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Charge transport in hopping systems under open-circuit conditions: computer simulation

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Abstract. Discharge under open-circuit conditions may be often used to find the drift mobility of charge carriers in thin films of low-mobility materials. The existing theories of the discharge assume domination of charge transport in the conduction band. In order to find whether the theories may be used for interpretation of experimental results in hopping systems a computer simulation was carried out. It has been shown that direct usage of the existing theories may lead to wrong values of drift mobility found from the discharge. The method of estimation of real value of mobility in the temperature range 100–500 K for various values of the decay parameter of the electron localized wavefunction and for various standard deviations of the energetic distribution of the localized states is presented in this paper.

1. Introduction

The mobility measurements in low-mobility disordered amorphous and strongly doped materials started many years ago [1, 2]. The main experimental method used for the measurements was the so-called time-of-flight (TOF) method. The method consists in measuring the transient current which is caused by movement of charge carriers injected into a sample at a surface. A sandwich-type sample is provided with well conducting (mainly metallic) electrodes. The injected charge move in the external electric field. The drift mobility μ may be obtained from the expression:

$$\mu = \frac{D^2}{V_0 t_{tr}} \tag{1}$$

where D is the sample thickness, V_0 is the voltage applied and t_{tr} is the transition time of charge carriers through the sample. A characteristic 'kink' on the current–time curve corresponds to the transition time. In the case of well defined band mobility of charge carriers the spatial concentration of moving charge carriers may be well described by a Gaussian distribution which broadens as a result of diffusion. Such a kind of charge transport is often called a Gaussian type of charge transport.

It was observed in the 1970s that some experimental data concerning disordered, amorphous or strongly doped materials could not be interpreted in this way because no characteristic 'kink' related to the transition time was detected. It was pointed out that such results were caused by non-Gaussian time development of the packet of charge carriers injected into the sample. In 1975 Scher and Montroll [3] presented their model of stochastic hopping

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to explain the observed dispersive transport. The time dependence of current in the Scher–Montroll (SM) model is given by:

$$I(t) \propto t^{-(1-\alpha)} \qquad \text{for } t < t_{tr} \tag{2}$$

$$I(t) \propto t^{-(1+\alpha)} \qquad \text{for } t > t_{tr} \tag{3}$$

where the disorder parameter α is a constant. It results from equations (2) and (3) that the transition time (corresponding to the time of transition of the fastest charge carriers) may be found from the double logarithmic plot $\log I(t)$ against $\log(t)$. During the next few years it was shown that both the multiple-trapping model [4–7] and trap-controlled hopping [8] also lead to the dispersive transport of charge.

The classical time-of-flight method sometimes cannot be used for very thin samples due to short-circuits originating during evaporation of the electrodes. In such a case the discharge under open-circuit conditions may be used to find the drift mobility of charge carriers [9–11].

The theoretical description of discharge under open-circuit conditions was proposed by Batra *et al* [12]. No top electrode is used in the open-circuit method. The free surface is charged using either a corona discharge [12–14] or a low-energetic electron beam [9–11]. The discharge of the system results from charge transport to the grounded electrode. The time dependence of the voltage is given by [12]:

$$V(t) = V_0 - \frac{\mu V_0^2}{2D^2} t \tag{4}$$

$$\frac{\mathrm{d}V}{\mathrm{d}T} = -\frac{\mu V_0^2}{2D^2} \tag{5}$$

for $t < t_{tr}$ and by:

$$V(t) = \frac{D^2}{2\mu} \frac{1}{t} \tag{6}$$

$$\frac{\mathrm{d}V}{\mathrm{d}t} = -\frac{D^2}{2\mu} \frac{1}{t^2} \tag{7}$$

for $t > t_{tr}$. The time corresponding to the voltage $V = 0.5\ V_0$ is the transition time of charge carriers which can be used in equation (1) to obtain the drift mobility.

The model proposed by Batra *et al* assumes domination of charge transport in the conduction band and neglects trapping and diffusion [12–14]. In this situation the question arises of whether the model can be used for description of discharge under open circuit conditions in hopping systems. It may be expected that the time development of packets of charge carriers in hopping systems differs from those in systems dominated by transport via extended states. Depending on the distribution of localized states this may lead either to dispersive transport or to semi-classical Gaussian-like transport showing a 'kink' on the current–time curve. However, even in the second case the time corresponding to the voltage $V = 0.5\ V_0$ may lead to wrong estimation of the drift mobility due to non-Gaussian time development of the charge packet. The aim of this paper is to solve the problem of applicability of equations (4)–(7) for open-circuit discharge in a hopping system in these cases, in which a 'kink' on the current pulses obtained from classical TOF experiments is observed. The problem of applicability of equations (4)–(7) for dispersive hopping systems will be a subject of further investigations.

