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Numerical investigation of non-ohmic hopping conduction in macroscopically inhomogeneous thin layers: strong electron–phonon interaction

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Abstract. The current–field and conductivity–field characteristics of random r hopping as well as r–$\mathcal{E}$ hopping systems with a strong electron–phonon coupling have been calculated numerically and discussed by Böttger and co-workers. They have assumed a random but macroscopically homogeneous distribution of centres over the sample. However, in most real cases an extremely thin layer can hardly be thought to be macroscopically homogeneous and the local hopping-centre density should depend on the distance from the electrodes. In the present work we apply the Böttger–Wegener procedure to random r hopping and r–$\mathcal{E}$ hopping systems with macroscopic average density exponentially dependent on the distance from the contact. Only a strong electron–phonon coupling will be considered, i.e. we shall consider small-polaron transport in a disordered solid. The influence of the inhomogeneity in the centre distribution on current–field and conductivity–field characteristics may be summarized as follows. Firstly, for r-hopping transport in homogeneous layers, Böttger and Wegener observe a decrease in the differential conductivity with increasing field, its local minimum being followed by an exponential increase; we confirm the result for homogeneous systems, whereas for inhomogeneous systems we find that, on increasing the degree of the site distribution inhomogeneity the local conductivity minimum is no longer followed by an exponential conductivity increase, but the system becomes ohmic (conductivity saturation), at least up to the fields consistent with the assumption of constant carrier concentration. Secondly, for r–$\mathcal{E}$ hopping in homogeneous layers, Böttger *et al* observe for not too high an energy spread in the hopping centres that the local conductivity maximum occurring just after the ohmic region is followed by a local minimum, the latter being followed by an exponential conductivity increase for still higher fields. At low temperatures, which is equivalent to a larger energy spread in the hopping centres, the height of the conductivity maximum increases, whereas the local minimum becomes shallower and disappears completely for sufficiently low temperatures. We find that in sufficiently non-uniform systems, for all temperatures, there is no longer a conductivity maximum, but the narrow low-field ohmic range is followed immediately by the conductivity decrease, and after reaching a negative local minimum the conductivity increases to a very low positive constant value, at least up to the fields consistent with the assumption of a field-independent carrier concentration.

1. Introduction

The non-ohmic hopping conductivity of disordered systems has been discussed in a number of papers (Böttger and Wegener 1984, Böttger *et al* 1985, 1986, Böttger and Bryksin 1979,

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1980, 1985, Nguyen Van Lien and Shklovskii 1981, Fishchuk 1982), both for r hopping (nearest-neighbour hopping) and for r–$\mathcal{E}$ hopping (variable-range hopping). The main results may be summarized as follows. For r-hopping transport it was found by Böttger and Wegener (BW) (1984) that the differential conductivity σ decreases with increasing applied external field E even at relatively low fields, and for sufficiently high fields the conductivity σ increases rapidly, so that a local minimum in the differential conductivity exists (for experimental results see Redfield (1975), Zabrodskii and Shlimiak (1977) and Bryksin *et al* (1980)). The phenomenon has been predicted theoretically by Böttger and Bryksin (1980) in the framework of the effective-medium approximation (cf also Böttger and Bryksin (1985)) and verified by the 'exact' numerical calculation performed for a sufficiently representative set of r-hopping centres (BW). For r–$\mathcal{E}$ hopping, Böttger *et al* (1985, 1986) observe that the local maximum occurring just after the ohmic region is followed by a local minimum, the latter being followed by an exponential conductivity increase for still higher fields. On increasing the width of the energy distribution of the centres (i.e. lowering the temperature), the height of the maximum increases, whereas the local minimum completely disappears for a sufficiently low temperature.

