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ELECTRIC TRANSPORT IN SPATIALLY NON-UNIFORM THIN DIELECTRIC LAYERS

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ABSTRACT: In the paper we summarise in short our works on the influence of the spatial non-uniformity of the transport centres density on the transient currents profiles, and on the stationary current-field characteristics of thin dielectric layers. We concentrate ourselves on the numerical results only:
1) Monte Carlo simulations of non-dispersive and dispersive transient currents in the classical small signal monopolar time-of-flight experiment, both for multiple-trapping, and hopping transport mechanisms;
2) Calculations of the high-field non-ohmic current-field, and differential conductivity-field characteristics within the Bottger-Bryksin model of 3D random resistor network, both for small, and large polarons in amorphous systems. The numerical results reveal some interesting features specific to the layer non-uniformity, which are very difficult to predict and/or describe within an analytical approach.

1. INTRODUCTION

Because of their direct applications in microelectronics, thin dielectric layers are of great interest in solid state physics. Electric transport properties are here of the fundamental importance [5, 7]. There are two basic mechanisms of electronic transport in the bulk of non-ideal solids: multiple-trapping, and hopping. The first of them, multiple-trapping mechanism (MT) consists in the electron (or hole) band transport, interrupted repeatedly by the carrier trapping at the material imperfections (localised states, so called traps). Once trapped (temporarily immobilised) carrier is after some random time thermally activated to the band of extended states, where it participates again in the band transport. The hopping mechanism (H) consists in the carrier jumps directly between localised states (tunnelling, or thermally activated tunnelling), without visiting the extended band states. There exists an extensive literature on both transport mechanisms in their applications to thin dielectric layers, placed between two planar contacts. Most of the work that has been done refers to the layers with random in space and in energy distributions of trapping/hopping centres, but the average total (integrated over energy) centres distribution is usually assumed to be uniform over the layer thickness L (independent on the distanced from the injecting contact). The experimental results are usually compared to the theoretical predictions, and in such

a way the material transport parameters are determined or estimated (traps density, their energetic distribution, trapping cross-section, band mobility for MT mechanism, or hopping centres density, their energetic distribution, Bohr radius for H mechanism). However, a real very thin layer (say $L \approx 0.1$ μm or less) can hardly have a spatially uniform distribution of trapping/hopping centres. There is a number of phenomena which can introduce in an uncontrolled way significant spatial variations of the centres total density, and their energetic distribution (diffusion of atoms from contacts or ambient atmosphere, chemical reactions, misfit between inter-atomic distances in the layer and the contacts, etc.). Obviously, any comparison of the experimental data to the theoretical formulas developed for spatially uniform layers fails, giving unrealistic values of the fitted parameters. Thus, it is necessary to understand the influence of spatial non-uniformity in the trapping/hopping centres distribution on the measured quantities. Among several most widely used experimental methods for studying the transport properties of thin layers are: the classical isothermal, and non-isothermal (thermally stimulated) time-of-flight (TOF) experiment, and the direct measurements of the stationary current-field, and conductivity-field characteristics. The present authors have contributed somewhat to the understanding of the above topics, extending certain analytical models on the case of spatially non-uniform layers, and performing extensive numerical simulations of the electron transport in such systems where no straightforward analytical solution exists.

In the present paper we summarise in short some our numerical results referring to transient (Sec. 2) and stationary (Sec. 3) currents in spatially non-uniform thin dielectric layers.

2. TIME-OF-FLIGHT (TOF) EXPERIMENT IN NON-UNIFORM LAYERS

In the isothermal TOF experiment an infinitesimally thin sheet of charge carriers is produced near one of the surfaces of a thin layer placed between two contacts, and subject to an external electric field. The subsequent motion of the carriers towards the collecting contact results in a transient current which is analyzed in an external circuit connected to the sample. The theory of the TOF experiment [2, 28, 29, 15] permits to perform a thorough analysis of the transient currents measured in the macroscopically uniform layers, especially as the MT transport mechanism is concerned. In this section we will consider the influence of spatial variations of the total trapping/hopping centres density on the TOF current profiles for both MT and H transport mechanisms.