2. Assumptions of the simulation

This paper concerns systems dominated with hopping transport in a narrow band of localized states at the Fermi level. The basic assumptions of the simulation are as follows:

(1) It was assumed that the transport of charge is due to hopping of charge carriers among localized states in a narrow band of states given by a normal distribution.
(2) The states constitute a three-dimensional regular network; the distance between the states was $R = 3$ nm. The size of the simulation sample was 0.3 μm $\times$ 0.3 μm $\times$ 0.6 μm (i.e. $100 \times 100 \times 200$ localized states). Every localized state i is characterized by its position in the network x_{lmn} and its energy E_i.
(3) The phonon frequency $\nu_{ph} = 10^{12}$ Hz.
(4) The value of $2\alpha R = 3, 5, 7, 9, 11, 13$ was taken (α is the decay of the electron localized wavefunction).
(5) The distribution of the state energies is the normal one. The standard deviation of energetic distribution σ_E was assumed to change in the range 0.01–0.16 eV.
(6) The temperature was assumed to change in the range 100–500 K.
(7) The probability of a jump between state i and state j was taken in the form [15]:

$$p_{ij} = \nu_{ph} \exp(-2\alpha R_{ij}) \exp\left(\frac{-(E_j - E_i - eF(x_j - x_i))}{kT}\right) \quad \text{for} \quad E_j - E_i - eE(x_j - x_i) > 0 \tag{8a}$$

$$p_{ij} = \nu_{ph} \exp(-2\alpha R_{ij}) \quad \text{for} \quad E_j - E_i - eE(x_j - x_i) \leqslant 0 \tag{8b}$$

where e is the electron charge, F is the intensity of electric field, E_i, E_j are the energies of i and j states and x_i, x_j are the positions of the states measured along the electric field.
(8) It is assumed that at the initial moment ($t = 0$) a number of carriers is injected into the sample and localized at the states of the first surface plane. The number of charge carriers was a few hundred, usually between 100 and 300. The shape of the current pulse was independent of the number of injected carriers provided that the number was great enough to give a smooth current pulse.
(9) The shape of current pulses was calculated using [16]:

$$i(t) \propto -\frac{1}{n_0}\frac{\mathrm{d}}{\mathrm{d}t}\left(\int_0^D n(x,t)\,\mathrm{d}x\right) + \frac{1}{n_0 D}\frac{\mathrm{d}}{\mathrm{d}t}\left(\int_0^D xn(x,t)\,\mathrm{d}x\right) \tag{9}$$

where $n(x, t)$ is the concentration of charge carriers at the distance x from the top electrode at the time t, n_0 is the total number of injected carriers and D is the sample thickness.

3. Short description of algorithm of simulation

The starting point of the simulation is the initial position of all charge carriers injected into the sample. The simulation consists of the following main steps:

(1) Registration of the initial position of all charge carriers.
(2) Calculation of probability of all jumps to the nearest neighbouring states for all the carriers.
(3) Stochastic selection of a jump for execution for every charge carrier. The selection takes into account the relative probability of all jumps resulting from equations (8*a*) and (8*b*).
(4) Calculation of the positions of every carrier after execution of the selected jumps and calculation of the time of execution of every executed jump.
(5) The history of each charge carrier can be calculated in this way. This history enables us to obtain the position of all carriers for a chosen set of moments (for instance the position for every 8 ns was registered in the case of data shown in figure 8). Having the positions it is possible to obtain the current pulse from equation (9).

