Postprint of: Tomaszewicz W., On weakly dispersive multiple-trapping transport, Journal of Non-Crystalline Solids, Vol. 355, Iss. 24–27 (2009), pp. 1414-1418, DOI: 10.1016/j.jnoncrysol.2009.05.039

On weakly dispersive multiple-trapping transport

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Abstract

The equations for multiple-trapping carrier transport, corresponding to the time-of-flight method, are approximately solved under assumption that the majority of carriers is in thermal quasi-equilibrium. The solutions show a Gaussian shape of the carrier packet. For dispersive transport regime, mean velocity of the carrier sheet decreases in time and its dispersion grows faster than the square root of time. The accuracy of obtained formulas is verified by a Monte Carlo calculations for exponential and Gaussian trap distributions. A satisfactory agreement is obtained up to the effective carrier transit-time, provided that trap density falls-off sufficiently fast in the energy gap. A new method of determining the energetic trap profiles in disordered solids from the time-of-flight measurements is proposed.

Key words:
time-of-flight method, dispersive transport, multiple-trapping model
PACS: 72.20.Jv, 72.80.Ng

1. Introduction

The time-of-fight (TOF) technique is a straightforward, frequently applied method of investigating the carrier transport in low-conductivity solids, both crystalline and amorphous. The sample is sandwiched between two electrodes with a constant voltage applied, and the excess carriers are generated by a short light pulse. The carrier motion in the sample induces a current transient in the measuring circuit. From the form of transient, as well as from its dependence on experimental parameters, the information about carrier transport mechanism can be inferred.

As regards disordered solids, there exist two basic mechanisms of carrier transport — multiple trapping (MT) and hopping. In the case of MT the transitions of carriers between extended states and localized states (traps) gap occur, whereas in the case of hopping the straightforward carrier transitions between localized states take place. For both mechanisms, the carrier transport may

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be either Gaussian or dispersive. The first transport regime is characterized by constant velocity and Gaussian shape of carrier sheet in a solid, the second one — by gradual decrease of mean velocity and extremely large dispersion of carrier packet. The first successful theory of dispersive transport was developed by Scher and Montroll [1], who attributed this phenomenon to very slow equilibration of charge carriers over localized states.

The Scher-Montroll theory initiated extensive investigations on dispersive transport (for earlier works, see the reviews [2, 3]). In spite of this, some problems seem to be still unresolved. In particular, this concerns the simplified description of MT dispersive transport, given by Tiedje and Rose [4] and by Orenstein and Kastner [5]. Their main idea was that, for specific trap distributions, the majority of trapped carriers is in thermal quasi-equilibrium with the free carriers. This approach was utilized in many subsequent papers. However, its validity was questioned by Arkhipov *et al* [6], since it does not describe the broadening of carrier packet. The main aim of the present paper is to resolve this controversy.

2. Transport equations

The present investigations are based on the standard MT model, assuming very small trap occupancy, electric field uniformity in the sample as well as negligible carrier diffusion. It should have in mind that the first assumption may be incorrect in the final stage of carrier transport, due to gradual filling of deeper traps. However, the analytical description of MT transport, taking into account the saturation of trap occupancy, is difficult. Only some special cases were studied so far [7].

In the following formulas, the free and trapped carrier densities are denoted by $n(z,t)$ and $n_t(z,t)$, respectively, where $z = x/\mu_0 E$ is the reduced space variable (x is the space variable, μ_0 — the free carrier mobility and E — the electric field strength) and t is the time variable. The MT carrier transport can be described by the continuity equation:

$$\frac{\partial}{\partial t}\left[n(z,t) + n_t(z,t)\right] + \frac{\partial n(z,t)}{\partial z} = 0, \tag{1}$$

and the equation relating the free and trapped carrier densities [8]:

$$n_t(z,t) = \int_0^t \Phi(t')\, n(z,t-t')\, \mathrm{d}t'. \tag{2}$$

Here, the function $\Phi(t)$ determines the probability that carrier is trapped in a time unit and remains in trap until time t. This function is given by the formula

$$\Phi(t) = C_t \int_0^\infty N_t(\varepsilon) \exp\left[-t/\tau_r(\varepsilon)\right] \mathrm{d}\varepsilon, \tag{3}$$

where C_t is the carrier capture coefficient, $N_t(\varepsilon)$ is the trap density at the energy level ε per unit energy, and

$$\tau_r(\varepsilon) = \nu_0^{-1} \exp(\varepsilon/kT) \tag{4}$$

is the mean lifetime of trapped carrier (ν_0 is the frequency factor, k — the Boltzmann constant and T — the sample temperature). The energy ε is measured from the edge of allowed band.