Let us now specify the centres distributions under consideration. In the first approximation the general trapping/hopping centres distribution in energy ε (measured down from the extended states edge), and in space (x – the distance from the injecting contact, $0 \le x \le L$), $N(x, \varepsilon)$, may be written in a factorised form as $N(x, \varepsilon) = N_0 S(x; D) f(\varepsilon)$ (or linear combination of such products). $S(x; D)$ is a predefined model one-parameter

(D) family of x-dependent functions, i.e. the shape functions describing the spatial variations of the fraction of centres, which are distributed in energy according to $f(\varepsilon)$. According to the general theory of electronic states in random systems [6] the most natural choices of the f – functions are: an exponential distribution ($\sim\exp(-\varepsilon/kT_c)$, where T_c is the characteristic temperature, k – the Boltzmann constant) in the case of MT mechanism, and the Gaussian distribution of standard deviation σ, in the case of H mechanism. As far as the $S(x, D)$ functions are concerned, we used mainly exponentially decreasing (increasing) in x spatial distributions, $S(x; D) \sim \exp(\pm x/D)$ (and their linear combinations), which correspond to defects of diffusive or radiative origin. The factorised form of $N(x, \varepsilon)$ is sufficiently flexible to cover a rather wide range of possible centres distributions in x and ε in real systems.

We consider a layer of thickness L placed between two planar contacts (at $x = 0$ and $x = L$, respectively), with an x-dependent concentration of trapping/hopping centres. At $t = 0$ a carrier is generated at $x = 0$, and an external electric field E enforces its motion towards the $x = L$ contact. Such individual carriers walks are averaged in order to obtain time evolution of the carrier packet density $n(x, t)$, and the electric current $j(t)$ in the external circuit can be calculated. The field E is held constant and uniform, so that no space charge effects are included.

The carrier packet $n(x, t)$ evolves in different ways for MT, and H mechanisms, and for each of them two limit cases may be distinguished: non-dispersive (Gaussian), and dispersive (non-Gaussian) transport. If the dispersion of times of flight (between $x = 0$, and $x = L$) of individual carriers is low, the initially generated sheet of carriers moves towards the collecting electrode as a Gaussian packet of increasing in time width [31]. As a result, the current measured in the external circuit is approximately constant, and decays rapidly at times close to the moment, at which the carriers packet centroid reaches $x = L$ (non-dispersive transport). On the other hand, if the times of flight of individual carriers differ by many orders of magnitude (due to deep trappings or hard hops), the current measured in the external circuit decays from $t = 0$ on, and the "effective time-of-flight" is related to the change of the current slope in the log-log scale [28]. Let us discuss in turn the cases of MT and H transport mechanisms, both in the dispersive, and non-dispersive limits.

2.1. Multiple-trapping transport mechanism

The theory of the isothermal TOF experiment in spatially uniform layers is due to [31] (non-dispersive transport), and to [28, 29] (dispersive transport). In the former case the transport equations in the form generalised on non-uniform systems can be solved analytically for $n(x, t)$, the latter being expressed by the hyperbolic Bessel function of the fist order of a complicated argument [20, 21]. Thus, the numerical work

in this simplest case of non-dispersive multiple-trapping transport is limited only to the calculation of $j(t)$ (integration of $n(x, t)$ in respect of x, and differentiation in respect of t [8]). In the case of the dispersive transport, however, no exact analytical solutions to the transport equations can be found, and only some approximate formulas can be derived using ad hoc simplifications of unknown accuracy. Here the Monte Carlo (MC) simulation [18] is an excellent way to obtain the current profiles (instead of the numerical solution of the system of partial differential-integral transport equations). The MC algorithm is very simple. One needs to generate at random, according to proper distribution functions, the free drift time in the conduction band before the trapping event, the depth of the encountered trap, and the carrier dwell time in the trap. The carrier moves towards the collecting electrode only when it remains in the conduction band. Averaging over 10^4-10^5 individual carriers random walks (each consisting in 10^3-10^4 trapping/detrapping events) gives almost exactly smooth current profiles. An example is shown in Figure 1 (exponential variations of the trap density in x).