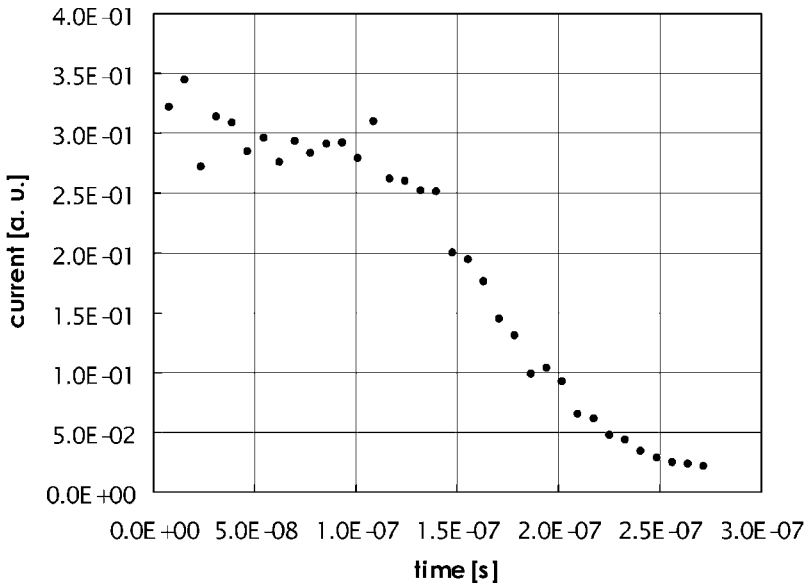

Figure 1. Current against time for $\sigma_E = 0.01$ eV, $T = 300$ K, $2\alpha R = 3$. Simulation carried out for the ordinary time-of-flight experiment. The current pulse is a typical example of Gaussian-like transport of charge.

4. Results of simulation

4.1. Simulation of transport under closed circuit conditions

In the case of complex computer simulations it is very important to test the algorithm used. In order to confirm the correctness of the algorithm used the simulation of a classical time-of-flight experiment was carried out. The simulation under closed circuit conditions was carried out for the voltage 6 V, which means that the electric field was equal to 10^7 V m^{-1}. Figures 1 and 2 show the current pulses for the standard deviation describing the width of the narrow band of states $\sigma_E = 0.01$ eV and 0.13 eV respectively. Figure 3 shows the data presented in figure 2 transformed to a double logarithmic scale. Figure 1 is a typical example of Gaussian-like transport of charge. The 'kink' on the current curve (in a linear scale) is quite clearly marked. Figure 2 corresponds to a classical dispersive transport, which is confirmed by figure 3. The curve shown in figure 3 may be described by equations (2) and (3), the sum of the slopes of the two parts being very close to -2. The following criteria resulting from the SM model were assumed to classify the pulses obtained from the simulation:

- No 'kink' on the current curve in a linear scale and the sum of slopes in the double logarithmic scale equal to -2 classifies the discharge as dispersive transport.
- Other cases are classified to non-dispersive transport.

Figure 4 shows the regions of dispersive and non-dispersive transport depending on the values of σ_E and temperature. In general the dispersive transport occurs for the higher values of σ_E and the lower temperatures. The classical (or Gaussian-like) type of transport, i.e. the transport showing a 'kink' in the current pulse, tends to occur at the lower values of σ_E and the higher temperatures. This result is in general agreement with the results obtained by Borsenberger *et al* [17]. Some results in the area close to the border line are difficult for classification because on one hand they do not have an easily noticeable 'kink' in the current

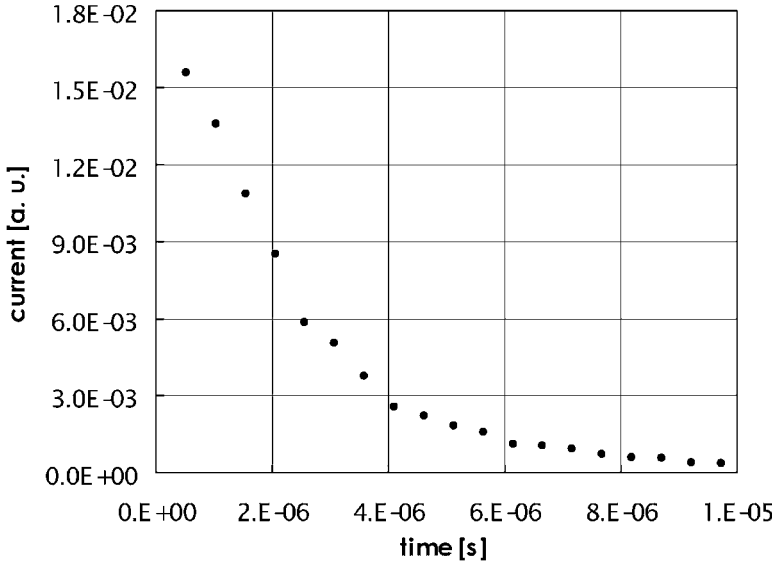

Figure 2. Current against time for $\sigma_E = 0.13$ eV, $T = 300$ K, $2\alpha R = 3$. Simulation carried out for the ordinary time-of-flight experiment. The current pulse is an example of dispersive transport (see also figure 3).

