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The effect of the density of states on the conductivity of the small-polaron hopping regime in disordered systems

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Abstract. We investigate the effect of the energy-dependent model densities of states on the DC conductivity of the small-polaron hopping regime in disordered systems. The DC conductivity behaviour is obtained for the high- and low-temperature cases, ignoring or taking into account correlations between bonds due to the energy of the common site.

1. Introduction

According to percolation theory (Ambegaokar *et al* 1971) the study of electron motion between localised states that are randomly distributed in energy and position, in disordered systems is equivalent to the study of the possibility of the passage of electric current through a network of impedances Z_{ij} that connect the different lattice sites i and j . The magnitude of these impedances depends on the site energies E_i , E_j and the distance R_{ij} between the two sites. The expression for the impedances Z_{ij} is given by

$$Z_{ij} = [(e^2/K)\Gamma_{ij}^0(R_{ij}, E_i, E_j)]^{-1} \quad (1)$$

where Γ_{ij}^0 is the average equilibrium transition rate, which depends exponentially on R_{ij} and E_i , E_j , e is the carrier's charge and K is the Boltzmann constant. Because of the exponential dependence of Z_{ij} , R_{ij} , E_i and E_j , the individual impedances change by many orders of magnitude. Therefore, the random network can be decomposed into three kinds of regions. (i) 'Short-circuits', which are characterized by the impedances $Z_{ij} < Z_c$. (ii) 'Open-circuits', which are characterized by high impedances $Z_{ij} > Z_c$ through which we have no passage of electric current. (iii) Impedances $Z_{ij} \simeq Z_c$, which interconnect small clusters of low impedances and form a conductive cluster through the material. The inverse of the magnitude of impedances $Z_{ij} \simeq Z_c$, Z_c^{-1} , characterizes the macroscopically observed conductivity of the material. The average equilibrium transition rate, Γ_{ij}^0 , is given by Ambegaokar *et al* (1971)

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

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where n_i^0 is the equilibrium occupation probability of the i th site and γ_{ij} is the intrinsic transition rate. This is of the general form

$$\gamma_{ij} = \gamma_0 \exp(-2\alpha R_{ij}) \exp\{f[(E_i, E_j)/2KT]\} \quad (3)$$

where $f(E_i, E_j)$ is a function of the site energies and α^{-1} is the spatial extent of the electronic wavefunction localized at a single site.

The analytical forms of the intrinsic transition rate for the small-polaron disordered regime, for the high- and low-temperature cases, are given by Emin (1975a, b), Triberis and Friedman (1981) and Triberis (1985a). Triberis and Friedman, in order to evaluate the intrinsic transition rate γ_{ij} in their studies, introduced the generalized molecular crystal model (GMCM). This has been described in some detail by Triberis and Friedman (1981) and Triberis (1985b). Here we summarize it very briefly.

The model Hamiltonian is

$$\langle m|H|n\rangle = \langle m|H_0 + V|n\rangle = E_{i,\{n_k\}} \delta_{ij} \delta_{\{n_k\},\{n_k'\}} + \langle m|V|n\rangle \quad (4)$$

where $\langle m|V|n\rangle$ is the overlap part, $|n\rangle = |i, \{n_k\}\rangle$ are the eigenstates of H , and H_0 is the zeroth-order (i.e. that for $J = 0$) Hamiltonian used by Holstein (1959). J is the standard electronic overlap integral of the tight binding theory (Holstein 1959). The corresponding eigenvalues are

$$E_{i,\{n_k\}} = E_i + \sum_k \hbar\omega_k(n_k + \frac{1}{2}). \quad (5)$$

Here, the totality of vibrational quantum numbers $\{\dots, n_k, \dots\}$ for the occupation of the site with position vector r_i , is represented by $\{n_k\}$,

$$E_i = \epsilon_i(0) - E_b(i) \quad (6)$$

is the electronic energy when the electron resides on site i , $\epsilon_i(0)$ is the local electronic energy and

$$E_b(i) = N^{-1} \sum_k (A_i^2/2M\omega_k^2) \quad (7)$$

is the small-polaron binding energy. N is the site number of lattice sites, and A_i is the electron-lattice interaction parameter.

Equations (6) and (7) show the essential features of the GMCM which are (i) a site-dependent local electronic energy, $\epsilon_i(0)$, and (ii) a site-dependent electron-lattice interaction parameter, A_i , and concomitant binding energy, $E_b(i)$. Then the expression for Z_{ij} takes the form

$$Z_{ij} = Z_0 \exp(\xi_{ij}) \quad (8)$$

where

$$Z_0 = KT/e^2\gamma_0 \quad (9)$$

$$\xi_{ij} = 2\alpha R_{ij} + E_{ij}/KT \quad (10)$$

and E_{ij} depends on the position of the sites i and j with respect to the Fermi level and on the form of the exponential dependence of the intrinsic transition rate on the site energies.