All the results summarized above have been obtained for a random but macroscopically homogeneous spatial distribution of hopping centres. However, in most real cases an extremely thin layer may hardly be thought of as macroscopically homogeneous. Despite the relaxation of the density of structural defects due to the differences in bond lengths, a macroscopically inhomogeneous average density of the centres may also arise from diffusion of atoms from the substrate and chemical reactions. All these reasons make it necessary to consider the case of random distributions of hopping centres, with their macroscopic density varying as a function of the distance from the injecting contact. Recently, such a spatial inhomogeneity in the distribution of centres has been proved to influence markedly the transient currents measured in classical time-of-flight experiments for hopping transport (Rybicki *et al* 1991a, 1992), as well as for multiple-trapping transport for the isothermal case (Rybicki and Chybicki 1988, 1989, 1990, Rybicki *et al* 1990a) and for the non-isothermal case (Tomaszewicz *et al* 1990, Rybicki *et al* 1991b). Thus, it seems interesting to investigate also the influence of the layer inhomogeneity on stationary current–field and conductivity–field characteristics. Preliminary results have been briefly reported by Rybicki *et al* (1990b, 1991c) and Mancini *et al* (1991). In the present paper we describe in detail the results of the numerical simulation of the above-mentioned current–field and conductivity–field characteristics for r-hopping and r–$\mathcal{E}$-hopping transport of small polarons in disordered systems, i.e. in the approximation of strong electron–phonon coupling (sections 3.1 and 3.2, respectively). In our model calculations we use the average density of hopping centres which exponentially decreases in space, and which for the case of r–$\mathcal{E}$-hopping transport has a Gaussian distribution in energy. However, before presenting the results, in the following section we briefly recall the basic equations describing hopping conductivity.

2. Basic equations

The basic equations describing hopping conductivity are here formulated in the form presented by Böttger and Bryksin (1985). For an electric field E of arbitrary strength, the density j of the DC hopping current is given by

$$j = \frac{1}{2\Omega} \sum_{m,m'}^{N} (R_m - R_{m'}) i(m', m) \tag{1}$$

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

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

where $V_{m'm} = V_{m'} - V_m$, $V_m = \mathcal{E}_m + eu_m$, $\mathcal{E}_m$ is the energy of the mth site, u_m is the potential of the external field E at the point $\boldsymbol{R}_m$, e is the elementary charge, $\beta = kT$, k is the Boltzmann constant, T is the temperature and $W_{m'm}$ is the symmetrized hopping probability. The latter, although in general given by an extremely complicated expression, in the limit of a strong electron–phonon coupling becomes simply

$$W_{m'm} = W_0 \exp[-2\alpha|\boldsymbol{R}_{m'm}|] \tag{3}$$

where α is the reciprocal Bohr radius, and the pre-factor W_0 depends weakly on the external electric field E, as well as on the site position $\boldsymbol{R}_m$ and energy $\mathcal{E}_m$. In the present paper we deal only with the case of strong coupling, leaving the limiting case of weak electron–phonon interaction to be discussed in subsequent work.

Our numerical results have been obtained by solving equations (1)–(3), together with the condition

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

The algorithm that we used follows the general guidelines describes by BW.

3. Numerical results and discussion

In this section we shall discuss in turn the cases of r-hopping transport (section 3.1) and r–$\mathcal{E}$-hopping transport (section 3.2), both for a strong electron–phonon interaction. Before going on to the results, we give here the values of the parameters, which are common to r-hopping and r–$\mathcal{E}$-hopping simulations. The concentration n of electrons in the system was chosen to be 0.5 and was assumed not to depend on the applied electric field. This means that the current–electric field characteristics were calculated under the normalization condition

$$N^{-1}\sum_m \rho_m = n = 0.5. \tag{5}$$

The qualitative behaviour of the calculated characteristics does not depend on the value of n, and the specific value $n = 0.5$ was chosen to maximize the current values. Following the previous work on the subject by BW and Böttger *et al* (1985, 1986), we deal with relatively diluted systems with $\alpha\mathcal{N}^{-1/3} = \frac{1}{15}(\mathcal{N} = N/\Omega)$.