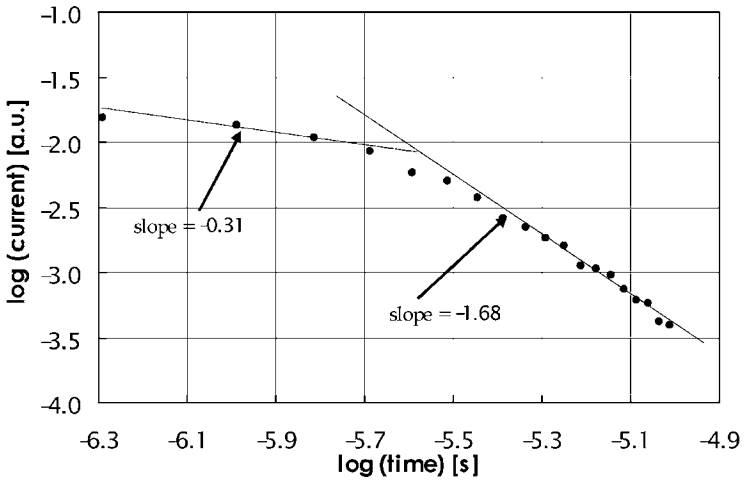

Figure 3. Log(I) versus log(t) for $\sigma_E = 0.13$ eV, $T = 300$ K, $2\alpha R = 3$. The sum of slopes of the two parts of current curve is very close to -2.

pulse, but on the other hand the sum of slopes in the double logarithmic scale is not equal to -2. These results were classified as examples of non-dispersive transport.

In the case of hopping transport in a narrow band of localized states the drift mobility may be described by [18]:

$$\mu = (1/6)(eR^2/kT)\nu_{ph} \exp(-2\alpha R) \exp(-W/kT) \tag{10}$$

where W is the effective activation energy related to the width of the band of localized states. It may be supposed that the activation energy W should be comparable to the standard deviation σ_E, but the relation is not known precisely. Comparing the values of mobility obtained from

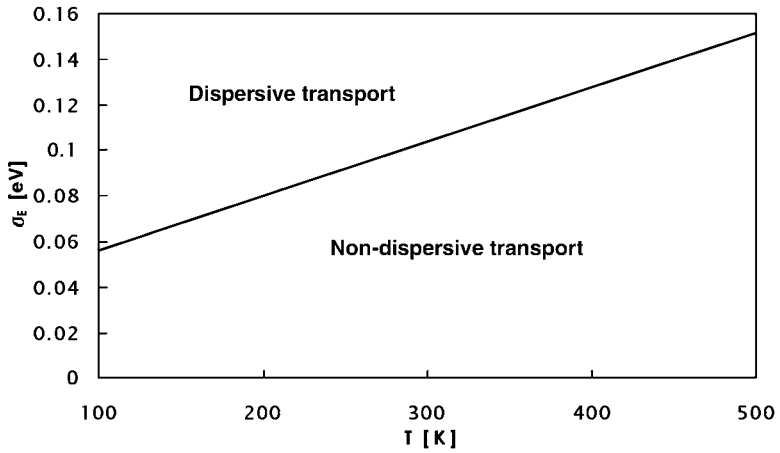

Figure 4. The transition from non-dispersive to dispersive transport in the investigated hopping systems. $2\alpha R = 3$.