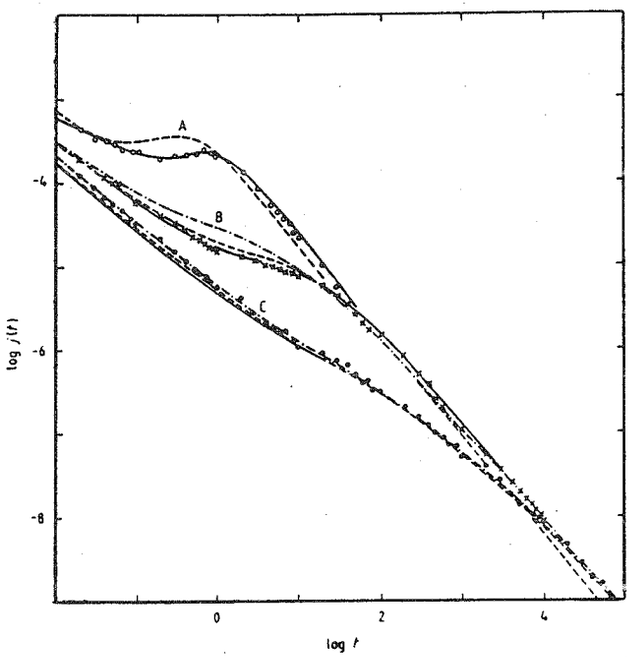

Fig. 1. Dispersive multiple-trapping TOF current profiles: points – MC simulation, lines – various approximate analytical expressions, suitable for a determination of the spatial trap distribution in the basis of the experimental results. $f(\varepsilon) \sim \exp(-\varepsilon/kT_c)$, $S(x;\ D) \sim \exp(-x/D)$, $L/D = 5.0$, dispersion parameter $T/T_c = 0.5$ (A), 0.33 (B), 0.2 (C) [20].

In the MT mechanism the efficient carrier mobility decreases on increasing trap density, and the dispersion degree increases on increasing width of the trap energetic distribution. Thus for monotonously decreasing in space trap concentration the carriers move faster near the collecting electrode, and the characteristic current maximum appears (under certain conditions) immediately before the effective time of flight. On the other hand, for increasing trap density the carriers motion is slowed down near the collecting electrode, leading to the current dumping. The MC results, despite their illustrative character allow also to establish the accuracy and the range of validity of the approximate analytical expressions (Figure 1, [20]). Because of the high flexibility of the MC method it was possible to investigate a wide range of possible energetic and spatial traps distributions in both isothermal [16, 22], and non-isothermal [23, 30] cases (the latter for an arbitrary sample heating scheduling).

2.2. Hopping transport mechanism

The hopping TOF transient currents are more difficult in theoretical treatment, and computer experiments are often performed in order to elucidate certain features of the hopping transport in materials characterised by on-diagonal or/and off-diagonal disorder ([1, 11-14] in each case for spatially uniform layers). The first MC simulations of the hopping TOF transient currents in spatially non-uniform systems have been performed by the present authors, and their co-workers. We report briefly on the results obtained for non-uniformly doped/defected crystalline layers (sec. 2.2.1), and amorphous layers (sec. 2.2.2).