As mentioned in the introduction, for model simulations we used an exponential spatial dependence of the average density $N_h(x)$ of hopping centres, where x is the distance measured from one of the electrodes, given by $0 < x < L$ (L is the layer thickness). In particular we assume that

$$N_h(x) = N_0 S(x) \tag{6}$$

where $S(x)$ is the distribution shape function; in our case

$$S(x) = \exp(-x/D) \tag{7}$$

where D is a characteristic length of the site concentration decay. The ratio L/D may be referred to as the degree of inhomogeneity of the distribution. In what follows we consider L/D to be in the range from 0.0 to 3.0. The results obviously do not depend on the layer polarization, i.e. the characteristics are invariant with respect to the transformation $S(x) \rightarrow S(L-x)$ (for the same random distribution of centres). As far as the influence of the random initial distribution is concerned, the characteristics for various random generations of the centre distribution lead to differences in numerical values, leaving the curve shape qualitatively unchanged. The quantitative discrepancies are most pronounced for macroscopically uniform distributions and almost do not exist for highly inhomogeneous structures.

As far as the energy distribution of centres is concerned, in the case of r–$\mathcal{E}$-hopping transport we shall consider a normal Gaussian distribution, with standard deviations in the range from $3kT$ to $16kT$.

3.1. r-hopping transport

For r-hopping transport we have performed calculations for cubes containing 100 and 500 centres. We present here the results obtained for 500-point simulations, which are of course more reliable. We would like to stress that the results for 100 points are qualitatively identical with those for 500 points for non-uniform structures ($L/D > 0.5$). In the case of a uniform ($L/D = 0.0$) or nearly uniform ($L/D = 0.5$) centre distribution, however, the results are extremely sensitive to the number of points in the simulation box and to their random distribution.

Figure 1 shows current–electric field characteristics for various degrees L/D of inhomogeneity in the range 0.0–3.0. The characteristics obtained for non-uniform centre distributions, with L/D in the range 1.0–3.0, reveal an N-like shape, i.e. they show a current maximum at fairly low fields, followed by current decay down to a minimum value, and by a subsequent exponential current increase. Such a behaviour is the same as obtained by BW for a uniform centre distribution. However, our calculations for a uniform average centre density ($L/D = 0$) do not coincide with the calculations of BW. Firstly, we could calculate only up to $E' = 0.4$, where $E' = \frac{1}{2}eE\beta\alpha^{-1}$ (for higher values of E', negative centre occupation probabilities were found). Secondly, in the field range that we scanned, our j versus E' characteristic increases monotonically, whereas BW observed an N-shaped curve, with the current maximum at about $E' = 0.2$, and the current minimum at about $E' = 0.4$ (figure 2 in the paper of BW; note the different arguments on the horizontal axis in their work and in the present work). Thus, in contrast with BW, we do not observe an N shape of the j versus E' characteristic for the uniform centre distribution. This difference could probably be attributed to the different numbers of points in the simulation box. Simulations for 100 points give neither a current minimum nor a differential conductivity minimum; for 500 points, as shown here (curves a in figures 1 and 2), there is no current minimum, but there is a distinct minimum in the differential conductivity; finally, the 1000-point simulation of BW permitted them to observe both a current minimum and a differential conductivity minimum. In view of such a size dependence we shall assume that the result of BW for the uniform centre distribution is more reliable than ours. The extremely strong size dependence of the characteristics in the case of the uniform distribution remains in contrast with the case of highly non-uniform centre distributions ($L/D \geqslant 1.0$), where the results reveal only

marginal size effects. Even a small degree of spatial inhomogeneity (cf curve b in figure 1, showing an intermediate behaviour between $L/D = 0$ and $L/D > 1$ curves) reduces markedly the size effects. Thus, we claim our results to be fully reliable for non-uniform systems ($L/D \geqslant 1.0$), and in what follows we shall concentrate on the influence of the spatial non-uniformity in the centre distribution on the sample conductivity.

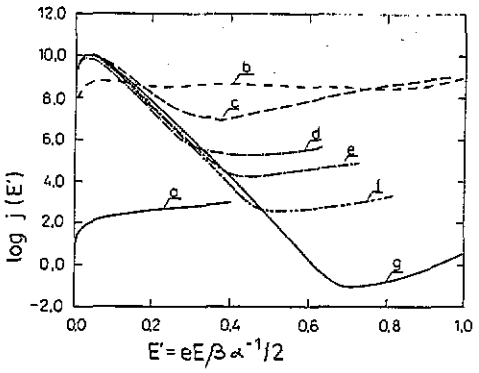

Figure 1. Dependence of the current–field characteristics on the inhomogeneity parameter L/D for r-hopping transport (system parameters: number N of centres, 500; average centres occupation $n = 0.5$, dilution $\alpha N^{-1/3} = 1/15$): curve a, $L/D = 0.0$ (macroscopically homogeneous centre distribution); 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, $L/D = 2.5$; curve g, $L/D = 3.0$.