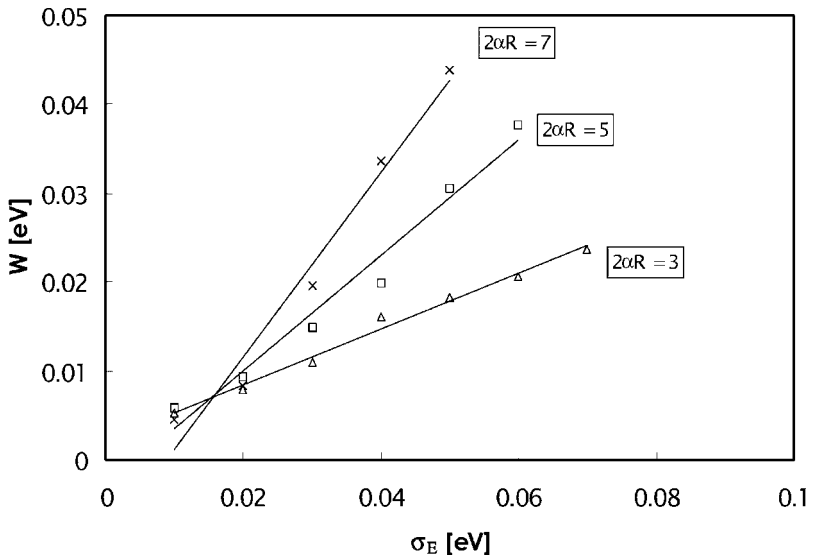

Figure 5. The relations between the activation energy of mobility and the standard deviation σ_E for various values of $2\alpha R$. The relations may be well described by the following linear equations: for $2\alpha R = 3$ $W = 0.3151\sigma_E + 0.0021$ eV; for $2\alpha R = 5$ $W = 0.6494\sigma_E - 0.003$ eV; for $2\alpha R = 7$ $W = 1.0395\sigma_E - 0.0092$ eV.

the simulation with the values resulting from equation (10) one should obtain confirmation (or negation) of correctness of the algorithm used. The problem is how to determine the value of W in the case of transport in a narrow band of localized states given by a normal distribution. For instance, assuming $W = 2\sigma_E$ or $W = 4\sigma_E$ we obtain the ratio of the resulting mobilities $\mu(2\sigma_E)/\mu(4\sigma_E)$ (at $T = 300$ K) close to one order of magnitude for $\sigma_E = 0.03$ eV and exceeding two orders of magnitude for $\sigma_E = 0.07$ eV. Such a sensitivity of the exponential function to the value of W makes the problem of comparison rather difficult. In this situation it is necessary to find the relation between the effective activation energy W and the standard

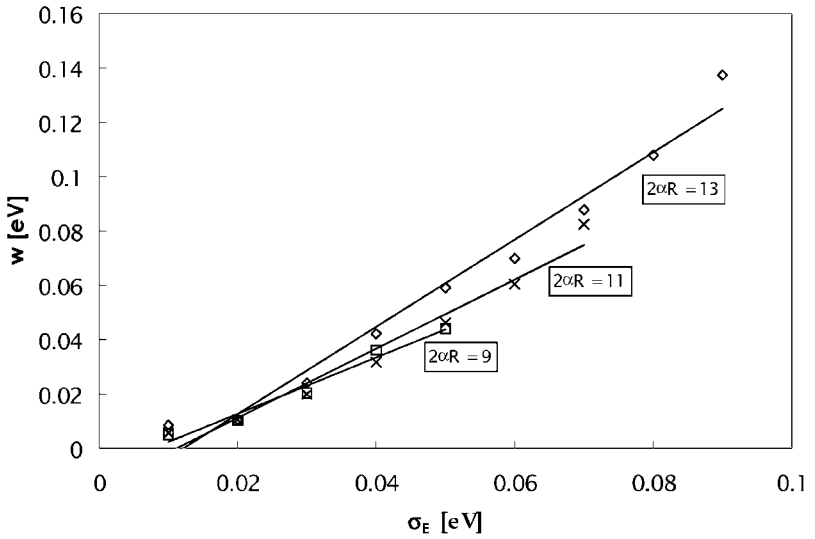

Figure 6. The relations between the activation energy of mobility and the standard deviation σ_E for various values of $2\alpha R$. The relations may be well described by the following linear equations: for $2\alpha R = 9$ $W = 1.0366\sigma_E - 0.0079$ eV; for $2\alpha R = 11$ $W = 1.2716\sigma_E - 0.0141$ eV; for $2\alpha R = 13$ $W = 1.6029\sigma_E - 0.0192$ eV.

deviation σ_E in order to compare the theoretical (μ_{theor}) and simulated (μ_{TOF}) values of drift mobility. Using the results of simulations of the TOF experiment these relations have been determined (by finding the activation energy for various values of σ_E). Figures 5 and 6 show the obtained relations between the activation energy W and the standard deviation σ_E for various values of the localization parameter $2\alpha R$. The relations prove to be well described by the linear formulae:

$$W = a\sigma_E + b \tag{11}$$

where a and b are parameters dependent on the value of $2\alpha R$ and are given by:

$$a = 0.1186 \times 2\alpha R + 0.037 \tag{12}$$

$$b = (-0.002 \times 2\alpha R + 0.0073)\ \text{eV}. \tag{13}$$

Using the above $W(\sigma_E)$ dependences the comparison of the mobility resulting from the simulation (μ_{TOF}) and equation (10) (μ_{theor}) was made. The ratio μ_{TOF}/μ_{theor} was found to be close to unity. Summarizing the results of the simulations of the TOF experiment we obtain:

- Gaussian-like pulses for smaller values of the standard deviation σ_E and higher temperatures;
- typical dispersive pulses for greater values of σ_E and lower temperatures (as expected, see for instance [17]). The dispersive pulses are well described by SM theory;
- values of the drift mobility being in good agreement with the theoretical expectations resulting from equation (10).

