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r-hopping transient currents in thin dielectric layers with macroscopically non-homogeneous spatial distribution of hopping centres

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Abstract. The present work is a contribution to the understanding of transient currents, which are usually discussed in the context of the classical time-of-flight (TOF) experiment. In particular, *r*-hopping pulse injection transient currents in dielectric layers with a macroscopically non-homogeneous distribution of hopping centres have been investigated with the aid of the Monte Carlo simulation, according to a new algorithm. Both the shape of the transient currents and the value of the effective TOF are found to depend strongly on the degree of macroscopic variations in the hopping-centre concentration over the layer thickness. For a smooth change in space centre concentration, the transient currents may be described by simple analytical expressions, suitable for the estimation of the spatial distribution of hopping centres from experimental data.

1. Introduction

One of the most powerful methods for investigation of the transport properties of thin dielectric layers is the classical time-of-flight (TOF) experiment (Scher and Montroll 1975, Arkhipov and Rudenko 1982, Rudenko and Arkhipov 1982a,b, Kao and Hwang 1981, Marshall 1983a, Weissmüller 1985). The well developed theoretical description, referring mainly to multiple-trapping transport, makes it possible to determine microscopic parameters, such as band mobility, trap concentration and its energetic distribution and trapping cross section. However, until recently, theoretical papers on the TOF experiment dealt only with the case of a spatially uniform average trap density. In real thin dielectric layers, at least in the near-surface region, the spatial distribution of traps may be macroscopically non-homogeneous, owing to the layer preparation, diffusion of atoms from contacts or ambient atmosphere, chemical reactions, etc (see, e.g., Kao and Hwang (1981, p 150), Samoć and Zboiński (1978)). For the multiple-trapping transport mechanism, the influence of the spatial non-homogeneity in the trap distribution on the transient currents measured in the constant-temperature TOF experiment has been investigated by Rybicki and Chybicki (1988, 1989a, 1990), Rybicki *et al* (1990a,b), and on the currents measured in the thermally stimulated TOF experiment by Tomaszewicz *et al* (1990) and Rybicki *et al*

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(1991). The present work is the first step towards understanding the influence of the layer spatial non-homogeneity on hopping-transport transient currents measured in the isothermal TOF experiment.

A theoretical description of the hopping-transport transient currents is rather difficult, and thus much attention has been paid to numerical studies (Böttger and Bryksin 1985). As far as the Monte Carlo simulation of the TOF experiment for r-hopping (nearest-neighbour hopping) transport is concerned, several studies have been published (Bässler 1981, Marshall 1978, 1981, 1983b, Adler and Silver 1982, Ries and Bässler 1987). None of these papers deals with the experimentally important case of spatially non-homogeneous density of hopping centres. In the present paper we study to some extent small-signal pulse injection transient r-hopping currents in dielectric layers with a spatially non-homogeneous density $N(x)$ of hopping centres. More preciesely, we deal with transient currents measured in the classical TOF experiment, for thin dielectric layers revealing an r-hopping mechanism of transport. Such a mechanism is found in disordered hopping systems of relatively narrow energetic distribution of hopping centres, especially at elevated temperatures (Böttger and Bryksin 1985, p 15). In the illustrative part of the paper (section 3), we present and discuss some examples of Monte Carlo results, showing a very strong dependence of both the shape of the transient currents and the value of the effective TOF on the degree of macroscopic variations in the hopping-centre concentration over the layer thickness. Because our algorithm for simulation of transient currents differs from those previously used (cf Marshall (1978) and Bässler (1981)), we describe it in some detail in section 2. Further, some simple analytical expressions describing approximately the simulation results are obtained (section 4). The potential usefulness of the presented simple analytical approach for estimating the spatial distribution of r-hopping centres from experimental data is underlined, which is the second aim of the present paper. Section 5 contains concluding remarks.