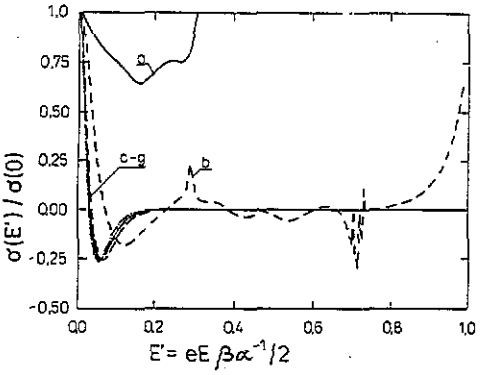

Figure 2. Differential conductivities $\sigma(E')/\sigma(E' = 0)$ calculated from characteristics a–g in figure 1.

Curves c–g in figure 1, calculated for systems with a marked degree of spatial non-uniformity in the r-hopping centre distribution have an N-like shape, as found by BW for the case of a uniform centre distribution. The quantities which depend markedly on L/D are the position and depth of the current minima. With increasing sample non-uniformity the current minima occur for systematically increasing fields, and the minimal current values become lower.

Figure 2 shows differential conductivities $\sigma(E')$ corresponding to the j versus E' characteristics of figure 1 (normalized to $\sigma(E' = 0)$). As is seen, the curves corresponding to $L/D \geqslant 1.0$ almost coincide, and thus the relative differential conductivity variations in the function of the applied field do not depend on the degree of spatial inhomogeneity in the centre distribution, assuming a specific shape common to sufficiently non-uniform systems. The point to be noted is that conductivity saturation occurs at a certain critical value E'_s of the applied field, which is only weakly dependent on L/D (for our 500-point simulations, $E'_s \simeq 0.2$; this value increases with increasing number of points in the simulation box, remaining independent of L/D, however). The non-uniform systems thus become ohmic, at least up to fields consistent with the assumption of constant carrier concentration. Such a behaviour is quite different from the case of uniform systems, where there is no conductivity saturation, but the local conductivity minimum is followed by an exponential conductivity increase. In order to understand such behaviour we have studied the histograms of the average occupation probabilities of the centres as a function of E', for values of L/D equal

to 0.0, 0.5, ..., 2.5, 3.0. Figures 3, 4 and 5 show several such histograms (for $L/D = 0.0$, 1.5 and 3.0, respectively) at several values of E'. The average occupation probabilities $P(i)$, $i = 1, \ldots, 10$ were calculated as the arithmetic means over subsequent slices $L/10$ thick. The histograms indicated in figures 3(a), 4(a) and 5(a) show average occupations for extremely low electric field ($E' = 0.005$). $P(i)$ is approximately constant over the layer thickness for the uniform centre distribution (figure 3(a)) and increases with increasing i when the centre average concentration decays with increasing i (figures 4(a) and 5(a)). Nowhere is the average occupation close to unity, and the value $E' = 0.005$ falls in the ohmic region for all values of L/D. Increasing the field leads to marked changes in $P(i)$. In particular, within the sample emerges a region with the average occupation probability close to unity. The field range, within which at least one of the $P(i)$-values effectively approaches the value of unity, corresponds to a subohmic portion of each j versus E' curve, i.e. to the decay of the differential conductivity to zero. For higher fields the extension of the region with $P(i)$ close to unity increases and, for more non-uniform structures, covers almost the whole bulk of the sample. This field range corresponds to the portions of decreasing current with increasing field, i.e. to the region of negative differential conductivity. Finally, the increasing field is again able to enforce effective carrier motion, and the current begins to increase slightly, so that the conductivity assumes small positive values; in this field interval the spatial extension of the $P(i) = 1$ region begins to shrink. The exponential conductivity increase at high fields expected for macroscopically uniform systems (Hill 1971, Mott 1971, Shklovskii 1972, 1976, Austin and Sayer 1974, Apsley and Hughes 1975, Pollak and Riess 1976) is not observed for the non-uniform systems investigated. It seems that the sample inhomogeneity shifts the region of the exponential current increase to field values which could not be consistent with the assumption of constant carrier concentration.