The above summary strongly justifies the conclusion that the algorithm used correctly simulates the charge transport in the assumed hopping system.

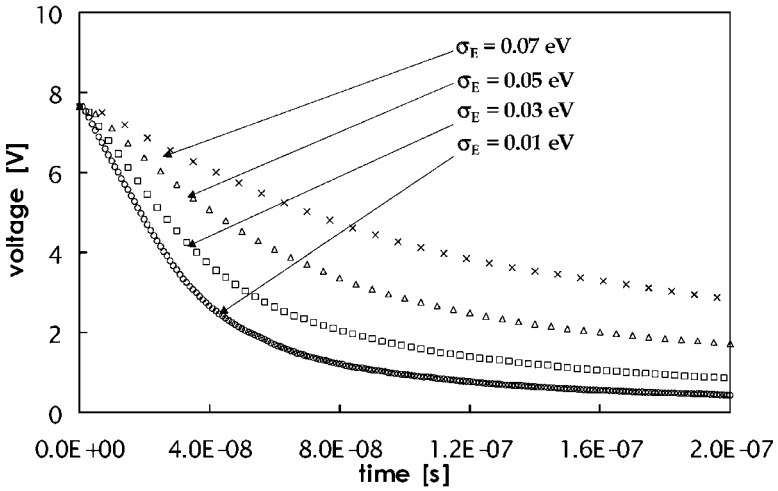

Figure 7. Discharge under open circuit conditions for various standard deviations of the energetic distribution of the localized states. $2\alpha R = 3$.

4.2. Simulation of discharge under open circuit conditions

Figures 7 and 8 show the results of simulated discharge under open circuit conditions. Figure 7 shows the results of simulation at 300 K for various values of the standard deviation σ_E and figure 8 shows the discharge for the standard deviation $\sigma_E = 0.08$ eV at various temperatures. It results from the data obtained that:

(1) The discharge is well described by equations (4)–(7) for the low values of the standard deviation σ_E, i.e. for these values of σ_E the discharge curve consists of two different parts including the straight-line one for the initial part of discharge. The contribution of the straight-line part of the discharge decreases with increasing standard deviation σ_E, which results from non-classical development of the charge packet.

(2) With increasing temperature the contribution of the straight-line part of the discharge increases, i.e. equations (4)–(7) describe the discharge better for the higher temperatures.

It results from the above statements that the increasing contribution of off-diagonal disorder (i.e. the increasing value of σ_E) leads to increasing discrepancy between the discharge pulses and the Batra model. According to equations (4)–(7) the transit time t_{tr} of charge carriers corresponds to the voltage $V(t_{tr}) = 0.5\ V_0$ (V_0 is the initial voltage). The transit time is used to obtain the drift mobility $\mu_{1/2}$ estimated from the open circuit discharge. This method of estimation of the drift mobility might be also used for the pulses which differ to some extent from the classical pulses resulting from the Batra model. The question arises of what is the relation between the mobility $\mu_{1/2}$ obtained from such an estimation of the transit time in the assumed hopping system and the real drift mobility. This relation (if known) should enable us to interpret the voltage pulses obtained under open circuit conditions in poorly conducting hopping systems.

