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J. Phys.: Condens. Matter 7 (1995) 8467–8475. Printed in the UK

Numerical investigation of non-Ohmic hopping conduction in macroscopically non-uniform thin layers: weak electron–phonon interaction

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Received 17 January 1995, in final form 28 March 1995

Abstract. The current–field and the conductivity–field characteristics of random hopping non-uniform systems in the limit of a weak electron–phonon coupling (band-like hopping transport in disordered solids) have been calculated numerically within the Bottger–Bryksin model. In particular, we consider thin layers placed between two planar Ohmic contacts, with exponential spatial variations in the total centre concentration over the layer thickness. We find that firstly for r-hopping transport in spatially uniform layers the current decreases with increasing field, reaching a constant saturation value, and on increasing the degree of non-uniformity the saturation current becomes lower and secondly both uniform and non-uniform r–ε-hopping systems are almost exactly Ohmic in the investigated field range; conductivity decreases on increasing the degree of non-uniformity of the centre distribution. The dependences on the degree of the layer non-uniformity in the limit of weak electron–phonon coupling are quite different from the dependences in the case of strong electron–phonon coupling.

1. Introduction

The non-Ohmic hopping conductivity for a strong electron–phonon interaction has been discussed in a number of papers (Böttger and Wegener 1984, Böttger *et al* 1985, 1986, Böttger and Bryksin 1979, 1980, 1985, Nguyen Van Lien and Shklovskii 1981, Fishchuk 1982, Mancini *et al* 1993). The main results may be summarized as follows. For r-hopping transport in macroscopically uniform systems the differential conductivity σ decreases with increasing applied external field E at relatively low fields, whereas for sufficiently high fields the conductivity σ increases rapidly. In non-uniform systems (in particular in systems with an exponential dependence of the hopping centre concentration on the distance from the contact, (Mancini *et al* 1993)) there may occur conductivity saturation instead of rapid growth, so that the system becomes Ohmic, at least up to fields consistent with the assumption of constant carrier concentration. For r–ε-hopping transport in unifrom systems the conductivity–field characteristics have an N-like shape; a local maximum is followed by a local minimum of the differential conductivity, and then an exponential increase occurs, at least at not too low temperatures (Böttger *et al* 1986). Here again the influence of

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the spatial non-uniformity of the sample is remarkable. In particular, for sufficiently non-uniform layers, there is no conductivity maximum at low fields, but the local negative minimum is followed by a saturated small positive value (Mancini *et al* 1993).

In view of the marked dependence of the conductivity–field characteristics on the spatial non-uniformity of the hopping centre distribution in the case of strong electron–phonon coupling, it seems interesting to investigate the predictions of the model Böttger and Bryksin also for the case of a weak electron–phonon interaction. Some preliminary results have been briefly reported by Mancini *et al* (1991). In the present paper we describe in detail the results of the numerical simulations of the current–field and conductivity–field characteristics for r-hopping, and r–ε-hopping transport in the limit of weak electron–phonon coupling (i.e. for band-like transport) in thin layers placed between two planar Ohmic contacts, with an average density of hopping centres (section 3) exponentially decreasing in space. Section 4 contains concluding remarks.

Before presenting our results, however, we recall briefly the basic equations describing hopping conductivity (Böttger and Bryksin 1985).

2. Basic equations and simulation algorithm

For an electric field E of arbitrary strength, the density j of the DC hopping current is given by (Böttger and Bryksin 1985)

$$j = \frac{1}{2\Omega}\sum_{m,m'}(\boldsymbol{R}_m - \boldsymbol{R}_{m'})i(m', m) \tag{1}$$

where $\boldsymbol{R}_m$ is the position of the mth hopping site, Ω is the volume of the system, and $i(m', m)$ is the current running from site m' to site m. The latter may be written as

$$i(m', m) = eW_{m'm}[\rho_{m'}(1-\rho_m)\exp(\beta V_{m'm}/2) - \rho_m(1-\rho_{m'})\exp(-\beta V_{m'm}/2)] \tag{2}$$