2.2.1. Non-uniformly doped or defected crystals

The simulation system is similar to the one used by Ries and Bassler [14]: a regular cubic lattice containing $N_x \times N_y \times N_z$ sites with periodic boundary conditions imposed in directions perpendicular to the applied field. A fraction c of the total number of the lattice nodes are chosen as hopping centres, and distributed along the direction of the external field E according to $S(x; D)$. The energies taken from the Gaussian distribution are then assigned to the transport sites. The remaining fraction 1 – c of the lattice nodes is labelled as host sites not participating in the transport process. The carrier packets $n(x, t)$ were obtained by averaging the random walks (between $x = 0$, and $x = L$) of $\approx 10^3$ individual carriers. An individual hop from a given occupied centre, say at $\mathbf{r}_0$, to one of the neighbouring empty centres, located at $\mathbf{r}_i$, $i = 1,...,342$ (from a $7 \times \times 7 \times 7$ cube centred on $\mathbf{r}_0$), has been realised as follows. According to the average jump rates ν_{0i} [14], random jump rates ν_i are chosen from an exponential distribution, and the most probable jump is accepted in the simulations. The MC simulations have been performed for a wide range of the system parameters: centres concentration c, $0.1 \le c \le 1.0$; standard deviation σ of the Gaussian distribution in energy, $0.0 \le \sigma \le 7.0$ kT; spatial exponential non-uniformity parameter L/D, $0.0 \le L/D \le 3.0$. The system dimensions were $N_x = N_y = 40$, $40 \le N_z \le 4000$.

For an decreasing in x hopping centres density the carriers moving towards $x = L$ enter the region of harder jumps, and their motion is slowed down (contrary to the MT mechanism). With increasing degree of non-uniformity, L/D in the exponential case, the average slopes before the effective time-of-flight increase, so that the increase of

L/D acts qualitatively as the decrease of the centres concentration c, or/and the increase of the energetic distribution width σ. However, there is an important difference in temporal variations of the current slope. In uniform structures the current initially decays more rapidly than just before the effective time of flight, due to the carrier relaxation in energy. On the contrary, in sufficiently non-uniform layers ($L/D \geq 1$) with a decreasing in space centres concentration, the effect is dominated by the influence of increasing average distance between the transport sites, and the current profile is steeper immediately before the time of flight than at much shorter times, when the most rapid carrier relaxation in energy occurs. After the effective time of flight the currents decrease more slowly for higher L/D, so that the increasing non-uniformity acts qualitatively again as the decrease of c, or/and the increase of σ.

On the other hand, the increasing with x centres density makes the carrier motion easier near the collecting electrode, and the effect of the spatial non-uniformity is quite different. For small times the current profiles are even steeper then for the corresponding transients in uniform systems, but at longer times after injection the rate of the current decay decreases to zero, and for sufficiently dense systems, a sufficient value of the non-uniformity parameter (and not too wide energetic centres distribution), the current increase is observed, which reflects an exponential increase of the effective carrier packet drift velocity near the collecting contact. The occurrence of the current maxima immediately before the final current decay in a quite wide range of the parameters is the most characteristic feature of the carrier transport in the layers with increasing in x total centres density (Fig. 2). After the effective time of flight the current slopes decrease with increasing degree of non-uniformity, L/D, similarly as in the case of decreasing in space centres density.