2. Simulation algorithm

The Monte Carlo simulation of transient currents corresponding to the isothermal TOF experiment for r-hopping transport was performed with the aid of two different algorithms. The older algorithm was developed by Marshall (1978), and the more recent algorithm by Bässler (1981) (cf Schönherr *et al* (1981)). The main difference between them lies in the manner of choosing the hop, which is assumed to be realized in nature; both workers calculate random hopping times to every neighbouring site, from which Marshall accepts the shortest as that realized in nature, whereas Bässler (working with hopping probabilities rather than with hopping times) accepts the most probable hop. In both algorithms, however, prior to performing random walks of carriers, the distribution of hopping centres is generated at random in a certain box and is kept fixed, at least for a given number of individual carrier walks. From a technical point of view the full information on the generated centre distribution is stored in a RAM during the program execution. Both Marshall and Bässler claim their results to be independent of the simulation box dimensions (rectangular prisms containing some 10^3 and 10^5 sites, respectively). In our case of a spatially macroscopically non-homogeneous centre distribution, however, in order to obtain results free of uncontrolled box-size effects, we would have to use the simulation boxes containing many more centres than in the above-mentioned studies. Firstly, the finite size of the simulation box leads to a systematic error because, conserving the rectangular shape of a simulation box representing the sample, the ratio of the number of near-surface centres (with an incomplete environment) to the number of bulk centres (with a complete environment) changes along the box length. Thus, the systematic error due to finite dimensions of the simulation box depends on the degree of spatial changes in the average hopping-centre density, making it difficult to recognize pure effects due to spatial non-homogeneity. Secondly, any reasonable representation of the random centre distribution with macroscopically smoothly varying average density requires rather a large number of centres within the simulation box. The numerical tests that we performed show that the box-size-independent results would be obtained for numbers of sites, the information for which can hardly be kept in the usually accessible RAM, and the usage of the disc memory makes the computer (VAX) run times unacceptable. Because of this we have developed a new algorithm (Rybicki and Chybicki 1989b), which is completely independent of size effects in directions perpendicular to the field direction and thus is particularly suitable for simulations of transport properties of thin layers in a direction perpendicular to the layer surface.

The individual hop in our algorithm is performed as follows. Let the carrier be localized at some time t at a distance x from the injecting contact (located in $x = 0$). The local homogeneous-density environment is generated in the form of a sphere centred at x, containing a given number n of neighbours. If the hopping-centre concentration is denoted by $N(x)$, the radius $R(x)$ of the sphere is given by $4\pi N(x)R^3(x)/3 = n + 1$. For each hop the new positions of n neighbours are generated at random within the local environment of radius $R(x)$. In particular, we choose their random spherical coordinates r_i and θ_i according to

$$r_i = R(x)X^{1/3} \tag{1}$$

$$\cos\theta_i = 2Y - 1 \qquad i = 1, \ldots, n$$

where X and Y are random numbers from the interval (0, 1). The third coordinate φ_i need not be specified. The hopping times t_i are generated according to (Marshall 1978)

$$t_i = -\log Z \exp(-2\alpha r_i - w r_i \cos\theta_i) \tag{2}$$

where Z is the random number from the interval (0, 1), α is the reciprocal Bohr radius, $w = qE/kT$ (q is the elementrary charge, E the external electric field, k the Boltzmann constant and T the sample temperature), and $r_i \cos\theta_i = x_i$ is the distance between two successive centres along the external electric field E. It is further assumed, following Marshall (1978), that the hop corresponding to $t_{\min} = \min_{(i)} t_i$, $i = 1, \ldots, n$ is realized. After the minimum-time hop is performed, the new position of the carrier becomes $x + \Delta x_{\min} = x + r_{\min}\cos\theta_{\min}$, where the subscript min corresponds to $t_{\min}$. The new position is being assumed at the moment $t + t_{\min}$. Such a procedure is repeated until the carrier reaches $x = L$, where L is the layer thickness. For initial hops, $x < 0$ may occur. In such a case, x is set to 0. The temporal and spatial evolution $n(x,t)$ of the injected carriers has been obtained by averaging over 10^4 random walks of individual carriers started at $t = 0$ and $x = 0$. The transient currents induced in the external circuit have been calculated on the basis of the well known formula (see, e.g., Leal Ferreira (1977))

$$j(t) = -\frac{1}{n_0}\frac{\mathrm{d}}{\mathrm{d}t}\int_0^L n(x,t)\,\mathrm{d}x + \frac{1}{n_0 L}\frac{\mathrm{d}}{\mathrm{d}t}\int_0^L x n(x,t)\,\mathrm{d}x \tag{3}$$

where n_0 is the number of injected carriers. Thus equation (3) gives the particle current per carrier.