Thus the percolation condition is

$$Z_{ij} \leq Z_c \quad (11)$$

or

$$\xi_{ij} \leq \xi_c. \quad (12)$$

Pairs of sites with energies E_i and E_j being at a distance R_{ij} belong to the percolation path if they obey the percolation condition (12), which can also be written as

$$\frac{R_{ij}}{r_m} + \frac{E_{ij}}{E_m} \leq 1 \quad (13)$$

where

$$r_m = \xi_c/2\alpha \quad (14)$$

$$E_m = KT\xi_c. \quad (15)$$

Any link with $R_{ij} > r_m$ will violate the inequality (13) and not be on percolation cluster regardless of values of E_i , E_j . Any link with $|E_{ij}| > E_m$ will violate the inequality (13) and not be on percolation cluster regardless of the values of R_{ij} .

It is ξ_c that characterizes the DC conductivity of the material, which varies as

$$\ln \sigma \sim -\xi_c. \quad (16)$$

Triberis and Friedman (1981, 1985) and Triberis (1985a) presented, using percolation arguments, two methods for the calculation of the conductivity of the small-polaron hopping regime in a disordered system, where firstly they ignored (uncorrelated hopping) and secondly they took into account correlations between bonds due to the energy of the common site (correlated hopping). To avoid cumbersome calculations in these studies, and in many others related to the subject (Austin and Mott 1969, Ambegaokar *et al* 1971, Shklovskii and Efros 1976, Serota *et al* 1986, Triberis 1987, 1988a), the densities of states (DOS) used were taken to be a constant over the energy range E_m i.e. (1) $N(E_i) = N(E_j) = N_0 = \text{constant}$, where in the small-polaron studies E_i , E_j are the small-polaron binding energy occupying site i or j , respectively, assuming the disorder energy to be mainly polaronic i.e. $|E_i| \simeq E_b(i)$ and $E_j \simeq E_b(j)$ (Triberis and Friedman 1981), and E_m is the maximum energy involved in the percolation condition.

They found that the DC conductivity varies as

$$\ln \sigma \sim -\xi_c = [-(T_0/T)^\nu] \quad (17)$$

where the analytical form for T_0 , and the value of ν depend on the case considered i.e. uncorrelated or correlated hopping, high or low temperatures.

The DC conductivity behaviour of a variety of chalcogenide glasses such as $\text{Ge}_x\text{Sb}_y\text{Se}_z$, As_xSe_y (Mohan and Rao 1985), As-Te based glasses (Triberis 1986), phosphorushybrate and phosphotungstate glasses (Selvaraj and Rao 1988) and the DC conduction in RF sputtered SiO_2 films (Meaudre *et al* 1983, Meaudre and Meaudre 1984, Triberis 1987, 1988a) have been very satisfactorily analysed using Triberis and Friedman's model (1981) in which correlations are ignored.

Recently Triberis (1988b) suggested that, in order to interpret consistently the conductivity of V_2O_5 thin layers deposited from gels (Bullot *et al* 1984), it is necessary to take correlations into account. The same model (Triberis and Friedman 1985) has been used to interpret the DC conduction of ternary Nb-W oxides (Ruscher *et al* 1988).

Finally the importance of correlations in the study of the conductivity has been ascertained analyzing experimental results on the DC conductivity of V_2O_5 - P_2O_5 glasses (Triberis 1990).

It was noticed (Triberis and Friedman 1981) that a more appropriate choice of the DOS for the actual physical situation could be one that takes into account the fact that the lower-energy states are less 'dense'.

This will affect the DC conductivity. It is T_0 , as well as the T -dependence of the conductivity, which is determined by the exponent ν , that are affected by the particular choice of the form of the DOS. Pollak (1972, 1978), Böttger and Bryksin (1976) have also reported on the effect of the DOS on the behaviour of the DC conductivity, in the case of variable range hopping at low temperatures including correlations. Different fractional laws for the DC conductivity from those obtained using the constant DOS have been observed in amorphous, heavily doped and strongly compensated semiconductors (Pollak 1972, 1978 and Böttger and Bryksin 1976). They pointed out that for these materials a DOS of the form $N(E) = N_0 E^n$ is more appropriate. In their studies the lattice deformation due to the presence of the carrier as well as uncorrelated hopping and high temperatures have not been considered.