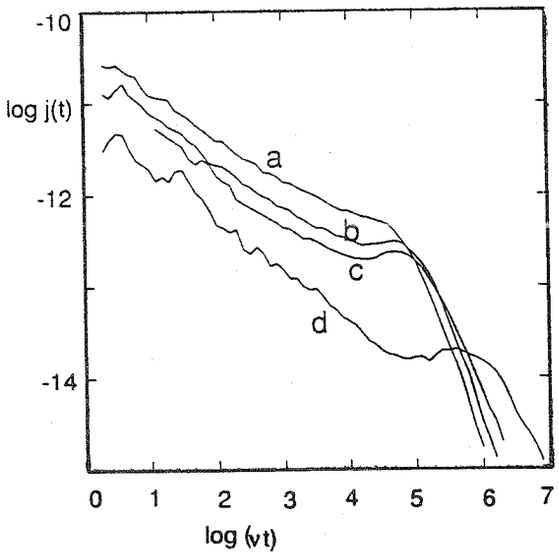

Fig. 2. R-ε-hopping transient currents, MC simulation, Gaussian energetic centres distribution with standard deviation $\sigma = 3.5\ kT$, $S(x;\ D) \sim \exp(x/D)$, $c = 0.2$, non-uniformity parameter $L/D = 0.0$ (a), 1.0 (b), 2.0 (c), 3.0 (d). Cubic lattice constant $d = 7 \times 10^{-10}$ m, wave-function overlap parameter $2d\alpha = 5.0$ (α – reciprocal Bohr radius), $T = 400$ K, $E = 1.1 \times \times 10^8$ V/m; the time is normalised to $1/\nu = \tau$, where τ is the average dwell time of carrier located at a site of undiluted (c = 1.0) cubic lattice with six nearest neighbours, current in arbitrary units [26].

The comparison of the curves for exponentially decreasing and increasing centres density (and the same L/D, c and σ) shows a remarkable polarity dependence of transient currents [26], similarly as in the case of the MT mechanism [18, 20]. The effective time of flight is an important characteristic of the current profiles. Its dependence on various parameters (in the base of the MC simulations) has been discussed in [26].

Most of the simulations have been performed for extremely thin layers (≈ 0.05 μm), and one could suspect that the strong influence of even moderate centres density non-uniformity on the current profiles is limited only to such ultrathin systems. This is not the case. Recently, we have performed extensive simulations on much greater samples (up to $L \approx 3$ μm, averaging 1000 individual carriers random walks over up to $\approx 10^7$ hopping centres in the simulation box). It turns out that all the characteristic features of the current profiles due to the centres density non-uniformity remain unchanged or even, under certain conditions, become more distinct [27].

2.2.2. Non-uniform amorphous systems

As far as amorphous hopping systems are concerned, a new algorithm has been developed [24]. Instead of performing carriers random walk on once generated centres distribution in the whole sample (up to $\sim 10^7$ hopping sites) we generate a random neighbourhood of each actually occupied centre. With its continuous configuration changes, the algorithm corresponds to the effective-medium-approximation. Moreover, the repeated fast jumps within isolated high density clusters (which only slightly influence the current profiles, but consume a lot of CPU time) are eliminated automatically, and the layer is really infinite in the directions perpendicular to the external field (no periodic boundary conditions need to be applied). The TOF transient current profiles in amorphous layers have been discussed in [24, 25]. The overall influence of the centres non-uniformity on the TOF currents is similar as in the lattice gas model of the previous section, and the detailed comparison of the cases of full, and limited off-diagonal disorder goes beyond the scope of the present paper.

3. NON-OHMIC HOPPING CONDUCTIVITY IN NON-UNIFORM LAYERS

The stationary current-field and conductivity-field characteristics of random three dimensional hopping systems, at arbitrary electric field strength can be calculated numerically within a model proposed in [3]. We have applied the model to the investigation of the characteristics of spatially non-uniform layers, mainly with exponential shape function $S(x;\ D) \sim \exp(-x/D)$, and Gaussian energetic centres distribution. Extensive numerical calculations have been performed for r-, and r-ε-hopping in the cases of a strong electron-phonon interaction (small polarons in amorphous solids), and a weak electron-phonon interaction (band-like transport). In what follows we limit ourselves to the small polaron transport (for band-like transport see [10], and references therein).