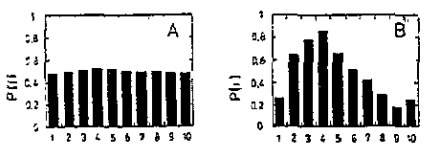

Figure 3. Histograms of the average occupation probability for $L/D = 0.0$ at various electric fields: (a) $E' = 0.005$; (b) $E' = 0.4$.

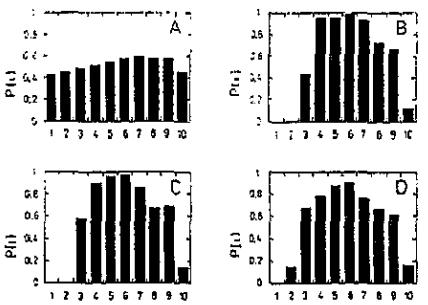

Figure 4. Histograms of the average occupation probability for $L/D = 1.5$ at various electric fields: (a) $E' = 0.005$; (b) $E' = 0.2$; (c) $E' = 0.4$; (d) $E' = 0.6$.

3.2. r–$\mathcal{E}$-hopping transport

The results of simultations in the case of the r–$\mathcal{E}$-hopping transport mechanism show much less pronounced box-size effects than those for the r-hopping mechanism. A number of tests leads us to consider the simulations performed for only 100 r–$\mathcal{E}$ hopping centres as fully representative.

Figures 6 and 7 show the dependence of the j versus E'' characteristics on the energy width of the Gaussian centre distribution, for L/D equal to 3.0 and 0.0, respectively. Here,

the field argument E'' is defined as $E'' = eEN^{-1/3}/(2\ \Delta\mathcal{E})$, where $\Delta\mathcal{E}$ is the threefold standard deviation of the Gaussian distribution. Figures 8 and 9 show the field dependences of the differential conductivity, extracted from the data presented in figures 6 and 7, respectively. For strongly non-uniform samples ($L/D \geqslant 2.0$) the j versus E'' characteristics calculated in rather a wide range of $\Delta\mathcal{E}$ have a well defined maximum (figure 6), in contrast with the uniform systems, where the current monotonically increases with increasing E'' (figure 7). The differential conductivities corresponding to the curves in figures 6 and 7 are shown in figures 8 and 9, respectively (normalized to $\sigma(E'' = 0)$). As is seen, for non-uniform structures the conductivity decreases to its negative minimum value at low fields and then increases asymptotically to a negative value close to zero. The width of the field range, where σ is negative and has an absolute value significantly different from zero, decreases systematically with increasing standard deviation of the energy distribution of the centres (or alternatively with decreasing temperature). On the contrary, uniform systems do not show negative conductivity, but only a shallow local minimum for not too wide an energy distribution (figure 9), in accordance with the results presented by Böttger *et al* (1986). Here again, in a similar way to the case of r hopping, an exponential increase in the conductivity at high fields, characteristic of uniform systems, is shifted to much higher fields of values inconsistent with the assumption of constant carrier concentration. Histograms of average occupation probabilities are presented in figures 10 and 11. For $L/D = 3.0$ and $\sigma_\mathcal{E} = 3kT$, a full-occupation region within the sample emerges at $E'' \simeq 1.0$ (figure 10(b)), which corresponds to the current maximum (figure 6). For higher fields the whole bulk of the sample has almost completely occupied centres (figures 10(c) and 10(d)). Up to the maximum field that we consider, the full-occupation region does not shrink, and no current increase is observed. On the contrary, for a macroscopically uniform spatial distribution of centres (figure 11), a narrow region of completely occupied centres appears for $E'' > 4.0$, which is probably high enough to enforce effectively hops longer than the width of the fully occupied slice, and so no current decrease is observed. Figures 12 and 13 show the currents $j(E'')$ and normalized differential conductivities $\sigma(E'')/\sigma(0)$, respectively, for $L/D = 1.5$. The latter value of the ratio L/D corresponds to the transition between the typical uniform-sample behaviour (figures 7 and 9) and the typical non-uniform-sample behaviour (figures 6 and 8).