As mentioned before, in contrast with the previously used algorithms, the algorithm described above avoids the surface effects on the box boundaries parallel to the external field. The generation of a new local random environment of each actual position eliminates both repeated easy jumps between two close centres and hard jumps between clusters, which were present for example in the Marshall algorithm. In order to gain some insight into the behaviours of the different algorithms, we have calculated some transients for the same parameters with the aid of our new algorithm, and Marshall's algorithm (Bässler deals with compositional disorder, whereas we, in a similar way to Marshall, consider purely positional disorder). It turned out that our algorithm gives current values somewhat higher than Marshall's (by a factor of 1.2–1.4), leaving all the qualitative features essentially unchanged (figure 1). Thus, the difference between the algorithm presented here and Marshall's algorithm leads to slightly different effective velocities of the carriers along the external electric field, the effective velocity being slightly larger in our algorithm.

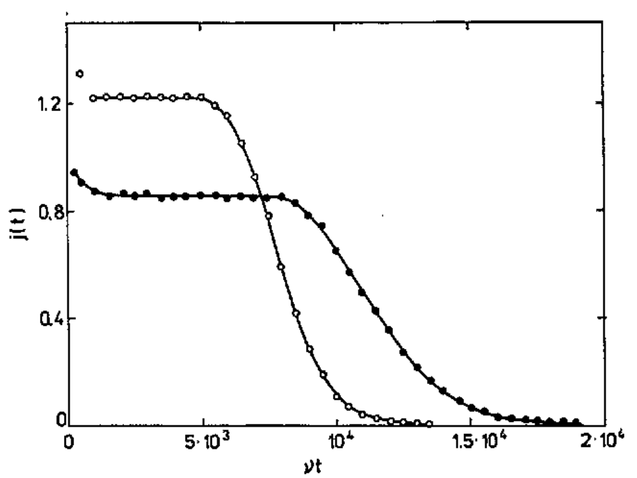

Figure 1. Comparison of transient currents calculated with the aid of the Marshall (1978) algorithm (●) and the algorithm described in the present work (○) for the same parameters ($\alpha' = 3$; $L = 50N_0^{-1/3}$; $\nu = 1\ \mathrm{s}^{-1}$).

3. Simulation results

The simulation has been performed for $\alpha' = 3, 5, 8$, where α is the reciprocal Bohr radius expressed as $N_0^{-1/3}/r_\mathrm{B}$, where r_B is the Bohr radius of the localized state and N_0 is the maximum centre concentration. In all cases, N_0 was equal to unity; thus the length is measured in units equal to the side length of a cube containing on average one hopping centre in the region of their maximum concentration. The spatial variations in the average density $N(x)$ of hopping centres were obtained by introducing the shape function $S(x) : N(x) = N_0S(x)$, $0 < x < L$. For illustrative calculations showing the influence of various spatial centre distributions on transient currents, the shape function $S(x)$ has been chosen to be exponential with a maximum centre density at one or both of the contacts, and a Gaussian spatial centre distribution with a maximum centre density in the middle of the layer thickness (cf the distributions discussed by Kao and Hwang (1981, ch 3)):

$$S(x) = \exp(-x/D) \tag{4}$$

$$S(x) = \exp[-(L-x)/D] \tag{5}$$

$$S(x) = \exp(-x/D_1) + \exp[-(L-x)/D_2] \tag{6}$$

$$S(x) = \exp[-(L/2-x)^2/D^2] \tag{7}$$

D, D_1 and D_2 being the non-homogeneity parameters. The layer thickness was $L = 150 N_0^{-1/3}$. The ratio L/D may be referred to as the non-homogeneity degree of the spatial r-hopping-centre distribution.