In order to investigate the effect of the DOS on the DC conductivity of the small-polaron hopping regime in disordered systems, in the present work we evaluate the DC conductivity using energy-dependent model DOS, consistent with the physical situation under study of the form: (II) $N(E) = N_0 E$, (III) $N(E) = N_0 E^n$, (IV) $N(E) = N_0 + \lambda E$, $\lambda > 0$ for uncorrelated hopping (section 2; low temperatures, $\hbar\omega_0 \gg KT$ section 2.1, and high temperatures, $\hbar\omega_0 \ll KT$, section 2.2) and correlated hopping (section 3; low temperatures, section 3.1, and high temperatures, section 3.2). For every form of the DOS used, N_0 is expressed in the appropriate for this form units. For completeness we also present previously published results for $N(E) = N_0 = \text{constant}$, for the cases of uncorrelated hopping (high temperatures) and correlated hopping (low and high temperatures).

In the following we assume that we have a band of localized states over the Fermi level ($E_F = 0$), taking into account the two possible configurations of the sites involved (i) $E_m > E_i > E_j > 0$ and (ii) $E_m > E_j > E_i > 0$, i.e. hops downwards or upwards in energy according to the energy distance of the two sites with respect to the Fermi level.

2. Uncorrelated hopping

2.1. The low-temperature regime

For this case the average number of impedances of magnitude Z_c or less connected to a given site, $P(Z_c)$, is given by (Triberis 1985a)

$$P(Z_c) = \theta = 2 \int_0^{E_m} N(E) \left(\int_0^{R'} 4\pi R^2 dR \right) dE \quad (18)$$

where

$$R' \equiv (r_m/E_m)(E_m - E). \quad (19)$$

Here, the indices have been dropped. For convenience, for θ we take the value 1.7 (Pollak 1972) and the limits of integrations are obtained from the corresponding percolation condition, which is

$$\frac{R}{r_m} + \frac{E}{E_m} \leq 1 \quad (20)$$

with

$$r_m \equiv \xi_c/2\alpha \quad (21)$$

$$E_m \equiv KT\xi_c. \quad (22)$$

It is ξ_c that determines the critical impedance, which is given by $Z_c = Z_0 \exp(\xi_c)$.

I. $N(E) = N_0 = \text{constant}$.

From (18) we obtain

$$T_0 = 6.49\alpha^3/N_0K \quad (23)$$

$$\nu = \frac{1}{4}. \quad (24)$$

This is in agreement with the results obtained by Ambegaokar *et al* (1971) and other workers, for the case of variable range hopping, with a difference appearing in the numerical factor in T_0 .

For energy-dependent DOS we have the following.

II. $N(E) = N_0E$.

$$T_0 = 5.71\alpha^{3/2}/N_0^{1/2}K \quad (25)$$

$$\nu = 2/5. \quad (26)$$

III. $N(E) = N_0E^n$.

$$T_0 = [0.27\alpha^3(n+1)(n+2)(n+3)(n+4)/N_0K^{n+1}]^{1/n+1} \quad (27)$$

$$\nu = \frac{n+1}{n+4}. \quad (28)$$

For $n = 0, 1$ we recover the results obtained using the model DOS (I) and (II) respectively.

IV. $N(E) = N_0 + \lambda E, \lambda > 0$.

For this case the percolation condition, $P(Z_c) = \theta$, is

$$\theta = \left(1 + \frac{0.2\lambda KT}{N_0}\xi_c\right) \frac{0.26N_0KT}{\alpha^3}\xi_c^4. \quad (29)$$

The enhancement of the conductivity due to the linear energy-dependent term, λE , appearing in the density of states, can be numerically obtained from (29). Pronounced deviations from the $T^{-1/4}$ behaviour due to this term do not seem to appear. In figure 1 we plot $-\xi_c(\sim \ln\sigma)$ against $T^{-1/4}$ for different values of $\lambda, (0, 10^{21}, 5 \times 10^{22}) \text{ eV}^{-2} \text{ cm}^{-3}$ applying (29). We have taken representative values of the parameters involved i.e. $\alpha^{-1} = 2 \text{ \AA}$, $N_0 = 5 \times 10^{20} \text{ eV}^{-1} \text{ cm}^{-3}$ and T in the range 50–120 K.