where $V_{m'm} = V_{m'} - V_m$, $V_m = \varepsilon_m + eu_m$, ε_m is the energy of the mth site, u_m is the potential of the external field E at the point $\boldsymbol{R}_m$, $\beta = 1/kT$, k is the Boltzmann constant, T is the temperature, ρ_m is the occupation probability of site m, $W_{m'm}$ is the symmetrized hopping probability, and e is the elementary charge. The latter in the limit of a weak electron–phonon interaction may be expressed as

$$W_{m'm} = W_0'[\sinh|V_{m'm}|\beta/2]^{-1}\exp(-2\alpha|\boldsymbol{R}_{m'm}|) \tag{3}$$

where α is the reciprocal Bohr radius, and the pre-factor W_0 depends only weakly on the external electric field E, as well as on the site position $\boldsymbol{R}_m$ and energy ε_m.

Our numerical results have been obtained from equations (1)–(3), where the occupation probabilities ρ_{ij} are the solution of

$$\sum_{m'} i(m', m) = 0 \tag{4}$$

together with the normalization condition

$$N^{-1}\sum_m \rho_m = n \tag{5}$$

where n is the concentration of electrons in the system, and N is the total site number within volume Ω. The algorithm that we used follows the general guidelines described by Böttger and Wegener (1984). Our slight modification consists in applying the periodic boundary conditions in directions perpendicular to the external electric field. In particular, in the directions perpendicular to the applied field we have constructed an infinite series of the

replicas of the simulation box, and the distances between the hopping sites were calculated using the minimum-image convention (see, e.g., Wood and Parker (1957)). Each of the curves shown below consists of 100 points, i.e. has been obtained by resolving equations (1)–(5) at $k\ \Delta E$, $k = 1,\ldots,100$ equidistant values within the field range considered.

3. Numerical results and discussion

First let us specify the values of the parameters, which are common for the curves. As in our previous paper (Mancini *et al* 1993, hereafter referred to as I) we made the following assumptions

(a) The concentration n of electrons in the system is equal to 0.5 and does not depend on the applied electric field.

(b) We deal with relatively diluted systems with $\alpha\mathcal{N}^{-1/3} = 15.0$ ($\mathcal{N} = N/\Omega$).

(c) To show the influence of the spatial variations of the average centre concentration on the current–field and conductivity field characteristics, we choose for our model simulations an exponential spatial dependence of the average density $N_h(x)$, of hopping centres given by

$$N_h(x) = N_0(D)\exp(-x/D) \tag{6}$$

where x is the distance measured from one of the electrodes ($0 < x < L$, with L the layer thickness) and D is a characteristic length of the site concentration decay. Such a distribution can originate from diffusive or radiative processes. The ratio L/D may be referred to as the degree of non-uniformity of the distribution. In what follows we consider L/D in the range from 0.0 to 2.5. The centre concentration $N_h(0)$ at $x = 0$ equal $N_0(D)$, and for each value of D is chosen in such a way that the total number N of hopping centres within the simulation box is constant ($N = 500$ for r hopping, and $N = 250$ for r–ε hopping). The results obviously do not depend on the layer polarization. As far as the influence of the random initial distribution is concerned, the characteristics for various random generations of the centre distribution lead to some differences in numerical values, leaving the curve shape qualitatively unchanged. In a similar way to the strong electron–phonon interaction (cf I), the quantitative discrepancies are most pronounced for macroscopically uniform distributions and almost do not exist for highly non-uniform structures.