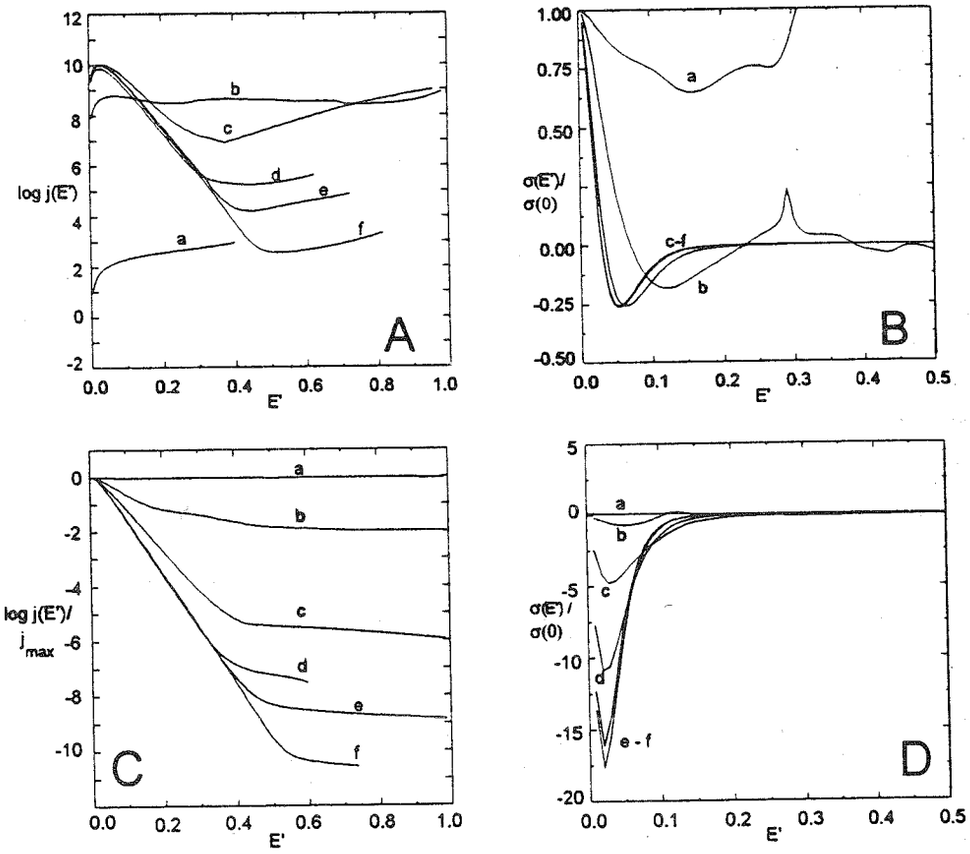

Fig. 3. A: current-field r-hopping characteristics ($E' = eE\beta\alpha^{-1}/2$, current in arbitrary units) in their dependence on the non-homogeneity parameter L/D: a) $L/D = 0.0$; b) $L/D = 0.5$; c) $L/D = 1.0$; d) $L/D = 1.5$; e) $L/D = 2.0$; f) $L/D = 2.5$, strong electron-phonon coupling; B: differential conductivities $\sigma(E')/\sigma(E' = 0)$ calculated from characteristics a-f of Figure 3A [9]; C, D: as in figures 3A, 3B, respectively, for a weak electron-phonon coupling [10].

The basic equations describing the hopping conductivity are as follows. For an electric field E of arbitrary strength, the density j of the dc hopping current is given by

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

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

where R_m is the position of the m-th hopping site, Ω – volume of the system, N – total site number within volume Ω, and $i(m', m)$ is the current running from site m' to site m, $V_{m'm} = V_{m'} - V_m$, $V_m = E_m + eu_m$, E_m – energy of the m-th site, e – elementary charge, u_m – potential of the external field E at the point R_m, $\beta = kT$, ρ_m – the occupation probability of site m, $W_{m'm}$ – the symmetrised hopping probability. The latter in the limits of strong and weak electron-phonon interactions are: $W_{mm'} = W_0 \exp(-2\alpha|R_{mm'}|)$, and $W_{mm'} = W_0' [\sinh(\beta|V_{mm'}|/2)]^{-1} \exp(-2\alpha|R_{mm'}|)$, respectively. The prefactor W_0 depends only weakly on E, R_m and ε_m. We resolve numerically the system of the Kirchhoff equations for each node of the equivalent random resistor network, that is, the system of 500-1000 non-linear algebraic equations for occupations ρ_m (or equivalently for local chemical potentials) for each value of E (usually about 1000 points).