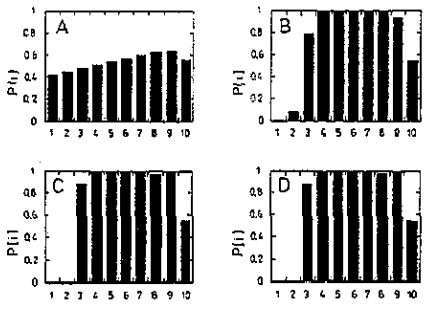

Figure 5. Histograms of the average occupation probability for $L/D = 3.0$ at various electric fields: (a) $E' = 0.005$; (b) $E' = 0.2$; (c) $E' = 0.8$; (d) $E' = 1.0$.

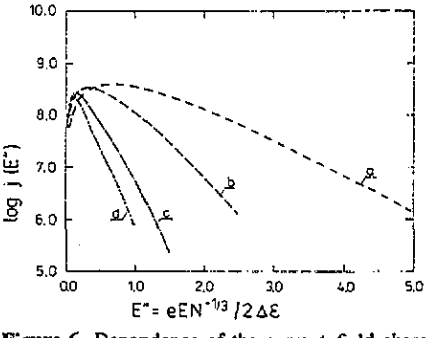

Figure 6. Dependence of the current–field characteristics on the width (standard deviation) $\sigma_\mathcal{E}$ of the Gaussian energy distribution of centres for an inhomogeneity parameter $L/D = 3.0$ (system parameters: number N of centres, 100; average centre occupation $n = 0.5$; dilution $\alpha\mathcal{N}^{-1/3} = \frac{1}{15}$): curve a, $\sigma_\epsilon = 3kT$; curve b, $\sigma_\mathcal{E} = 6kT$; curve c, $\sigma_\mathcal{E} = 12kT$; curve d, $\sigma_\mathcal{E} = 16kT$.

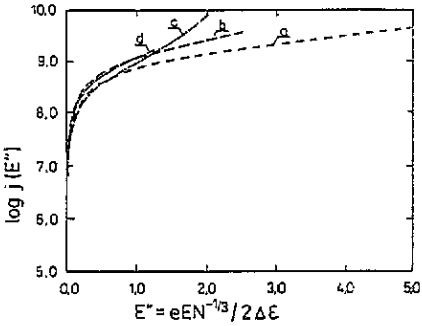

Figure 7. Dependence of the current–field characteristics on the width (standard deviation) σ_ε of the Gaussian energy distribution of centres for a macroscopically uniform centre distribution ($L/D = 0.0$) (system parameters: number N of centres, 100; average centre occupation $n = 0.5$; dilution $\alpha N^{-1/3} = \frac{1}{15}$): curve a, $\sigma_\varepsilon = 3kT$; curve b, $\sigma_\varepsilon = 6kT$; curve c, $\sigma_\varepsilon = 12kT$; curve d, $\sigma_\varepsilon = 16kT$.

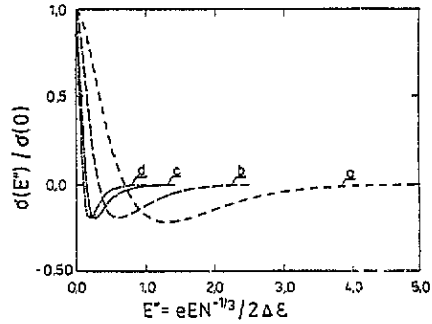

Figure 8. Differential conductivities $\sigma(E'')/\sigma(E'' = 0)$ calculated from characteristics a–d in figure 6.