The currents obtained are shown in figures 2–4 and later also in figures 6 and 7 for α' and L/D specified in the captions. As easily seen, the influence of the spatial non-homogeneity of centre distribution on transient currents is very distinct (note the log–log scale). The influence of the spatial non-homogeneity on the effective TOF is more pronounced for larger α'. However, a non-dispersive character of the currents is conserved (the existence of a sharp current decay after the effective TOF) (Marshall 1978). For a hopping-centre density (4) exponentially decreasing in x, the carriers with increasing x enter the regions of lower centre concentration, which results in exponentially longer hopping times, and thus in a strong decrease in the current (reversely for the centre distribution (5)). Examples of spatial distributions of the density of propagating charges at several chosen moments of time are shown in figure 5. The carrier packets shown as full circles, open circles, crosses and open squares correspond to the same time and different spatial distributions of hopping centres; full circles and open circles correspond to the homogeneous distribution N_0 = constant, and crosses and open squares to the centre distributions (4) and (5), respectively. The decrease in the current in the regions of decreased centre density remains in opposition to the case of multiple-trapping transport (Rybicki and Chybicki 1988, 1989a, Rybicki *et al* 1990a,b). In the latter case the trap concentration decreasing (increasing) in x results in the increase (decrease) in the current. Despite this obvious qualitative difference, r-hopping transient currents are much more sensitive to the non-homogeneity parameter L/D than are multiple-trapping currents. The transient currents obtained for the centre distributions (6) and (7) are presented in figures 6 and 7, respectively. In these figures we have also shown the influence of the local environment size taken for simulation. The curves through open circles have been calculated for $n = 36$ neighbours, whereas the curves through full circles (in a similar way to figures 2–4) for $n = 8$ neighbours. For $n > 36$ the effect of the local environment size becomes almost saturated.

After this rather illustrative section we now pass to a very simple analytical approach to r-hopping transient currents, giving the experimentalist a potential tool for a rough estimation of the r-hopping-centre distribution over the layer thickness on the basis of TOF data.

4. Analytical considerations

The effective velocity v_{eff} that the carriers assume under the action of the external electric field (related in a straightforward way to the current induced in the external

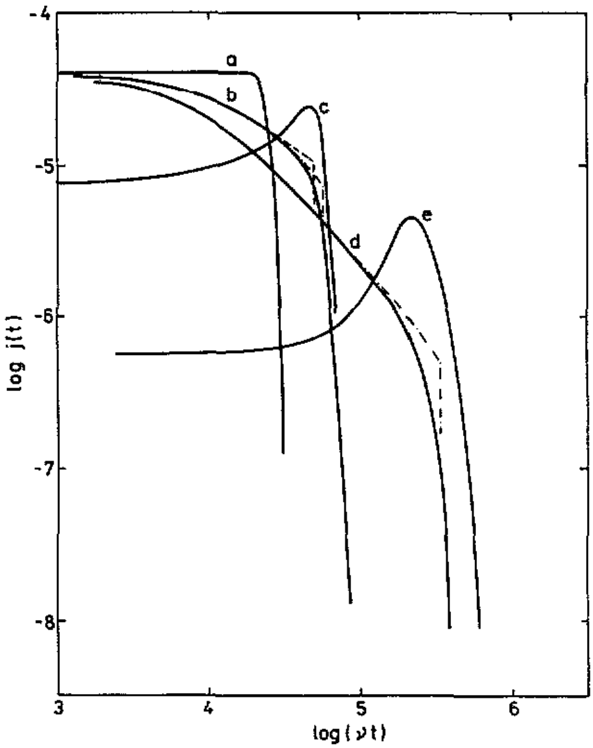

Figure 2. r-hopping transient currents for $\alpha' = 3.0$ and exponential spatial centre distributions (4) and (5): curve (a), $L/D = 0$ (homogeneous hopping-centre distribution); curve (b), distribution (4), $L/D = 1.0$; curve (c), distribution (5), $L/D = 1.0$; curve (d), distribution (4), $L/D = 2.0$; curve (e), distribution (5), $L/D = 2.0$; —, Monte Carlo simulation; – – –, currents calculated with the aid of numerical solution of (11) and (12); –·–·–, with the aid of the approximate equation (15).