3.1. r- and r–ε-hopping transport; uniform systems

To the best of our knowledge, no extensive calculations of the current–field characteristics for a weak electron–phonon interaction have been performed. Only two such characteristics appear in the literature (those given by Böttger and Wegener (1984) and Böttger and Bryksin (1985)), both calculated for the case of r-hopping transport. The curves differ markedly (figures 1A and 1B, respectively), and no explanation of the discrepancies is given. Our current–field characteristics for uniform systems without any spread in the centre energy ($\sigma = 0.0kT$) agree with those obtained by Böttger and Wegener (1984) (figure 1A). The curve in figure 1A requires some comments. For $\sigma = 0.0kT$, equation (3) gives an infinite value of the symmetrized hopping probability and thus cannot be used for the calcualtion of the current at $E \simeq 0$. In order to perform the first step ΔE, we use the corresponding expression for a strong electron–phonon interaction: $W_{mm'} = W_0\exp(-2\alpha|R_{mm'}|)$. Thus at $E = 0$ the current zero (by definition), and at $E = 1 \times \Delta E$ it assumes immediately a finite value, so that the initial linear current increase seen in figure 1A has no physical meaning. The calculations performed with various values of the field increment ΔE show, however,

that neither the low-field limit of the current nor the characteristics' shape scarcely depend on ΔE, if only the same numerical precision is held.

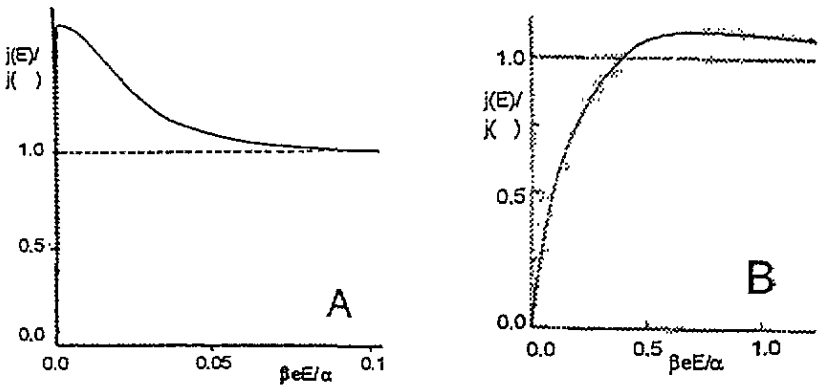

Figure 1. Current–field characteristics for a weak electron–phonon interaction in macroscopically uniform layers with no spread in energy levels: A, after Böttger and Wegener (1984); B, after Böttger and Bryksin (1985).

As far as the curve in figure 1B (quoted here after Böttger and Bryksin (1985) and described therein as a characteristic obtained 'without any energetic disorder') is concerned, we were unable to reproduce its shape within simulations with σ equal exactly to zero. From the physical point of view, however, energy distributions with σ-values in the range $0 < \sigma < 1kT$ could be considered as distributions with 'no' energy spread of the levels (σ is defined here as the sixfold variance of the Gaussian energy distribution). By applying the algorithm for r–ε hopping with $\sigma \ll 1kT$, one avoids formally the division by zero in (3). The denominator, however, turns out to be a small number, leading again to numerical problems, so that the $\sigma \to 0$ limit is still not reachable. No convergence has been obtained for $\sigma \leqslant 0.025kT$ above $E' \approx 0.005$ ($E' = \beta eE/\alpha$). For the energy distribution width $\sigma = 0.25kT$ we were able to calculate the j–E characteristic for $N = 250$ hopping centres up to about $E' \approx 0.01$. The typical shape of the characteristics obtained in this way for several random generations of the centres within the simulation box is similar to that of Böttger and Bryksin (1985) (figure 1B) and thus we conclude that the latter was probably obtained for an r–ε-hopping system with a very narrow energy centre distribution.

After commenting on the literature results for unifrom systems let us turn to the presentation of our results on the influence of the spatial non-uniformity of the centre distribution. The current–field characteristics for r-hopping transport (calculated by using the strong electron–phonon interaction formula for the first step at $E' = 0$), and for r–ε-hopping transport, both depending on the layer non-uniformity parameter L/D, are discussed in sections 3.2 and 3.3, respectively.