The current intensity $I\,(t)$, registered in TOF experiment, equals the conduction current intensity in the sample, averaged over its thickness. Therefore

$$I(t) = \frac{I_0}{n_0 \tau_0} \int_0^{\tau_0} n(z,t)\,\mathrm{d}z, \tag{5}$$

where n_0 is the density of generated carriers, averaged over sample thickness, $\tau_0 = d/\mu_0 E$ and $I_0 = e n_0 \mu_0 E S$ are, respectively, the carrier time-of-flight and the initial current intensity in a trap-free sample (with d being the sample thickness, e — the elementary charge and S — the sample cross-sectional area). For times smaller than the effective carrier transit time τ_e through the sample, Eq. (5) may be rewritten as

$$I(t) = I_0 \frac{\mathrm{d}\bar{z}\,(t)}{\mathrm{d}t}, \quad t < \tau_e, \tag{6}$$

where the dash denotes averaging over spatial carrier distribution. The transit time τ_e is implicitly given by the formula

$$\bar{z}\,(\tau_e) = \tau_0. \tag{7}$$

3. Thermal quasi-equilibrium approximation

The progress of carrier thermalization in trapping states is characterized by the demarcation level $\varepsilon_0\,(t)$ [4, 5, 8], defined implicitly by the formula $\tau_r\,[\varepsilon_0\,(t)] = 1.8t$, which gives

$$\varepsilon_0\,(t) = kT \ln\,(1.8\nu_0 t)\,. \tag{8}$$

The level approximately separates the traps with equilibrium ($\varepsilon < \varepsilon_0\,(t)$) and non-equilibrium ($\varepsilon > \varepsilon_0\,(t)$) occupancy.

In the case of weakly dispersive transport, when the approximate thermal equlibrium between free carriers and majority of trapped carriers is established, Eq. (2) describing trapping/detrapping kinetics can be simplified. If the trap density decreases sufficiently fast in the energy gap, the main contribution to the integral in Eq. (3) should proceed from the energy region $\varepsilon < \varepsilon_0\,(t)$. The argument of exponential function in the integrand is then much larger than unity for almost all values of energy ε, and the function $\Phi\,(t)$ should differ significantly from zero only for a very small time values. The free carrier density in Eq. (2) can be then replaced by the initial terms of its Taylor series,

$$n\,(z, t - t') \approx n\,(z,t) - t' \frac{\partial n\,(z,t)}{\partial t}. \tag{9}$$

This results in approximate equation, describing carrier trapping/detrapping processes,

$$n_t\,(z,t) \approx \left[\Theta^{-1}\,(t) - 1\right] n\,(z,t) - \tau_s\,(t)\,\frac{\partial n\,(z,t)}{\partial t}, \tag{10}$$

where the functions:

$$\Theta^{-1}(t) = 1 + C_t \int_0^{\infty} N_t(\varepsilon)\,\tau_r(\varepsilon)\left[1 - \exp\left[-t/\tau_r(\varepsilon)\right]\right] \mathrm{d}\varepsilon, \tag{11}$$

$$\tau_s(t) = C_t \int_0^{\infty} N_t(\varepsilon)\,\tau_r^2(\varepsilon)\left\{1 - \left[1 + t/\tau_r(\varepsilon)\right]\exp\left[-t/\tau_r(\varepsilon)\right]\right\} \mathrm{d}\varepsilon. \tag{12}$$

In Eqs. (11) and (12) the last factors in integrands may be approximated by the unit step function, $H\left[\varepsilon_0(t) - \varepsilon\right]$, provided that the functions $N_t(\varepsilon)\,\tau_r(\varepsilon)$ and $N_t(\varepsilon)\,\tau_r^2(\varepsilon)$ vary sufficiently slowly with energy. Then:

$$\Theta^{-1}(t) \approx 1 + C_t \int_0^{\varepsilon_0(t)} N_t(\varepsilon)\,\tau_r(\varepsilon)\,\mathrm{d}\varepsilon, \tag{13}$$

$$\tau_s(t) \approx C_t \int_0^{\varepsilon_0(t)} N_t(\varepsilon)\,\tau_r^2(\varepsilon)\,\mathrm{d}\varepsilon. \tag{14}$$

It should be stressed that both integrals are calculated over the energy interval $0 \le \varepsilon \le \varepsilon_0(t)$, where trapped carriers are in thermal quasi-equilibrium.