There are two ways to find the relation between the mobility $\mu_{1/2}$ obtained from the simulation under open circuit conditions and the real drift mobility. The first one is to compare $\mu_{1/2}$ and the mobility resulting from equation (10). The second one is to compare $\mu_{1/2}$ and the mobility obtained from the simulation of the classical TOF experiment μ_{TOF}. In the latter case the mobility obtained for the TOF experiment is considered to be the real drift mobility of

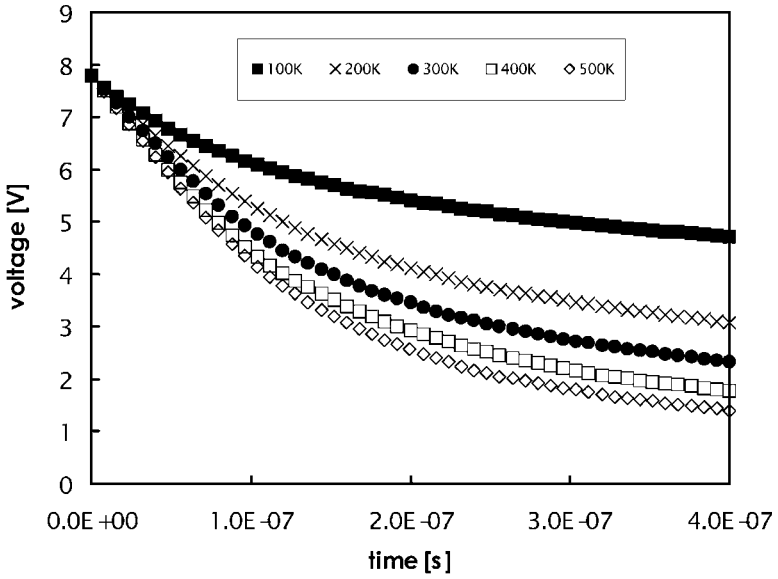

Figure 8. Discharge under open circuit conditions for various values of temperature. The energetic standard deviation $\sigma_E = 0.08$ eV, $2\alpha R = 3$.

Table 1. The ratio $\mu_{real}/\mu_{1/2}$ for various values of the localization parameter $2\alpha R$ and the standard deviation σ_E at $T = 100$ K.

$2\alpha R$	$\sigma_E = 0.01$ eV	0.02 eV	0.03 eV	0.04 eV	0.05 eV	0.06 eV	0.07 eV	0.08 eV
3	1.58	1.639	2.03	2.11	—	—	—	—
5	1.81	2.35	2.77	3.82	4.74	—	—	—
7	2.19	3.18	3.94	3.62	5.01	—	—	—
9	2.02	2.78	—	—	—	—	—	—
11	1.80	—	—	—	—	—	—	—
13	1.38	—	—	—	—	—	—	—

charge carriers (μ_{real}) in the assumed hopping system. Using the first way one must take into account that any inaccuracy of determination of $W(\sigma_E)$ (W is necessary to use equation (10)) leads to some systematic error of the comparison. The second way of comparison enables us to avoid any systematic errors, because the only difference between the two simulations is the kind of simulated experiment. In other words the influence of non-Gaussian development of the packet of charge carriers on the discharge under open circuit conditions is the only factor taken into account in the second way of comparison. For this reason the second way of comparison was chosen in order to test the applicability of Batra's equations for interpretation of open circuit discharge in the hopping system. The presented results (see tables 1–5) are limited to the cases of Gaussian-like transport (i.e. to those cases for which a detectable 'kink' on current pulses in the classical TOF experiment was found). The cases of dispersive transport will be a subject of further investigations.

The following conclusions result from the data presented in tables 1–5:

(1) The ratio $\mu_{real}/\mu_{1/2}$ depends both on the standard deviation σ_E and the localization parameter $2\alpha R$. The latter dependence is not pronounced strongly and has its maximum for $2\alpha R$ between 5 and 9.

Table 2. The ratio $\mu_{real}/\mu_{1/2}$ for various values of the localization parameter $2\alpha R$ and the standard deviation σ_E at $T = 200$ K.

$2\alpha R$	$\sigma_E = 0.01$ eV	0.02 eV	0.03 eV	0.04 eV	0.05 eV	0.06 eV	0.07 eV	0.08 eV
3	1.28	1.329	1.73	1.57	2.05	—	—	—
5	2.08	1.94	1.50	2.59	2.43	2.17	—	—
7	1.96	2.17	2.18	2.36	—	—	—	—
9	1.81	1.85	2.22	—	—	—	—	—
11	1.74	2.07	2.31	2.41	2.77	—	—	—
13	1.538	1.86	1.84	2.02	1.93	3.02	—	—

Table 3. The ratio $\mu_{real}/\mu_{1/2}$ for various values of the localization parameter $2\alpha R$ and the standard deviation σ_E at $T = 300$ K.