3.2. r-hopping transport; non-uniform systems

Figure 2 shows the current–field and the differential conductivity–field characteristics for various values of the non-uniformity parameter L/D in the range 0.0–2.5, as a function of a dimensionless field argument $E' = eE\beta/\alpha$, $\beta = 1/kT$. The current–field characteristics (figure 2A) are normalized to the initial value of the current $j_1 = j(E = 0.01)$. In each case, j_1 was also the maximum value of the current, so that $j_1 = j_{max}$. This maximum value is followed by a current decay down to the minimum (saturation) value. The quantities which

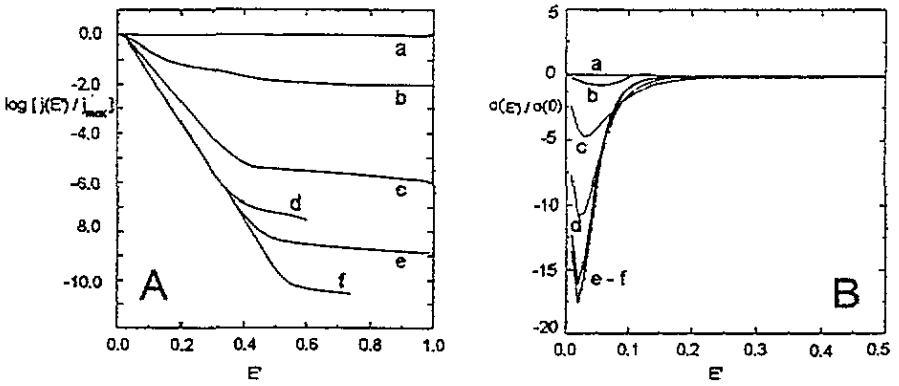

Figure 2. A, current–field characteristics for various values of the non-unformity parameter L/D: curve a, $L/D = 0.0$; curve b, $L/D = 0.5$; curve c, $L/D = 1.0$; curve d, $L/D = 1.5$; curve e, $L/D = 2.0$; curve f, $L/D = 2.5$. B, differential conductivities $\sigma(E')/\sigma(E' = 0)$ calculated from characteristics a–f of A. Weak electron–phonon coupling; $N = 500$.

depend markedly on L/D are the values of the saturation current, and the field at which the saturation occurs. With increasing sample non-uniformity the current reaches its saturation value for a systematically increasing field. The final current values is lower for higher L/D. Thus, as far as the current–field characteristics are concerned, increasing L/D changes the curves only qualitatively. The effect is, however, extremely strong (the saturation currents differ by about ten orders of magnitude as L/D changes in the range 0.0–2.5). For a strong electron–phonon interaction (cf I), the presence of the spatial non-uniformity changes the characteristics also qualitatively.

Figure 2B shows differential conductivities $\sigma(E')$ corresponding to the j versus E' characteristics in figure 2A (normalized to $\sigma(E' = 0)$). The curves corresponding to different L/D-values retain their typical shape; the conductivity decreases, reaches its minimum and then approaches a saturated small negative value (at E_{sat}). The depth of the conductivity minimum depends dramatically on L/D. E_{sat} also depends on L/D and is lower for more non-uniform systems. This behaviour is quite different from the case of a strong electron–phonon interaction, where all the σ versus E' curves with $L/D > 1.0$ almost coincide (cf I), so that the relative differential conductivity variations as a function of the applied field do not depend on the degree of non-uniformity of the centre distribution, the conductivity satuaration field E'_{sat} is almost L/D independent, and saturation takes place only in sufficiently non-uniform systems. In a similar way to in I, we have constructed the histograms of the average occupation probabilities $P(i)$ of the centres as a function of E', for each value of L/D. The average occupation probabilities $P(i)$, $i = 1,\ldots, 10$ were calculated as the arithmetic media over subsequent slices $L/10$ thick. Figures 3 and 4 show several such histograms (for $L/D = 0.0$ and 2.5, respectively) at several values of E'. The histograms in figures 3A and 4A show the average centre occupations for a very low electric field ($E' = 0.01$). $P(i)$ is approximately constant over the layer thickness for the uniform centre distribution (figure 3A) and increases with increasing i for the centre average concentration decaying with increasing i (figure 4A). Nowhere is the average occupation close to 1. Increasing field leads to marked changes in $P(i)$. For $L/D \geqslant 1.0$ the field range in which the average occupation in the central part of a layer tends to about one (over about 10% of the layer thickness) corresponds to the conductivity decay towards its minimum value. On increasing the field, although the spatial extension of the region with