circuit) depends in non-homogeneous layers on the local centre density and may be written as

$$v_{\rm eff}(x) = \mu_{\rm eff}(x)E \qquad x \leqslant L \tag{8}$$

where $\mu_{\rm eff}(x)$ is the x-dependent effective mobility of the propagating carrier packet and $x(t)$ is the actual position of the centroid of the hopping carrier packet. $\mu_{\rm eff}(x)$ may be written as (Marshall 1981)

$$\mu_{\rm eff}(x) = (q\nu/6kT)\rho^2(x)\exp\left[-2\alpha\rho(x)\right] \tag{9}$$

where $\rho(x)$ is the average distance between r-hopping centres given by

$$\rho(x) = [N(x)]^{-1/3} \tag{10}$$

and ν is the frequency factor. Thus we have

$$\mu_{\rm eff}(x) = A_0[N_0S(x)]^{-2/3}\exp\{-2\alpha[N_0S(x)]^{-1/3}\} \tag{11}$$

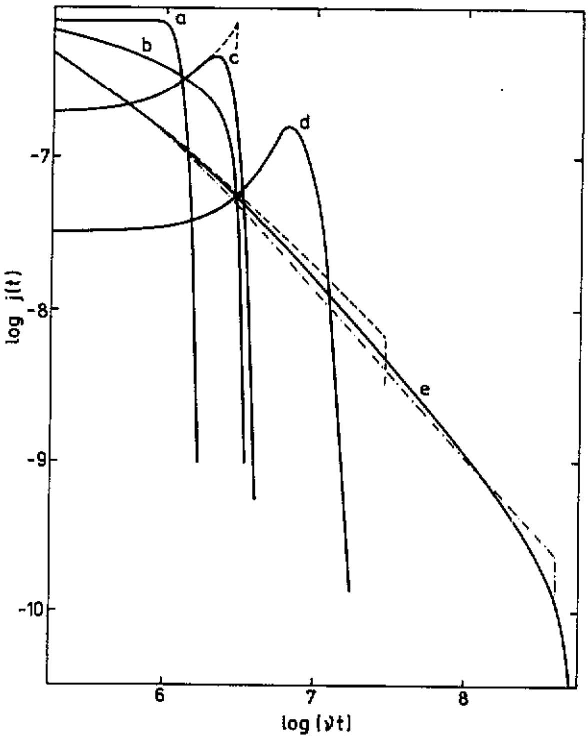

Figure 3. r-hopping transient currents for $\alpha' = 5.0$ and exponential spatial centre distributions (4) and (5): curve (a), $L/D = 0$ (homogeneous distribution); curve (b), distribution (4), $L/D = 0.5$; curve (c), distribution (5), $L/D = 0.5$; curve (d), distribution (5), $L/D = 1.0$; curve (e), distribution (4), $L/D = 2.0$. The meaning of the curves is as in figure 2.

with $A_0 = q\nu/6kT$. Clearly, the actual position of the carrier packet centroid $x(t)$ is

$$x(t) = E\int_0^t \mu_{\rm eff}(x(\tau))\,\mathrm{d}\tau \tag{12}$$

and thus equations (11) and (12) constitute an integral equation for $x(t)$, which cannot be solved without $S(x)$ defined *a priori*. However, for a given shape function $S(x)$ the inegral equations (11) and (12) may be solved at least approximately. For example for $S(x)$ given by (4) we have

$$\overline{\mu}_{\rm eff}(t) = A_0 N_0^{-2/3} \exp\left(\frac{2}{3D}\int_0^t E\overline{\mu}_{\rm eff}(\tau)\,\mathrm{d}\tau\right) \times \exp\left[-2\alpha N_0^{-1/3}\exp\left(\frac{1}{3D}\int_0^t E\overline{\mu}_{\rm eff}(\tau)\,\mathrm{d}\tau\right)\right] \tag{13}$$

where $\overline{\mu}_{\rm eff}(t) \equiv \mu_{\rm eff}(x(t))$. For $L < D$ the exponent in the second exponential factor may be linearized, and thus

$$\overline{\mu}_{\rm eff}(t) = \mu_0 \exp\left(-\frac{2(\alpha'-1)}{3D}\int_0^t E\mu_{\rm eff}(\tau)\,\mathrm{d}\tau\right) \tag{14}$$

