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Low-temperature relaxations of charge carriers in disordered hopping systems

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Abstract. Energetic relaxation, drift, diffusion and recombination of charge carriers are considered at low temperatures in disordered hopping systems. It is shown that, due to its scaling form, an exponential energetic distribution of the density of localized states (DOS) generates very specific transport characteristics which cannot be obtained for other types of DOS functions. In contrast with other distributions, only an exponential DOS function allows (i) the introduction of a low-temperature analogue of the Einstein relation between carrier mobility and diffusivity for some initial time domain of relaxation and (ii) the description of carrier drift in strong electric fields in terms of a field-dependent effective temperature.

1. Introduction

In the low-temperature limit ($T \rightarrow 0$) carrier jumps to deeper states represent the only possible mode of charge transport. In the absence of an electric field, this impedes carrier equilibration within an energetically disordered system of hopping sites. In other words, low-temperature carrier diffusion and recombination are always non-equilibrium (dispersive) processes. The presence of an electric field does not abolish the necessity to jump down in energy, but a carrier can now move to a shallower state with respect to the band edge by executing a jump along the field. Such jumps become dominant if the energetic distribution of the density of states (DOS) decreases sharply with energy and if most carriers already occupy states close to the edge of this distribution. Under such conditions an equilibrium transport regime can be established even at $T = 0$. This means that applying an electric field can radically change characteristics of low-temperature drift and diffusion.

An analysis of many important electronic processes, such as carrier generation and geminate recombination, requires a self-consistent consideration of both drift and diffusion of carriers [1–3]. An important point of the analysis is the question of what kind of relation between the mobility μ and the diffusivity D can be used under the given conditions. In general, the impossibility of a quasi-equilibrium energetic distribution of carriers at low temperatures makes this question irrelevant: processes of carrier drift and diffusion must be considered independently without assuming any relationship between μ and D. Moreover, introducing these kinetic coefficients under non-equilibrium conditions becomes questionable since the dispersive drift and diffusion are described by equations different from the Fokker–Planck equation for the quasi-equilibrium transport regime [1, 2]. Some low-temperature analogue of the Einstein relation between μ and D may, nevertheless, be

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used [4–8] if both the carrier packet mean position $\langle x\rangle$ and its dispersion $\sigma = [\langle \Delta x^2\rangle]$ have similar time dependencies and if the former is proportional to the electric field strength F. As long as an equilibrium carrier diffusion is impossible at low temperatures, the above conditions can be fulfilled only under the non-equilibrium transport regime. Thus, the questions one should address for the case of low-temperature transport, are: (i) can a dispersive drift that is linear with respect to the field exist at low temperatures and, if so, (ii) what conditions limit its existence?

In the present paper low-temperature energetic relaxation, drift, diffusion, and recombination of excess carriers within a disordered hopping system are considered. It is shown that applying an electric field is equivalent to increasing the temperature as far as the energetic distribution of localized carriers is concerned [9–11]. The field-dependent effective temperature T_F is calculated for an exponential DOS function. Time dependences of the functions $\langle x\rangle(t)$ and $\sigma(t)$ are found to be similar within some initial time domain of relaxation. It is also proven that the low-temperature carrier drift velocity reveals a non-linear dependence upon the field even at weak fields, but that an initial time domain of linear drift can be distinguished.

2. Energetic relaxation and diffusion of carriers in the absence of electric field

Under non-equilibrium conditions a carrier occupies a particular localized state at a given time t if the probability of making a jump into a deeper state has remained low up to t. The probability w for the carrier to remain until the time t in the initial state separated from the nearest deeper neighbour by the distance r is given by the Poisson distribution

$$w(r,t) = \exp[-\nu_0 t \exp(-2\gamma r)] \tag{1}$$

where ν_0 is the attempt-to-jump frequency and γ is the inverse localization radius. The probability density W of finding the nearest deeper neighbour for a state with the energy E at the distance r is again determined by the Poisson formula:

$$W(E,r) = \exp[-n(E,r)]\frac{\mathrm{d}n(E,r)}{\mathrm{d}r} \tag{2}$$

where $n(E,r)$ is the density of localized states with energies exceeding E within the sphere of radius r. (In this paper the energy scale is chosen so that deeper states have higher energies.) Combining equations (1) and (2) and integrating over r yields the probability $f(E,t)$ for a localized state with the energy E to be occupied at time t as a result of a sequence of uncorrelated relaxation steps:

$$f(E,t) = \int_0^\infty \mathrm{d}r\ \frac{\mathrm{d}n(E,r)}{\mathrm{d}r}\exp[-n(E,r)]\exp[-\nu_0 t\exp(-2\gamma r)]. \tag{3}$$

The function $f(E,t)$ plays the role of a non-equilibrium distribution function for low-temperature carrier relaxation. Unlike the equilibrium Fermi–Dirac or Boltzmann distributions, it strongly depends upon the DOS function in a particular material.