almost fully occupied centres increases, the differential conductivity begins to increase. In contradistinction to the case of a strong electron–phonon interaction, the increasing field is not able to enforce the effective carrier motion, and the current always decays, so that the conductivity has a small negative value up to the maximum fields (assumption of constant carrier concentration). The described behaviour of non-uniform systems is only quantitatively different from the behaviour of macroscopically uniform samples, where the conductivity also remains negative, but the conductivity minimum is much less deep than for the non-uniform samples. The average occupation histograms show that, for uniform systems, full occupation is never reached. Up to the highest fields, the average centre occupation does not exceed 0.7 in any of the slices. The shallow conductivity minimum (occurring at $E' = 0.06$), however, corresponds to somewhat higher centre occupations (about 10% of the layer thickness has an average centre occupation of about 0.8 in the field range $0.05 < E' < 0.1$).

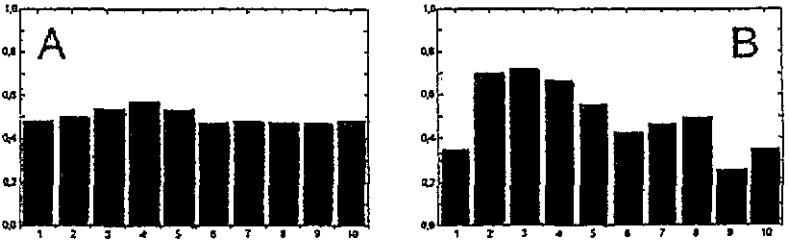

Figure 3. Histograms of the average occupation probability for $L/D = 0.0$: A, $E' = 0.01$; B, $E' = 1.0$.

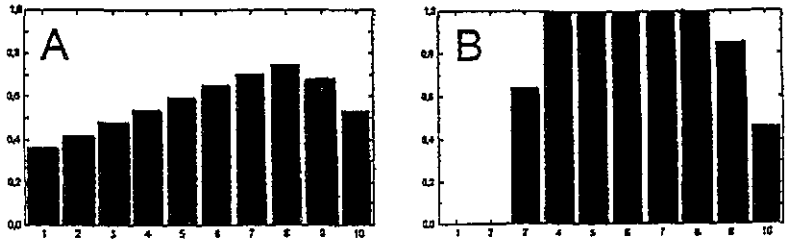

Figure 4. Histograms of the average occupation probability for $L/D = 2.5$: A, $E' = 0.01$; B, $E' = 0.7$.

3.3. r–ε-hopping transport; non-uniform systems

As far as r–ε-hopping transport is concerned, no published results are known to the present authors. In this case, as mentioned in section 3.1, great numerical problems appear. We were able to perform numerically reliable calculations for random systems containing only $N = 250$ hopping centres, rarely arriving at fields higher than $E' = 0.01$ (for typical values of α and β this corresponds to about 10^4 V m^{-1}). A sample containing such a small number of centres, distributed at random in space, as well as in energy, is a rather poor representation of real systems, even if periodic boundary conditions are imposed. Thus, for each set of input parameters, the calculations were repeated for several random initial generations of the centres within the simulation box. It turned out that, although

the numerical values of the calculated currents depend strongly on the random position generation, the qualitative tendencies in changing σ and L/D are reproducible. In the figures below, we show exemplary but typical curves.