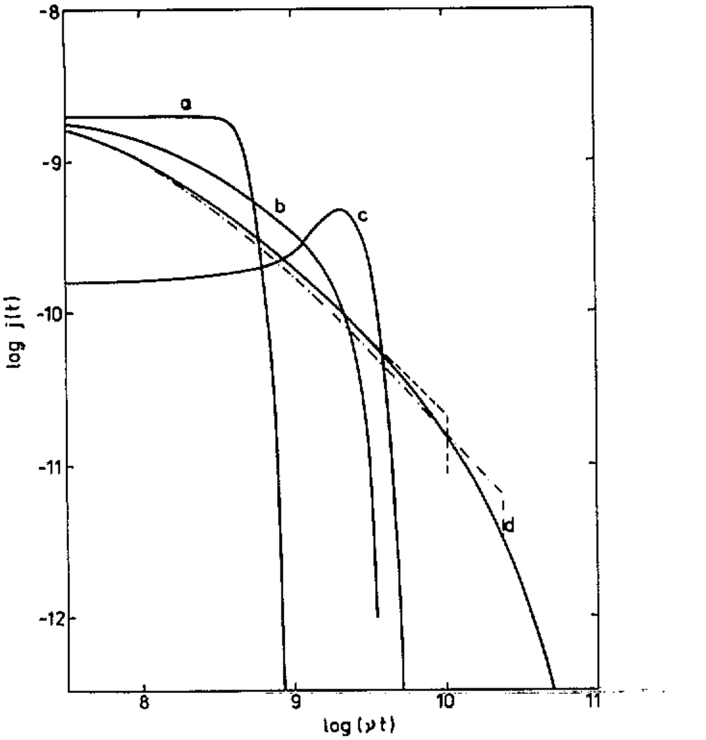

Figure 4. r-hopping transient currents for $\alpha' = 8.0$ and exponential spatial centre distributions (4) and (5): curve (a), $L/D = 0$ (homogeneous distribution); curve (b), distribution (4), $L/D = 0.5$; curve (c), distribution (5), $L/D = 0.5$; curve (d), distribution (4), $L/D = 1.0$. The meaning of the curves is as in figure 2.

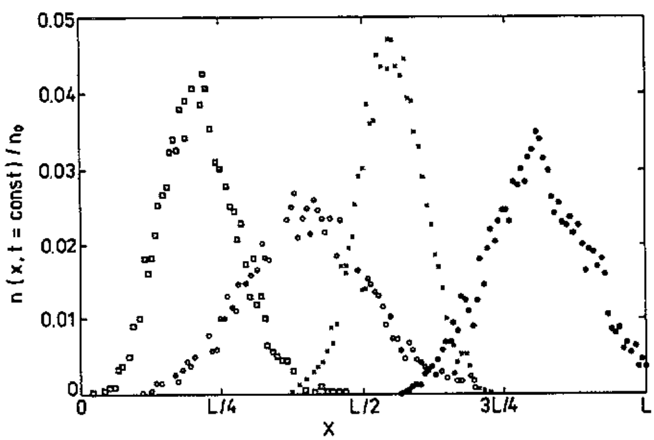

Figure 5. Spatial distributions of the carrier packets: ●, $\alpha' = 3$, $\nu t = 2 \times 10^4$, homogeneous centre distribution; ×, $\alpha' = 3$, $\nu t = 2 \times 10^4$, distribution (4), $L/D = 1$; □, $\alpha' = 3$, $\nu t = 2 \times 10^4$, distribution (5), $L/D = 1$; ○, $\alpha' = 8$, $\nu t = 2 \times 10^8$, homogeneous centre distribution. Note the increasing packet dispersion with increasing α'.