The equations equivalent to Eq. (10), with the last term omitted, were obtained in [4, 5] under assumption of exact thermal equilibrium between free carriers and carriers trapped in the energy region $\varepsilon \le \varepsilon_0(t)$. As already indicated, this approach was criticized [6], because it does not describe the spatial carrier dispersion. The mentioned term approximately takes into account the deviations of carrier densities from their equilibrium values. It will be seen that presence of the term results in the finite spread of carrier packet.

4. Solution of transport equations

The approximate solution of the equations identical to (1) and (10) was already obtained in the paper [9], dealing with non-isothermal carrier transport, and has the form of:

$$n(z,t) \approx \frac{n_0 \tau_0 \Theta(t)}{2\left[\pi \xi(t)\right]^{1/2}} \exp\left\{-\frac{\left[z - \zeta(t)\right]^2}{4\xi(t)}\right\}, \tag{15}$$

$$n_t(z,t) \approx \left[\Theta^{-1}(t) - 1\right] n(z,t), \tag{16}$$

where the functions

$$\zeta(t) = \int_0^t \Theta(t')\,\mathrm{d}t', \tag{17}$$

$$\xi(t) = \int_0^t \tau_s(t')\,\Theta^3(t')\,\mathrm{d}t'. \tag{18}$$

Thus, in considered approximation, the carrier packet has a Gaussian shape. The 'centroid' and the RMS spread of carrier distribution are given respectively by the formulas

$$\bar{z}(t) = \zeta(t), \tag{19}$$

$$\sigma(t) = [2\xi(t)]^{1/2}\,. \tag{20}$$

The above results constitute straightforward extension of those obtained for Gaussian carrier transport [10, 11]. In such a case the functions $\Theta(t)$ and $\tau_s(t)$ are constant, which implies that $\bar{z}(t) \propto t$ and $\sigma(t) \propto t^{1/2}$.

Inserting the free carrier density (15) into integral (5) one obtains the formula for intensity of current transient,

$$I(t) = \frac{I_0\Theta(t)}{2}\left\{1 + \mathrm{erf}\left[\frac{\tau_0 - \zeta(t)}{2\xi^{1/2}(t)}\right]\right\}, \tag{21}$$

where $\mathrm{erf}(\ldots)$ is the error function. The initial current decay and the effective carrier transit time τ_e, corresponding approximately to the transition to faster final current decay, are given by

$$I(t) \approx I_0\Theta(t)\,, \quad t < \tau_e, \tag{22}$$

$$\zeta(\tau_e) = \tau_0. \tag{23}$$

From Eqs. (15)-(17) as well as Eqs. (22)-(23) it follows that the effective mobility of carrier packet, determined by the trapping/detrapping events, is given by $\mu_{eff}(t) = \mu_0\Theta(t)$. The concept of effective carrier mobility in the case of dispersive transport was introduced in [4, 5] and the present study confirms its validity.

5. Functions $\bar{z}(t)$ and $\sigma(t)$ for model trap distributions

In this section the formulas, determining the 'centroid', $\bar{z}(t)$, and the RMS dispersion, $\sigma(t)$, of carrier packet for some model distributions of traps are given. These functions almost completely characterize the carrier transport in the initial time interval, $t \le \tau_e$. In particular, the $\bar{z}(t)$ function determines the form of current transient for $t < \tau_e$, as well as the transit time τ_e (see Eqs. (6)-(7)).