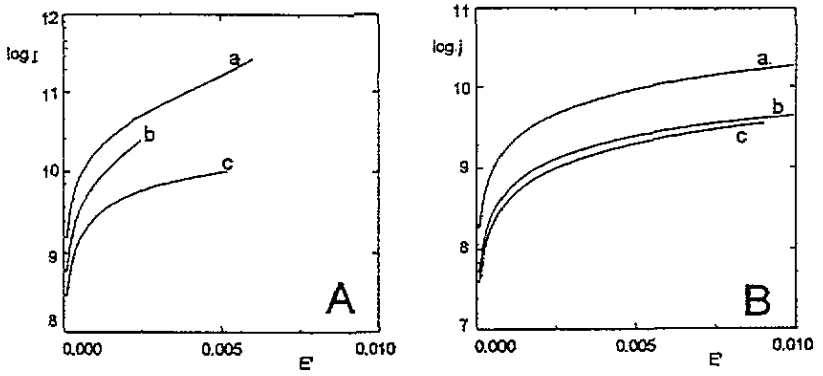

Figure 5. Current–field characteristics for r–ε-hopping in the limit of a weak electron–phonon interaction: A, $\sigma = 0.25kT$; B, $\sigma = 12.0kT$. Curves a, $L/D = 0$; curves b, $L/D = 1.0$; curves c, $L/D = 1.5$. Number of centres in the simulation box $N = 250$.

Figure 5 shows the current–field characteristics for various widths σ of energy distribution of the centres (figures 5A and 5B), and various values of the macroscopic non-uniformity parameter L/D (curves a, b and c). In all the cases the current increases with increasing field. Such behaviour is quite different from the results obtained for ideal ($\sigma = 0.0kT$) r hopping. In the latter case the current decreases monotonically with increasing field, reaching a low saturation value at much higher fields. In the field range ($E' \leqslant 0.01$) considered here, ideal ($\sigma = 0.0kT$) r-hopping currents remain almost constant, decreasing slightly on increasing the field (by a few per cent). Such behaviour can be explained by the field dependence of the symmetrized transition probability $W_{mm'}$ between the centres. Without any energy disorder of hopping centres, the transition probability decreases simply on increasing the field as $\sinh[e|u_m - u_{m'}|\beta/2]^{-1}$, and the calculated currents are monotonically decreasing functions of E'. In this case, only the nearest-neighbour hops are effective. The presence of the energy disorder allows variable-range hopping, which promotes the carrier transport effectively; so in the low-field region the currents increase with increasing field. As seen from figure 5, for each value of σ the currents become lower when the L/D parameter increases, because of harder hops in the lowest centre concentration region.

Figure 6 shows the dependence of the current–field characteristics on the energy distribution width σ, or alternatively on the temperature. Here the currents become lower on increasing σ/kT. In fact, for a wider energy spread of the centres there are few neighbouring centres of comparable energy, the deeper centres are occupied, and the shallower centres, although empty, are too greatly separated in energy, so that a high activation energy is necessary. As a result, for wider energy distributions the electron hops become more difficult.

Figure 7 shows differential conductivities calculated from several current–field characteristics on figures 5 and 6. The systems are almost Ohmic, and the dependences of the conductivity values on L/D and σ agree with the preceding discussion. In contradistinction, differential conductivity in the case of a strong electron–phonon interaction, instead of being constant, decays markedly over the same field range (cf I).

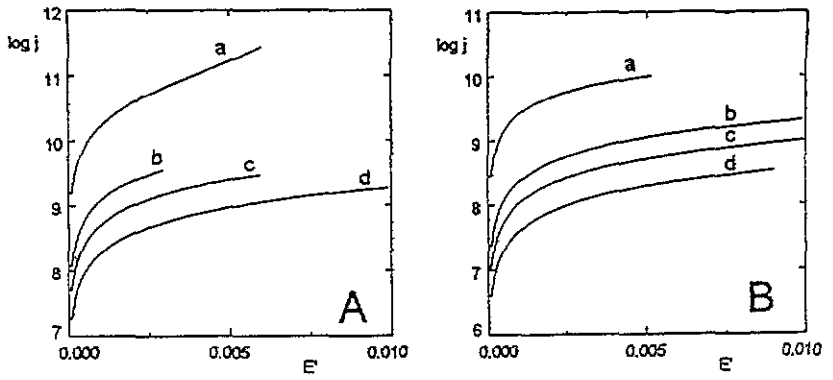

Figure 6. Current–field characteristics for r–ε hopping in the limit of a weak electron–phonon interaction: A, $L/D = 0$; B, $L/D = 1.5$. Curves a, $\sigma = 0.25kT$; curves b, $\sigma = 3.0kT$; curves c, $\sigma = 6.0kT$; curves d, $\sigma = 12kT$. The number N of centres in the simulation box equals 250.