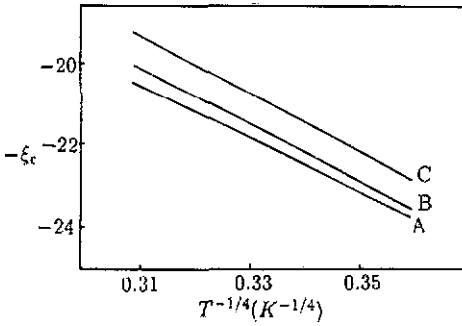


Figure 1. Plots of $-\xi_c$ ($\sim \ln \sigma$) against $T^{-1/4}$ for different values of λ . Curve A: $\lambda = 0$; B: $\lambda = 10^{21} \text{ eV}^{-2} \text{ cm}^{-3}$; C: $\lambda = 5 \times 10^{22} \text{ eV}^{-2} \text{ cm}^{-3}$.

2.2. The high-temperature regime

Here, $P(Z_c)$ is given by (Triberis and Friedman 1981)

$$P(Z_c) = \theta = \int_0^{E_m} \left[\int_{\Delta}^{\Delta'} \frac{1}{N_s} N(E - \Delta) N(E) \left(\int_0^{R'} 4\pi R^2 dR \right) dE \right] d\Delta \tag{30}$$

where

$$\Delta' \equiv (E_m + \Delta)/2 \tag{31}$$

$$R' \equiv (r_m/E_m)[E_m - (2E - \Delta)]. \tag{32}$$

The limits of integrations are obtained from the corresponding percolation condition

$$\frac{R}{r_m} + \frac{E_i}{E_m} + \frac{E_j}{r_m} \leq 1 \tag{33}$$

where

$$r_m \equiv (\xi_c/2\alpha) \tag{34}$$

$$E_m \equiv K'T\xi_c \tag{35}$$

$$K' \equiv 4/3K \tag{36}$$

$$\Delta \equiv E_j - E_i. \tag{37}$$

I. $N(E) = N(E - \Delta) = N_0 = \text{constant}$.

We obtain (Triberis and Friedman 1981)

$$T_0 = 8.5 N_s^{1/2} \alpha^{3/2} / N_0 K \tag{38}$$

$$\nu = 2/5. \tag{39}$$

For energy-dependent DOS we have the following.

II. $N(E) = N_0 E$.

$$T_0 = 6.45 N_s^{1/4} \alpha^{3/4} / N_0^{1/2} K \tag{40}$$

$$\nu = \frac{4}{7}. \tag{41}$$

III. $N(E) = N_0 E^n$.

$$\nu = \frac{2n + 2}{2n + 5} \tag{42}$$

while an analytical expression for T_0 cannot be obtained. For $n = 0, 1$ we recover, for ν , the results obtained using the model DOS (I) and (II), respectively.

IV. $N(E) = N_0 + \lambda E, \lambda > 0$.

For this case the percolation condition, $P(Z_c) = \theta$, is

$$\theta = [1 + 0.042(\lambda KT/N_0)^2 \xi_c^2] 0.023(N_0 KT/N_s^{1/2} \alpha^{3/2})^2 \xi^5. \tag{43}$$

This can be treated numerically.

3. Correlated hopping

3.1. The low-temperature regime

For this case, the average number of impedances of magnitude Z_c or less, connected to a site of energy $E_i, P(Z_c/E_i)$, is given by (Triberis 1985a)

$$P(Z_c/E) = \int_0^{E_i} N(E_j) dE_j \left(\int_0^{R'} 4\pi R^2 dR \right) + \int_{E_i}^{E_m} N(E_j) dE_j \left(\int_0^{R''} 4\pi R^2 dR \right) \tag{44}$$

where

$$R' \equiv (r_m/E_m)(E_m - E_i) \tag{45}$$

$$R'' \equiv (r_m/E_m)(E_m - E_j). \tag{46}$$

Averaging $P(Z_c/E_i)$ with respect to E_i , we obtain the percolation condition

$$P(Z_c) = \frac{\int_0^{E_m} P^2(Z_c/E_i) N(E_i) dE_i}{\int_0^{E_m} P(Z_c/E_i) N(E_i) dE_i}. \tag{47}$$

I. $N(E) = N_0 = \text{constant}$.

We obtain (Triberis 1985a)

$$T_0 = 17.8\alpha^3/N_0 K \tag{48}$$

$$\nu = \frac{1}{4}. \tag{49}$$

For energy-dependent DOS we have the following.

II. $N(E) = N_0 E$.

$$T_0 = 11.63\alpha^{3/2}/N_0^{1/2} K \tag{50}$$

$$\nu = \frac{2}{5}. \tag{51}$$

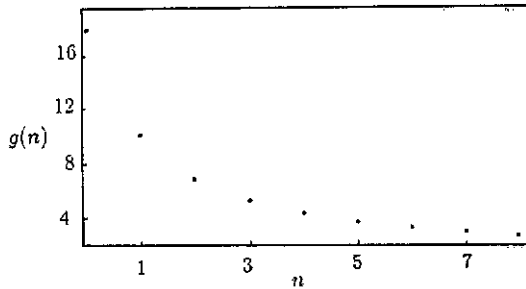


Figure 2. $g(n)$ -values for different n , (0, 1, 2, ..., 8).