The mentioned functions are calculated for the exponential distribution,

$$N_t(\varepsilon) = \frac{N_{tot}}{kT_c}\exp\left(-\frac{\varepsilon - \varepsilon_t^0}{kT_c}\right), \quad \varepsilon \ge \varepsilon_t^0, \tag{24}$$

and for the special case of Gaussian distribution,

$$N_t(\varepsilon) = \frac{2N_{tot}}{\pi^{1/2}kT_c}\exp\left[-\left(\frac{\varepsilon - \varepsilon_t^0}{kT_c}\right)^2\right], \quad \varepsilon \ge \varepsilon_t^0. \tag{25}$$

In both expressions N_{tot} stands for the total density of traps, the characteristic temperature T_c determines the rate of trap density decrease with energy and ε_t^0 denotes the lower limit of trap distribution ($N_t(\varepsilon) = 0$ for $\varepsilon < \varepsilon_t^0$). The 'cut-off' in the trap density is introduced for calculational convenience in the numerical simulations of carrier transport. The simulation results are presented in the next section. The majority of formulas is derived under the assumptions that $t \gg \tau_r^0$ (with $\tau_r^0 = \tau_r\left(\varepsilon_t^0\right)$) and, in the case of approximate formulas, that $\Theta\left(t\right) \ll 1$.

For exponential trap distribution (24) and dispersive transport regime, corresponding to the value of parameter $\alpha = T/T_c < 1$, the functions $\bar{z}\left(t\right)$ and $\sigma\left(t\right)$ may be calculated exactly. This can be done, making use of the general solutions of MT equations in terms of Laplace transforms [10, 12], as well as of some results given in [1]. The obtained formulas have the form:

$$\bar{z}\left(t\right) = \frac{\tau_t}{\Gamma^2\left(1+\alpha\right)\Gamma\left(1-\alpha\right)}\left(\frac{t}{\tau_r^0}\right)^{\alpha}, \quad \alpha < 1, \tag{26}$$

$$\sigma\left(t\right) = \left[\frac{2}{\Gamma\left(1+2\alpha\right)} - \frac{1}{\Gamma^2\left(1+\alpha\right)}\right]^{1/2}\frac{\tau_t}{\Gamma\left(1+\alpha\right)\Gamma\left(1-\alpha\right)}\left(\frac{t}{\tau_r^0}\right)^{\alpha}, \quad \alpha < 1, \tag{27}$$

where $\tau_t = 1/C_t N_{tot}$ denotes the mean trapping time of free carriers and $\Gamma\left(\ldots\right)$ is the Euler gamma function. On the other hand, the functions $\bar{z}\left(t\right)$ and $\sigma\left(t\right)$, calculated from Eqs. (19)-(20), are as follows:

$$\bar{z}\left(t\right) \approx \frac{\left(1-\alpha\right)\tau_t}{1.8\alpha^2}\left(\frac{1.8t}{\tau_r^0}\right)^{\alpha}, \quad \alpha < 1, \tag{28}$$

$$\sigma\left(t\right) \approx \left[\frac{\left(1-\alpha\right)^3}{1.8\alpha^3\left(2-\alpha\right)}\right]^{1/2}\tau_t\left(\frac{1.8t}{\tau_r^0}\right)^{\alpha}, \quad 0.5 < \alpha < 1. \tag{29}$$

Therefore, the exact and approximate expressions for $\bar{z}\left(t\right)$ and $\sigma\left(t\right)$ differ solely in multiplicative coefficients, which proves to some extent the correctness of present approach. It is seen that the velocity of carrier packet ‘centroid’, $\mathrm{d}\bar{z}\left(t\right)/\mathrm{d}t$, decreases in time, which is the characteristic feature of dispersive transport.

For exponential distribution of traps with $\alpha > 1$ the carrier transport regime is commonly regarded as Gaussian. In this case the exact formulas for $\bar{z}\left(t\right)$ and $\sigma\left(t\right)$ probably cannot be obtained. The approximate formulas have the form:

$$\bar{z}\left(t\right) \approx \frac{\left(\alpha-1\right)\tau_t t}{\alpha\tau_r^0}, \quad \alpha > 1, \tag{30}$$

$$\sigma\left(t\right) \approx \left[\frac{\left(\alpha-1\right)^3}{0.9\left(2-\alpha\right)\left(3-\alpha\right)}\right]^{1/2}\frac{\tau_t}{\alpha}\left(\frac{1.8t}{\tau_r^0}\right)^{\left(3-\alpha\right)/2}, \quad 1 < \alpha < 2, \tag{31}$$

$$\sigma\left(t\right) \approx \left[\frac{2\left(\alpha-1\right)^3}{\left(\alpha-2\right)}\right]^{1/2}\frac{\tau_t}{\alpha}\left(\frac{t}{\tau_r^0}\right)^{1/2}, \quad \alpha > 2. \tag{32}$$

Since the velocity of the carrier packet ‘centroid’ is constant, the carrier transport may be in fact considered Gaussian. However, for $1 < \alpha < 2$ the dispersion of carrier packet increases faster than the square root of time, contrary to the case of pure Gaussian transport.

