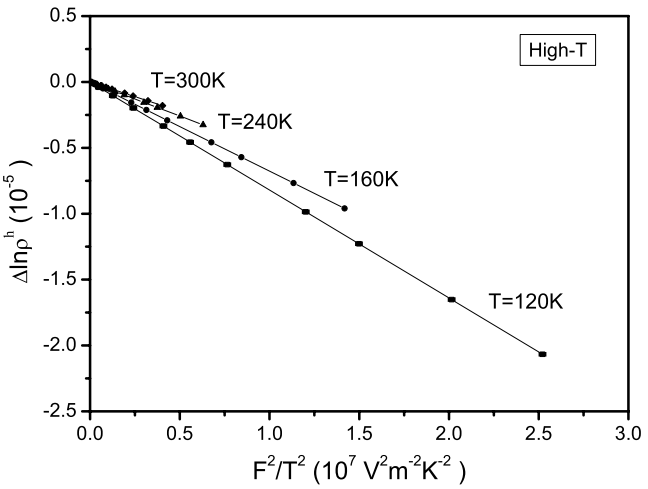

Figure 3. $\Delta \ln \rho^{\mathrm{h}}$-versus-$F^2/T^2$ for the high temperature regime ($T = 120, 160, 240, 300$ K).

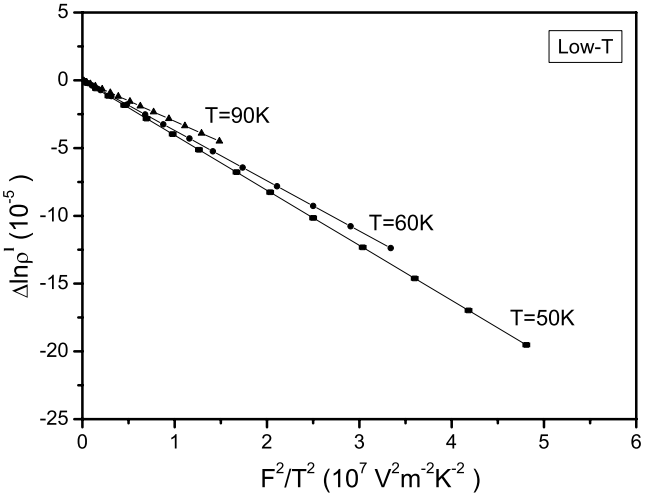

Figure 4. $\Delta \ln \rho^{\mathrm{l}}$-versus-$F^2/T^2$ for the low temperature regime ($T = 50, 60, 90$ K).

electric field values, for the low-T regime. Specifically, we show the $\Delta \ln \rho^{\mathrm{l}}$-versus-$T^{-1/2}$ for $T = 50$, 60, 90 K and $F = 0.6 \times 10^5$ V m^{-1} (full squares), $F = 1.6 \times 10^5$ V m^{-1} (full circles), $F = 2.5 \times 10^5$ V m^{-1} (full triangles) and $F = 3.5 \times 10^5$ V m^{-1} (overturned full triangles). In the inset we show the $\ln \rho^{\mathrm{l}}(0, T)$-versus-$T^{-1/2}$ ohmic behavior. T_0^{l} is approximately equal to 1814 K, which is comparable with the corresponding value reported by Aleshin *et al* [31] for the case of variable range hopping.

The fit of the experimental data, for $F = 0$, supports the idea of small polaron transport according to the $T^{-2/3}$ law, at high temperatures, and according to the $T^{-1/2}$ law, at low temperatures.

We notice that for very low electric fields the hopping conductivity conforms with the ohmic law. Increasing the electric field the conductivity presents non-ohmic characteristics due to the contribution of the electric field to the thermally assisted hopping conductivity. The transition from the ohmic to the non-ohmic behavior starts at smaller values of the electric field at lower temperatures. We also notice that the slope of the curves, which describes the rate of the increase of the conductivity, is greater at lower temperatures.

In figure 3 we show the interplay of the electric field and the temperature plotting $\Delta \ln \rho^{\mathrm{h}} = \ln \rho^{\mathrm{h}}(F, T) - \ln \rho^{\mathrm{h}}(0, T)$ as a function of F^2/T^2, for the high-T regime i.e. for $T = 300$ K (full rhombus), $T = 240$ K (full triangles), $T = 160$ K (full circles) and $T = 120$ K (full squares).

In figure 4 we show the interplay of the electric field and the temperature plotting $\Delta \ln \rho^{\mathrm{l}} = \ln \rho^{\mathrm{l}}(F, T) - \ln \rho^{\mathrm{l}}(0, T)$ as a function of F^2/T^2, for the low-T regime i.e. for $T = 90$ K (full triangles), $T = 60$ K (full circles) and $T = 50$ K (full squares).

Figures 3 and 4 show the competitive role of the electric field and the temperature in the behavior of the electrical conductivity. The increase of the electric field weakens the effect of the temperature driving the system from the thermally activated hopping to an electric field induced hopping conductivity.

Our theoretical results are also consistent with previous theoretical reports. It is well established [67–69] that in 1D systems there are rare fluctuations in the random distribution of localized states that create unavoidable highly resistive breaks on the hopping network. Raikh and Ruzin [69], investigated the fluctuations on the hopping conductance of 1D systems. They reported that breaks that are shaped as diamonds and have a temperature dependent size, have resistances that far exceed that of typical links. These breaks at very low electric fields have a dominant contribution to resistivity which exhibits an ohmic behavior. Fogler *et al* [27] theoretically investigated the effect of a finite electric field on the resistivity of a disordered one-dimensional system in the variable range hopping regime and shed more light to the role of these breaks. According to them as F increases the large resistors are progressively eliminated leading to the non-ohmic behavior of the resistivity. Moreover, according to Raikh and Ruzin [69],

the size of the diamond-shaped breaks is inversely proportional to temperature. Thus, for the same applied field, the higher the temperature, the smaller the breaks, and consequently the greater the conductivity, especially for the range of F values where the breaks have a dominant contribution to conductivity, i.e. for very low fields.

Recently, Ma *et al* [29] reported on the field dependence of the DC conductivity of 1D systems with off-diagonal disorder. As they pointed out, at very low electric fields, the field has almost no effect on the conductivity, while increasing the electric field a non-ohmic behavior is observed.

In the present investigation we focused upon moderate electric fields. Increasing further the electric field's intensity it is expected that the field assisted hopping will prevail upon the thermally assisted character of transport and conductivity will become totally field dependent. Additionally, in the present work we did not take into account correlation effects due to the energy of a common site (site j) between two successive hops, from i to j and from j to k. The inclusion of correlations could affect the behavior of the conductivity. These will be presented in the near future.

6. Summary

In summary, we developed a theoretical model for the temperature dependence of the electrical conductivity under the influence of moderate electric fields, when small polarons are transported in a disordered 1D environment, at high and low temperatures.

The analytical expressions for the electrical conductivity, obtained as a function of the electric field and the temperature, are applied on experimental findings concerning charge transport in polydiacetylene quasi-1D single crystals. It is concluded that small polaron hopping could be the responsible transport mechanism for the observed electrical conductivity. It is shown that the electric field and the temperature act competitively upon the behavior of the electrical conductivity. At very low electric fields the hopping conductivity conforms with the ohmic law while increasing the electric field the conductivity presents non-ohmic characteristics. The transition from the ohmic to the non-ohmic behavior starts at smaller values of the electric field at lower temperatures. The rate of the increase of the conductivity is greater at lower temperatures.

Our conclusions are also in accordance with theoretical results which are referred to variable range hopping.

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