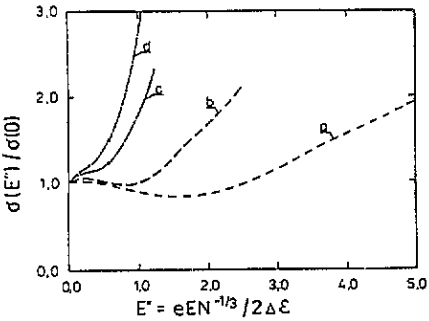

Figure 9. Differential conductivities $\sigma(E'')/\sigma(0)$ calculated from characteristics a–d in figure 7.

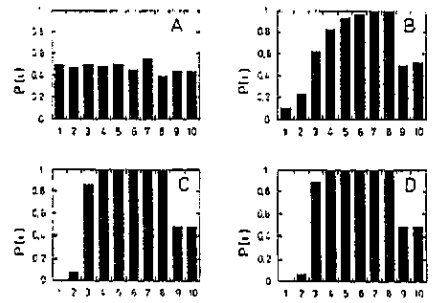

Figure 10. Histograms of the average occupation probability for $L/D = 3.0$ at various electric fields (width σ_ε of the Gaussian distribution, $3kT$): (*a*) $E'' = 0.0$; (*b*) $E'' = 1.0$; (*c*) $E'' = 4.0$; (*d*) $E'' = 5.0$.

4. Concluding remarks

The model of hopping transport described in section 2 is rather difficult to treat numerically. In order to obtain fully realistic results one should perform calculation involving—as we estimate—at least 10^4 hopping centres in the simulation box. For such a number of centres, one could claim to obtain results free of box-size effects. However, increasing markedly the number of sites in the simulation box not only would increase strongly the difficulty of operating numerical methods but also would demand extremely long CPU times; therefore, such calculations are in practice impossible. 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 both significant quantitative and significant qualitative changes due to quasicontinuous variations in the average centre concentration over the sample thickness.

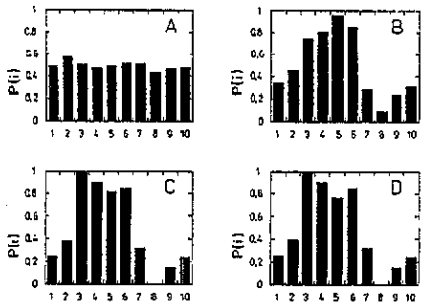

Figure 11. Histograms of the average occupation probability for $L/D = 0.0$ (uniform spatial centres distribution) at various electric fields (width σ_ε of the Gaussian distribution, $3kT$): (*a*) $E'' = 0.0$; (*b*) $E'' = 1.0$; (*c*) $E'' = 4.0$; (*d*) $E'' = 5.0$.

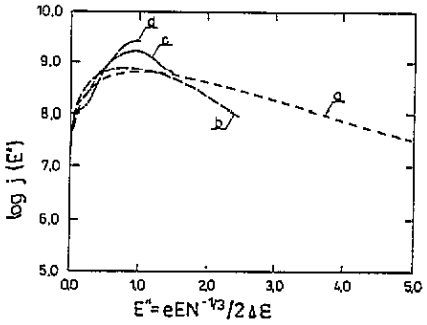

Figure 12. Dependence of the current–field characteristics on the width (standard deviation) σ_ε of the Gaussian energy distribution of centres for an inhomogeneity parameter $L/D = 1.5$ (system parameters: number N of centres, 100; average centre occupation $n = 0.5$; dilution $\alpha N^{-1/3} = \frac{1}{15}$); curve a, $\sigma_\varepsilon = 3kT$; curve b, $\sigma_\varepsilon = 6kT$; curve c, $\sigma_\varepsilon = 12kT$; curve d, $\sigma_\varepsilon = 16kT$.

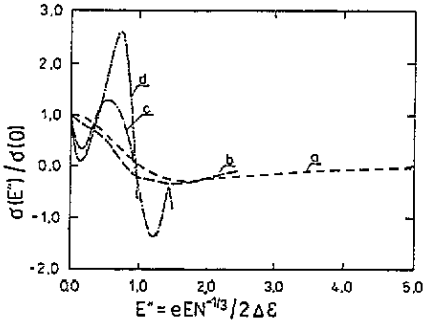

Figure 13. Differential conductivities $\sigma(E'')/\sigma(0)$ calculated from characteristics a–d in figure 12.

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