For Gaussian trap distribution (25), the formulas determining considered functions are:

$$\bar{z}(t) \approx \frac{\exp\left(-1/4\alpha^2\right)\tau_t t}{\left[1+\operatorname{erf}(1/2\alpha)\right]\tau_r^0}, \tag{33}$$

$$\sigma(t) \approx \frac{\exp\left(1/8\alpha^2\right)\left\{2\left[1+\operatorname{erf}(1/\alpha)\right]\right\}^{1/2}\tau_t}{\left[1+\operatorname{erf}(1/2\alpha)\right]^{3/2}}\left(\frac{t}{\tau_r^0}\right)^{1/2}, \tag{34}$$

where the parameter $\alpha = T/T_c$ and $\operatorname{erf}(\ldots)$ is the error function. These formulas are derived under more restrictive assumption than before, that is $t \gg \exp\left[(1+\alpha)/\alpha^2\right]\tau_r^0$. The carrier transport in this time region is Gaussian for arbitrary value of α. However, if the density of traps decays slowly with energy, so that $\alpha < 1$, the time of carrier thermalization might be very long. In this case gradual transition from dispersive to Gaussian transport regime should occur. The idea of such transition was primarily introduced in [13].

6. Numerical results

In order to verify the accuracy of formulas determining current transients and related quantities, Monte Carlo simulations of MT carrier transport are performed. The utilized procedure is similar to that described in [14]. In the following figures numerical results (denoted by points) are compared with analytical results (denoted by lines). In calculations the exact formulas (11)-(12) for the functions $\Theta(t)$ and $\tau_s(t)$ are used. The integrals (17)-(18), determining the functions $\zeta(t)$ and $\xi(t)$, are computed numerically.

Figs. 1 and 2 show several current transients, obtained for exponential (24) and Gaussian (25) trap distributions, respectively. The arrows mark the effective transit time τ_e, calculated from Eq. (23). As expected, for both distributions the accuracy of analytical solutions improves with the increase of parameter α. In the time interval $t < \tau_e$ the relative difference between current intensities, calculated numerically and analytically for smaller values of α, remains constant for exponential distribution and decreases for Gaussian one. In the time region $t \geq \tau_e$ the accuracy of analytical results is better for the Gaussian distribution. This is explained by the fact that in the case of exponential distribution the ratio of carrier densities, captured in the energy intervals $\varepsilon < \varepsilon_0(t)$ and $\varepsilon > \varepsilon_0(t)$ (with $\varepsilon_0(t) - \varepsilon_t^0 \gg kT$), is constant and carrier transport still remains dispersive. In the case of Gaussian distribution the mentioned ratio increases in time and the transition from dispersive to Gaussian transport regime takes place.

Fig. 3 presents the time evolution of the total carrier density, $n_{tot}(z,t) = n(z,t) + n_t(z,t)$, in the sample for exponential trap distribution with $\alpha = 0.75$. In this case the spatial carrier distribution, computed numerically, differs remarkably from Gaussian distribution, given by Eqs. (15)-(16). Nevertheless, the positions of ‘centroids’ of both distributions are nearly the same, which explains the similarities of current transients, calculated analytically and numerically for $t < \tau_e$.

Fig. 4 shows the trapped carrier densities per energy unit, $n_t'(z,t,\varepsilon)$, averaged over sample thickness, for exponential distribution of traps with $\alpha = 0.75$. One should expect that the energetic distribution of trapped carriers is given by the relationships: $\overline{n_t'}(t,\varepsilon) \propto N_t(\varepsilon)\,\tau_r(\varepsilon)$ for $\varepsilon < \varepsilon_0(t)$ and $\overline{n_t'}(t,\varepsilon) \propto N_t(\varepsilon)$ for $\varepsilon > \varepsilon_0(t)$. In the case of exponential trap distribution the relationships take the form of: $\overline{n_t'}(t,\varepsilon) \propto \exp(\varepsilon/kT - \varepsilon/kT_c)$ for $\varepsilon < \varepsilon_0(t)$ and $\overline{n_t'}(t,\varepsilon) \propto \exp(-\varepsilon/kT_c)$ for $\varepsilon > \varepsilon_0(t)$. In figure the sloping full and dashed lines correspond respectively to these relationships, whereas the horizontal lines mark the position of demarcation level $\varepsilon_0(t)$. It is seen that the energetic distribution of trapped carriers, computed numerically, is in good agreement with above predictions.