Figure 3A shows current-electric field characteristics for various degrees of exponential non-uniformity (L/D) for a strong electron-phonon interaction in layers with no energetic disorder (so called r-hopping, or nearest-neighbour hopping). The characteristics obtained for non-uniform centres distributions with L/D in the range 1.0-2.5, reveal an N-like course, showing a current maximum at fairly low fields, followed by a current decay down to a minimum value, and by a subsequent exponential current increase. The overall shape of the curves remains similar. The quantities which depend remarkably on L/D are the position and the depth of the current and conductivity minima. With increasing sample non-uniformity the current minima occur for increasing fields, and the minimal current values become lower.

Figure 3B shows differential conductivities $\sigma(E')$ corresponding to the j vs E' characteristics of figure 3A (normalised to $\sigma(E' = 0)$). The curves corresponding to $L/D > 1.0$ practically coincide, and thus the relative differential conductivity variations as the function of the applied field do not depend on the degree of spatial non-homogeneity in the centres distribution, assuming a specific shape, common to all sufficiently non-uniform systems. The point to be noted is that the conductivity saturation occurs at a certain critical value of the applied field, E'_c, only weakly dependent on L/D. Thus, the non-uniform systems become ohmic at E'_c, and remain ohmic up to the highest 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 the exponential conductivity increase. There is also no negative-conductivity region, in contradistinction to the non-uniform systems. For comparison, Figures 3C, and 3D show the characteristics as in Figures 3A, and 3B, respectively, for a weak electron-phonon coupling.

Figure 4 refers to systems with a Gaussian energetic distribution of centres (so called r-ε-hopping, or variable-range hopping). Figure 4A shows several characteristics for rather strongly non-uniform system ($L/D = 3.0$, exponential spatial centres distribution), whereas Figure 4B shows the corresponding conductivity-field characteristics (for standard deviations of the energetic distribution equal to 3 kT, 6 kT, 12 kT, and 16 kT). The curves obtained for spatially uniform systems ($L/D = 0.0$) are presented in Figures 4C, and 4D, respectively. As it is seen, in uniform layers for not too high an energy spread in the hopping centres, 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 (Fig. 4D). 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 [4].