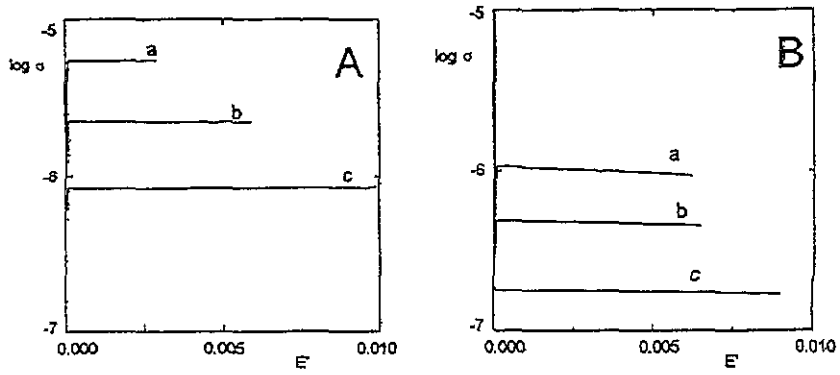

Figure 7. Differential conductivities calculated from several current–field characteristics: A, $L/D = 0.0$; B, $L/D = 1.5$, Curves a, $\sigma = 3.0kT$; curves b, $\sigma = 6.0kT$; curves c, $\sigma = 12.0kT$.

The average centre occupations are only slightly field dependent (figure 8). The most pronounced changes occur for low energy distribution width σ, and high non-uniformity degree L/D (cf the histogram in figure 8B). Here again the appearance of the bulk region of enhanced average occupation leads to some decrease in the conductivity (curve a in figure 7B).

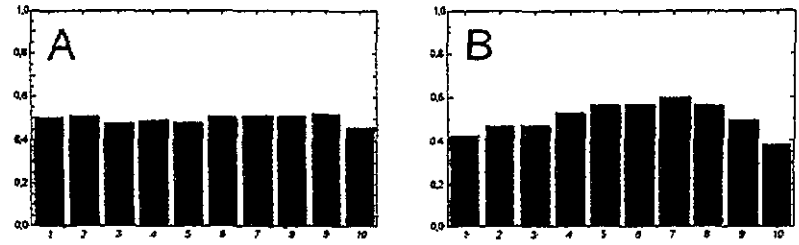

Figure 8. Histograms of the average occupation probability for $\sigma = 3.0kT$, $L/D = 1.5$: A, $E' = 0.007$; B, $E' = 0.01$.

4. Concluding remarks

The model of hopping transport in the limit of weak electron–phonon interaction proposed by Böttger and Bryksin turns out to be very difficult for numerical treatment. In order to obtain fully realistic results, one should perform calculations involving many more centres in the simulation box. Moreover, the mathematical formulation of the problem is not applicable directly to the limit of ideal r-hopping (discrete energy level), at least at $E' \approx 0$. We think, however, that even somewhat oversimplified numerical results reveal at least qualitative behaviours of real physical systems. On the basis of the results discussed above, we can state that there are significant quantitative and qualitative changes due to quasi-continuous variations in the average centre concentration over the sample thickness in the case of a weak electron–phonon interaction. The dependences on the degree of the layer non-uniformity in the limit of weak electron–phonon coupling are quite different from the dependences in the case of strong electron–phonon coupling.

Acknowledgments

The work has been sponsored by KBN, grant 2 P 302 16004. The possibility of performing our simulations at Centro Interdipartimentale di Calcolo of the Camerino University (Italy) and at the Task Computer Centre (Gdansk, Poland) is kindly acknowledged.

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