7. Conclusions and final remarks

In this paper, approximate description of the current transients, registered by the TOF method, is given for the case of MT quasi-equilibrium carrier transport. The obtained formulas are verified by comparison with some exact formulas, as well as with results of simulations of MT transport for model trap distributions. The agreement is quite satisfactory, provided that trap density decreases sufficiently fast with energy.

The main aim of TOF measurements in disordered solids is the determination of energetic distribution of traps and their parameters. Several methods of analysis of experimental data may be used for this purpose (see, for example, the review [15]). Basing on results obtained in present paper, yet another method can be proposed. Making use of Eqs. (8), (13) and (22) and assuming that $\Theta(t) \ll 1$, one obtains the formula:

$$\frac{\mathrm{d}}{\mathrm{d}t}\left[\frac{Q_0}{I(t)}\right] \approx 1.8\tau_0 C_t N_t\left[\varepsilon_0(t)\right], \quad t < \tau_e. \tag{35}$$

Here, Q_0 is the total charge generated in the sample, equal to the area under the curve $I(t)$ *versus* t. The above formula enables us to calculate the function $\mu_0^{-1} C_t N_t[\varepsilon_0(t)]$. The value of frequency factor ν_0, necessary for determination of demarcation energy $\varepsilon_0(t)$ (see Eq. (8)), may be found from the TOF measurements at several temperatures. For proper value of ν_0, all calculated trap distributions should coincide, provided that the temperature dependencies of μ_0, C_t and ν_0 are negligible.

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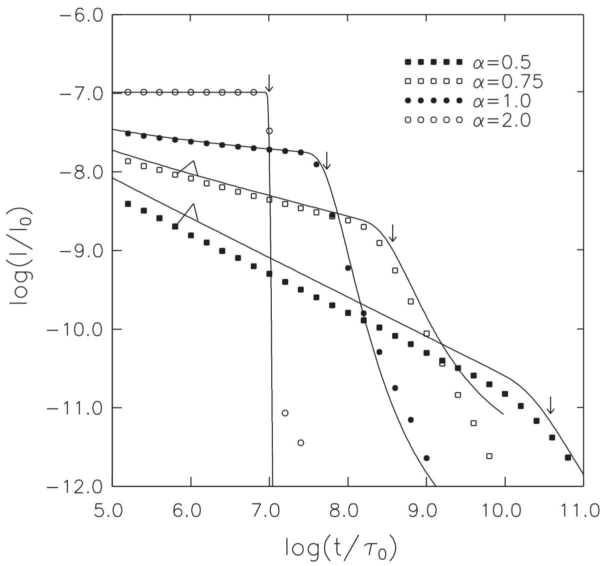

Figure 1: Current transients, calculated for exponential trap distribution (24) and several values of $\alpha = T/T_c$. Other calculation parameters: $\tau_0/\tau_t = 10^4$, $\nu_0\tau_0 = 10^6$, $\varepsilon_t^0/kT = 20$. Points and lines denote numerical and analytical results, respectively, whereas arrows indicate effective transit times τ_e.