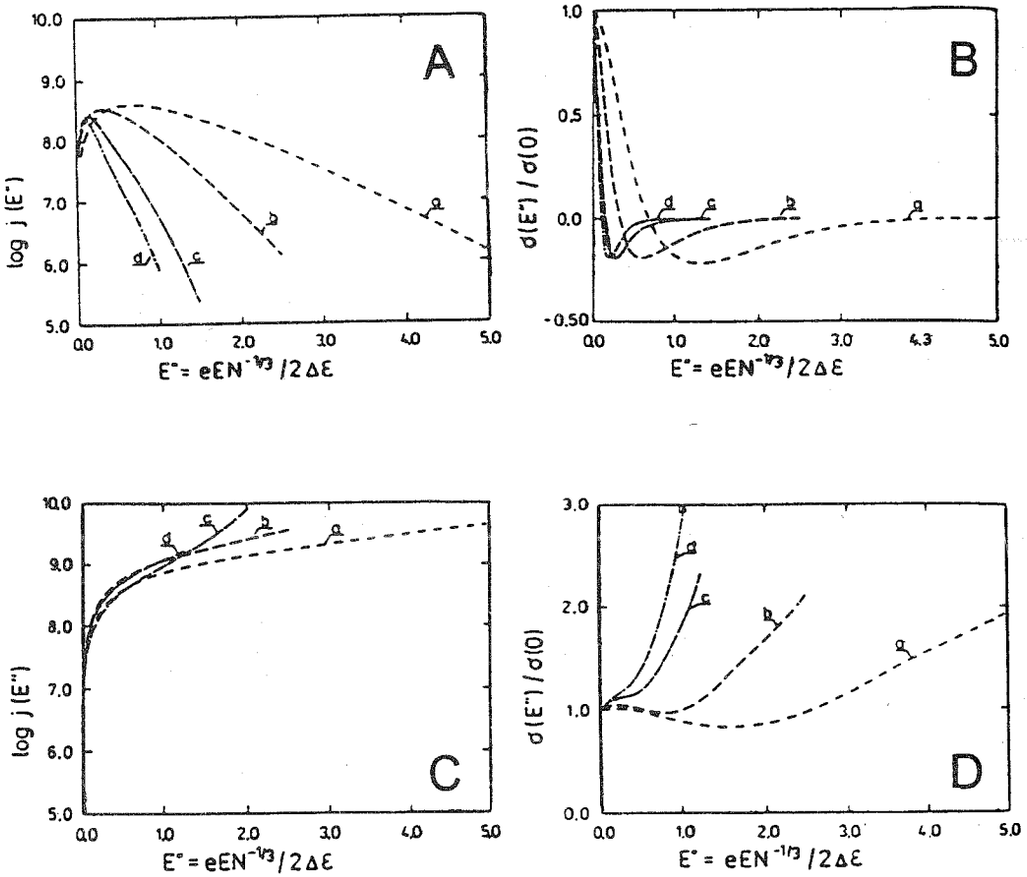

Fig. 4. A: current-field ($E'' = eEN^{-1/3}/2\Delta\varepsilon$, N-centres average concentration, $\Delta\varepsilon$ - sixfold standard deviation σ of the centres energetic Gaussian distribution, current in arbitrary units) characteristics for the non-uniformity parameter L/D = 3.0; a) σ = 3 kT, b) σ = 6 kT, c) σ = 12 kT, d) σ = 16kT, strong electron-phonon interaction; B: differential conductivities $\sigma(E'')/\sigma(E'' = 0)$ calculated from characteristics a-d of Figure 4A; C, D: as in figures 4A, 4B, respectively, for uniform centres distribution (L/D = 0.0) [9].

Our numerical calculations reveal that in sufficiently non-uniform systems, for all temperatures, there is no longer a conductivity maximum, but a very 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 constant value, at least up to the fields consistent with the assumption of a field-independent carrier concentration (Fig. 4B). Such a behaviour of conductivity-field characteristic means, that the currents (Fig. 4A) assume their maximum value at fairly low fields, and are strongly damped at higher fields. The effect is due to the appearance of a wide spatial region of almost completely occupied centres, where the currents $i(m, m')$ between the centres tend to zero.

4. CONCLUDING REMARKS

Transient currents measured in the classical TOF experiment are highly sensitive to the spatial macroscopic-scale variations of the total centres concentration, both for MT, and H transport mechanisms. The detailed shape of the transients depend in a very complicated way on the system dilution, the width of the energetic centres distribution, and the spatial variations of the centres concentration, and because of difficulties in the analytical treatment, the MC method turns out to be the only effective tool for the study of the TOF current profiles in their dependence on various parameters. The low field and low injection TOF experiment is the best method for the sampling of spatial changes of transport centres parameters. Transient currents measured in the conditions of step-like switching on of the electric field are sensitive rather to the moments of the distribution function $S(x; D)$, then to its exact shape. Thus, from such experiments only the centroid position of the S-function could be determined [17, 19]. Also the discharge currents seem to yield only a very limited information on the layer non-uniformity [16].

As far as the stationary H-transport characteristics are concerned, the current-field characteristics, as well as the differential conductivity-field characteristics depend strongly, both quantitatively, and qualitatively on the degree of the spatial non-uniformity in the centres distribution over the layer thickness. The numerical implementation of the Bottger-Bryksin model reveals very interesting features specific to non-uniform layers. The most striking result is the appearance (in sufficiently non-uniform systems) of wide field-ranges of negative differential conductivity, which follow the ranges of positive differential conductivity at lower fields. Thus, the model predicts the opening-switch-like characteristics (which, in general, are of a great technological interest) of extremely thin, spatially non-uniform H-transport layers, placed between ideal Ohmic contacts.

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