III. $N(E) = N_0 E^n$.

$$T_0 = g(n)(\alpha^3/N_0 K^{n+1})^{1/(n+1)} \tag{52}$$

$$\nu = \frac{n + 1}{n + 4}. \tag{53}$$

We have evaluated $g(n)$ analytically. Because of its rather complicated form in figure 2 we present $g(n)$ for different values of n , (0, 1, 2, ..., 8). For $n = 0, 1$ we recover the results obtained using model DOS of the form (I) and (II), respectively.

IV. $N(E) = N_0 + \lambda E, \lambda > 0$.

For this case the percolation condition, $P(Z_c) = \theta$, is

$$\theta = \left(1 + 0.13 \frac{\lambda K T}{N_0} \xi_c \right) 0.095 \frac{N_0 K T}{\alpha^3} \xi_c^4. \tag{54}$$

This can be treated numerically.

3.2. The high-temperature regime

Here, $P(Z_c/E_i)$ is given by (Triberis and Friedman 1985, Triberis 1985a)

$$P(Z_c/E_i) = \int_0^{E'} N(E_j) dE_j \left(\int_0^{R'} 4\pi R^2 dR \right) + \int_{E_i}^{E'} N(E_j) dE_j \left(\int_0^{R'} 4\pi R^2 dR \right) \tag{55}$$

where

$$R' \equiv \frac{r_m}{E_m} (E_m - E_i - E_j) \tag{56}$$

$$E' \equiv E_m - E_i. \tag{57}$$

$P(Z_c)$ is given by (47) using $P(Z_i/E_i)$ given by (55).

I. $N(E) = N_0 = \text{constant}$.

We obtain (Triberis and Friedman 1985, Triberis 1985a)

$$T_0 = 12.5 \alpha^3 / N_0 K \tag{58}$$

$$\nu = \frac{1}{4}. \tag{59}$$

For energy-dependent DOS we have the following.

$$\text{II. } N(E) = N_0 E.$$

$$T_0 = 4.2\alpha^{3/2}/N_0^{1/2}K \quad (60)$$

$$\nu = \frac{2}{3}. \quad (61)$$

$$\text{III. } N(E) = N_0 E^n$$

$$\nu = \frac{n+1}{n+4} \quad (62)$$

while an analytical expression for T_0 cannot be obtained. For $n = 0, 1$ we recover, for ν , the results obtained using model DOS of the form (I) and (II), respectively.

$$\text{IV. } N(E) = N_0 + \lambda E, \lambda > 0.$$

For this case the percolation condition, $P(Z_c) = \theta$, is

$$\theta = \left(1 + 0.74 \frac{\lambda KT}{N_0} \xi_c\right) 0.13 \frac{N_0 KT}{\alpha^3} \xi_c^4. \quad (63)$$

This can be also treated numerically.

4. Comparison with other theories—conclusions

In the present work we investigated the effect of the DOS of the form $N(E) = N_0 E^n$ and $N(E) = N_0 + \lambda E, \lambda > 0$, on the DC conductivity of the small-polaron hopping regime in disordered systems. Analytical expressions for the DC conductivity, which generally varies as $\ln \sigma \sim -(T_0/T)^\nu$, have been obtained.

For the case of correlated hopping our result for ν (i.e. $\nu = \frac{n+1}{n+4}$) agree with those predicted by other workers (Pollak 1972, 1978, and Böttger and Bryksin 1976), for the case of variable range hopping.

In their studies the lattice deformation due to the presence of the carrier, which is a basic ingredient of the present work, have not been taken into account. Neither they nor any other workers have reported analytical expressions for T_0 , as we do, for energy-dependent DOS

According to the present work, for the same temperature range (i.e. low or high T): (i) the exponent ν , which determines the T -dependence of the conductivity, increases with n , and (ii) T_0 diminishes with n .

Finally, for a given model DOS when correlations are taken into account ν does not change with the temperature range. For uncorrelated hopping, ν changes with the temperature range and in particular ν (high T) $>$ ν (low T).

In this work we investigated the effect of energy-dependent model DOS on the DC conductivity of the small-polaron hopping regime in disordered systems. The choice of the DOS is expected to affect not only the DC conductivity but transport coefficients such as the thermoelectric power as well. This will be presented in a future publication.

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