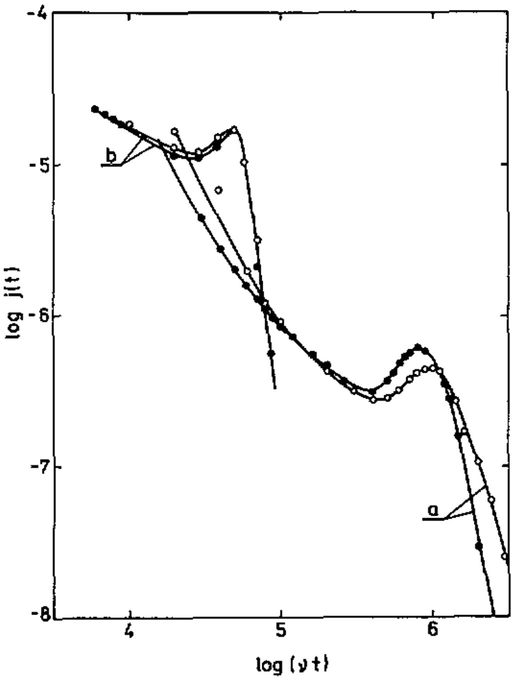

Figure 6. Monte Carlo r-hopping transient currents for $\alpha' = 3.0$ and double-exponential spatial centre distribution (6): curve (a), $L/D_1 = L/D_2 = 5$; curve (b), $L/D_1 = L/D_2 = 3.0$; ●, $n = 8$ neighbours in each local environment; ○, $n = 36$ neighbours in each local environment.

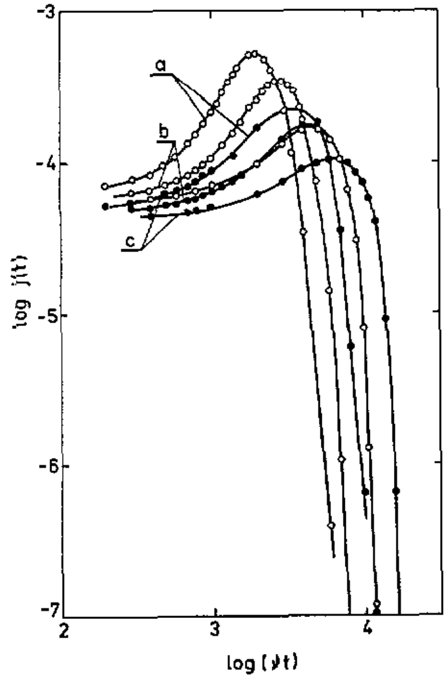

Figure 7. Monte Carlo r-hopping transient currents for $\alpha' = 3.0$ and Gaussian spatial centre distribution (7): curve (a), $L^2/4D^2 = 3.0$; curve (b), $L^2/4D^2 = 2.0$; curve (c), $L^2/4D^2 = 1.0$; ●, $n = 8$ neighbours in each local environment; ○, $n = 36$ neighbours in each local environment.

where $\mu_0 = A_0 N_0^{-2/3} \exp(-2\alpha')$. The logarithm of both sides of equation (14) is reduced, on differentiation with respect to t, to a simple separated ordinary differential equation, which is solved immediately as

$$\mu_{\mathrm{eff}}(t) = \mu_0 / \{[2(\alpha' - 1)/3D]\mu_0 E t + 1\}. \tag{15}$$

The latter expression, together with (8), yields the electric current $i(t) = e n_0 v_{\mathrm{eff}}(x(t))$. The corresponding formula for the hopping-centre distribution (5) may be obtained in the same way. Transient currents obtained from (8) with μ_{eff} calculated by numerically solving (11) and (12) are shown in figures 2–4 as broken curves, and those based on the approximate solution (15) as chain curves. In both cases the pre-factor μ_0 has been taken from the Monte Carlo simulation. In our simple model the packet spatial dispersion has been neglected, and thus the current falls to zero at the moment $x = L$ is reached. This means that our analytical results are not expected to be precise for times close to the effective TOF.

5. Concluding remarks

In the present work we have shown with the aid of the Monte Carlo simulation the remarkable influence of the spatial non-homogeneity in the r-hopping-centre distribution on transient currents measured in the classical TOF experiment. The r-hopping current–time characteristics turn out to be much more sensitive to the layer non-homogeneity than are the multiple-trapping characteristics. Thus, in any interpretation of the TOF measurements for hopping transport in the framework of a uniform centre spatial distribution, special care must be paid to produce really homogeneous samples or, alternatively, the measurement results must be interpreted with the aid of models taking into account the spatial variations in centre density. Regarding the latter possibility, as is easily seen, our very simple model for r-hopping transients described in section 4 leads for times shorter than the effective TOF to quite good agreement between the analytic formulae and the Monte Carlo results, at least for a slowly varying centre density. This suggests that the model could be used to determine, or at least to estimate, the spatial r-hopping-centre distribution on the basis of experimental TOF data. In order to do this, one should assume $S(x)$ in the form of a family of one-parameter (or more) functions and fit them to get the closest coincidence between the measured and calculated curves.

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