$2\alpha R$	$\sigma_E = 0.01$ eV	0.02 eV	0.03 eV	0.04 eV	0.05 eV	0.06 eV	0.07 eV	0.08 eV
3	1.31	1.38	1.58	1.41	1.62	1.85	1.83	—
5	2.06	2.43	2.50	2.17	2.65	2.38	—	—
7	2.61	2.56	2.31	2.57	1.88	—	—	—
9	1.81	1.85	2.22	—	—	—	—	—
11	1.64	1.80	1.78	1.96	2.15	2.2	1.89	—
13	1.78	1.74	1.66	2.11	1.97	2.06	2.19	2.12

Table 4. The ratio $\mu_{real}/\mu_{1/2}$ for various values of the localization parameter $2\alpha R$ and the standard deviation σ_E at $T = 400$ K.

$2\alpha R$	$\sigma_E = 0.01$ eV	0.02 eV	0.03 eV	0.04 eV	0.05 eV	0.06 eV	0.07 eV	0.08 eV
3	1.34	1.30	1.46	1.41	1.49	1.50	1.53	—
5	1.99	1.65	1.88	2.11	2.22	2.64	—	—
7	2.83	2.80	2.90	2.50	3.16	—	—	—
9	1.93	2.16	1.90	2.16	2.13	—	—	—
11	1.68	1.84	1.76	1.69	1.83	1.78	1.71	—
13	1.54	1.72	1.70	1.96	1.78	1.97	1.94	1.89

Table 5. The ratio $\mu_{real}/\mu_{1/2}$ for various values of the localization parameter $2\alpha R$ and the standard deviation σ_E at $T = 500$ K.

$2\alpha R$	$\sigma_E = 0.01$ eV	0.02 eV	0.03 eV	0.04 eV	0.05 eV	0.06 eV	0.07 eV	0.08 eV
3	1.08	1.01	1.08	0.96	0.98	1.04	1.04	—
5	2.02	2.14	2.06	1.78	2.11	2.44	—	—
7	2.01	2.15	2.26	2.59	2.06	—	—	—
9	1.93	2.17	1.90	2.16	2.13	—	—	—
11	1.69	1.82	1.80	1.70	1.81	1.89	1.71	—
13	1.55	1.62	1.65	1.91	1.60	2.02	1.83	—

(2) The ratio $\mu_{real}/\mu_{1/2}$ tends to increase with increasing value of σ_E.

(3) The ratio $\mu_{real}/\mu_{1/2}$ tends to decrease with increasing temperature. For 100 K the ratio becomes quite remarkable and is close to 5 for $2\alpha R = 5$–7 and $\sigma_E = 0.04$–0.05 eV.

Measurements of the drift mobility in diamond-like carbon films [9–11] are an example of application of the discharge under open circuit conditions for hopping systems. At $T = 300$ K the mobility was measured to be 7.1×10^{-6} cm^2 V^{-1} s^{-1}, the activation energy in the temperature range between 200 K and 300 K was found to be 0.03 eV. The value of $2\alpha R$

for amorphous carbon structures was estimated to be in the range 5–7 [19] (it is necessary to know the localization parameter $2\alpha R$ in order to estimate σ_E which in turn is necessary to apply the data shown in the tables). Using the corresponding values of $\mu_{real}/\mu_{1/2}$ shown in table 3 we find the expected real value of the mobility equal to 1.8×10^{-5} cm^2 V^{-1} s^{-1} at $T = 300$ K.

5. Conclusions

The following conclusions resulting from the presented results may be put forward:

- Using the model presented by Batra *et al* (equations (4)–(7)) for interpretation of discharge under open circuit conditions in hopping narrow-band systems with off-diagonal disorder may lead to a wrong estimation of drift mobility. The ratio of the real mobility μ_{real} to the mobility obtained from application of the Batra model $\mu_{1/2}$ depends on the temperature, the standard deviation σ_E describing the energetic distribution of localized states and on the localization parameter $2\alpha R$.
- The ratio $\mu_{real}/\mu_{1/2}$ is close to 5 for $T = 100$ K and $2\alpha R = 5$–7. For the higher temperatures the ratio is close to 2 for the most values of $2\alpha R$ except for $2\alpha R = 3$, for which the ratio becomes close to 1 for the higher temperatures. In the last cases the Batra model well describes the discharge under open circuit conditions in the discussed hopping system.

It may be supposed that the ratio $\mu_{real}/\mu_{1/2}$ should increase for these higher values of the standard deviation σ_E for which the dispersive transport occurs. Applicability of the discharge under open circuit conditions for the region of dispersive transport will be a subject of further investigations.

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