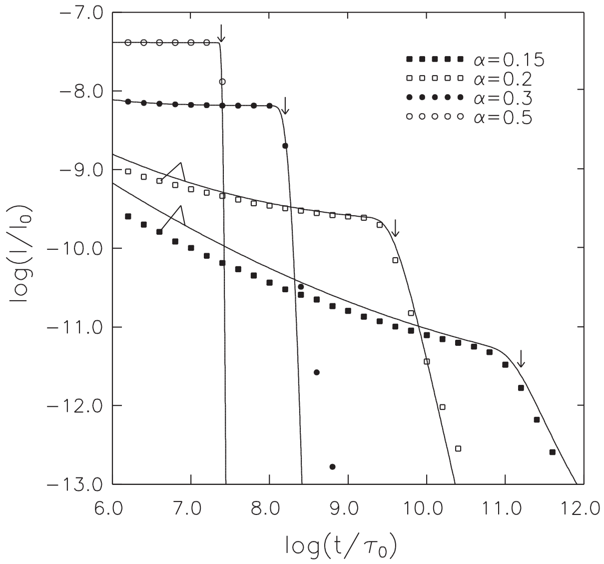

Figure 2: Current transients, computed for Gaussian trap distribution (25) and several values of $\alpha = T/T_c$. Other calculation parameters: $\tau_0/\tau_t = 10^4$, $\nu_0\tau_0 = 10^6$, $\varepsilon_t^0/kT = 20$. The notations are as in Fig. 1.

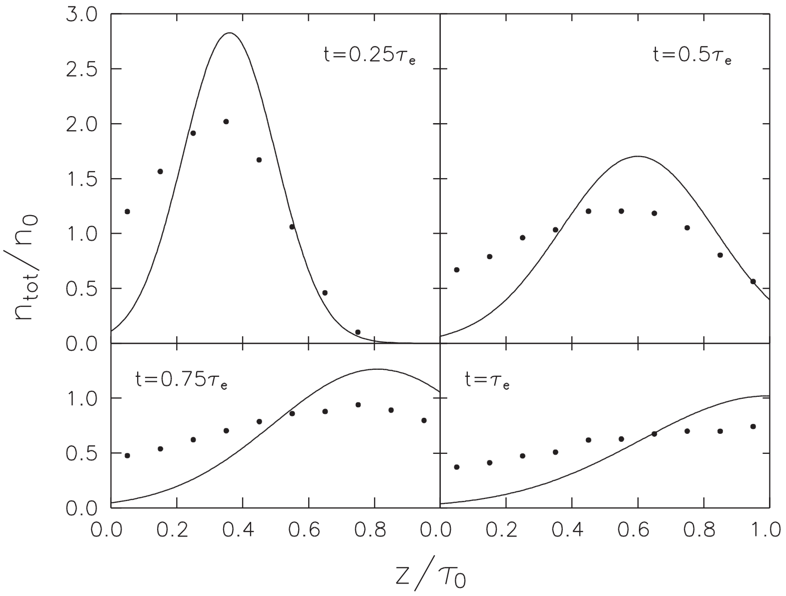

Figure 3: Spatial carrier distribution for several times, calculated for exponential trap distribution (24) with $\alpha = 0.75$. The values of remaining parameters are as in Fig. 1. Points and lines mark numerical and analytical results, respectively.

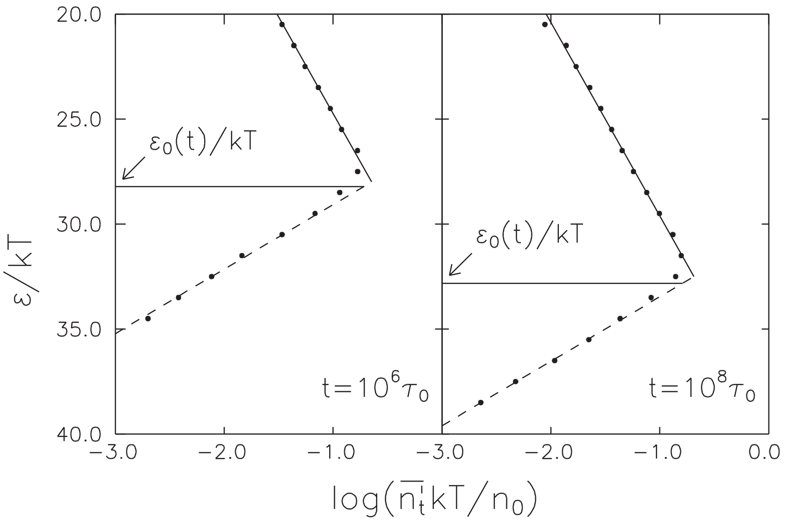

Figure 4: Energetic distribution of carriers for two times, computed for exponential trap distribution (24) with $\alpha = 0.75$. The values of remaining parameters are as in Fig. 1. Points and lines denote numerical and analytical results, respectively. Sloping full and dashed lines refer, respectively, to quasi-equlibrium and non-equilibrium carrier distributions.