The last exponential term in the integrand of equation (3) represents a very steep function of the variable r around $r = r_j$:

$$\begin{aligned} &\exp[-\nu_0 t\exp(-2\gamma r)] \cong 0 \qquad r < r_j(t) \\ &\exp[-\nu_0 t\exp(-2\gamma r)] \cong 1 \qquad r > r_j(t) \end{aligned} \qquad r_j(t) = (1/2\gamma)\ln(\nu_0 t) \tag{4}$$

that allows one to reduce equation (3) to [8]

$$f(E,t) = \exp\left\{-n\left[E, \frac{1}{2\gamma}\ln(\nu_0 t)\right]\right\}. \tag{5}$$

Equations (4) and (5) imply that at a given time t carriers occupy those localized states which have no neighbours with energies $E' > E$ at a distance shorter than $r_j(t)$. This allows the designation of such states as 'currently metastable states'. In the absence of an electric field the function $n(E, r)$ takes the form

$$n(E, r) = \frac{4\pi r^3}{3} \int_E^\infty \mathrm{d}E'\ g(E') \tag{6}$$

where $g(E)$ is the usual density-of-states function. The distribution function described by equation (5) then reduces to

$$f(E, t) = \exp\left[-\frac{\pi}{6\gamma^3}\,[\ln(\nu_0 t)]^3 \int_E^\infty \mathrm{d}E'\ g(E')\right]. \tag{7}$$

Like the Fermi–Dirac distribution, the function $f(E, t)$ reveals a rather sharp edge at some demarcation energy $E_d(t)$ determined by the condition

$$\frac{\pi}{6\gamma^3}\,[\ln(\nu_0 t)]^3 \int_{E_d(t)}^\infty \mathrm{d}E\ g(E) = 1. \tag{8}$$

For an exponential DOS function with the total density of localized states

$$N_t = \int_0^\infty \mathrm{d}E\ g(E)$$

and the characteristic energy of the distribution E_0

$$g(E) = \frac{N_t}{E_0} \exp\left(-\frac{E}{E_0}\right) \tag{9}$$

equation (8) yields [12]

$$E_d(t) = 3E_0 \ln\left[\left(\frac{\pi N_t}{6\gamma^3}\right)^{1/3} \ln(\nu_0 t)\right]. \tag{10}$$

Energetic relaxation of carriers in a hopping system is coupled to the spreading of their spatial distribution. The time dependence of the carrier packet dispersion can be estimated as

$$\frac{\mathrm{d}\sigma(t)}{\mathrm{d}t} = \frac{1}{3}\nu_j(t)\left[r_j(t)\right]^2 = \frac{\nu_0}{3}\exp\left[-2\gamma r_j(t)\right]\left[r_j(t)\right]^2 \tag{11}$$

where r_j and ν_j are the characteristic jump distance and the characteristic frequency of jumps at time t, respectively. Using equation (4) in equation (11) then yields

$$\frac{\mathrm{d}\sigma(t)}{\mathrm{d}t} = \frac{1}{12\gamma^2 t}\,[\ln(\nu_0 t)]^2. \tag{12}$$

Integrating equation (12) gives a universal function $\sigma(t)$:

$$\sigma(t) = \frac{1}{36\gamma^2}\,[\ln(\nu_0 t)]^3 \tag{13}$$

which depends neither upon a particular choice of the DOS distribution nor upon the total density of localized states. This universality follows from prescribed time dependencies for both the jump distance and the jump rate at low temperatures since a carrier makes a jump as soon as it finds an energetically accessible site at a distance less than $r_j(t)$ [5, 13, 14]. Note that the above results are valid for times

$$\nu_0 t > 1 \tag{14}$$

longer than the average dwell time for the first carrier jump after excitation at $t = 0$ that allows one to introduce the demarcation energy—see equation (10).

3. Low-temperature carrier recombination

The results obtained above for the distribution function can easily be extended to describing kinetics of monomolecular carrier recombination at low temperatures. Indeed, monomolecular recombination is equivalent to carrier trapping by localized states which are deeper than any states contributing to the hopping transport. Under these conditions the distribution function given by equation (7) takes the form

$$f_R(E,t) = \exp\left[-\frac{\pi}{6\gamma^3}\,[\ln(\nu_0 t)]^3\left(N_R + \int_E^\infty \mathrm{d}E'\ g(E')\right)\right] \tag{15}$$

where N_R is the concentration of recombination centres. The density of carriers, $p(t)$, which survive until the time t can be calculated as the total density of carriers occupying the hopping sites but not recombination centres:

$$\begin{aligned} p(t) &= p_0\int_0^\infty \mathrm{d}E\ g(E)f_R(E,t)\Big/\left(\int_0^\infty \mathrm{d}E\ \left[g(E) + N_R\delta(E-E_R)\right]f_R(E,t)\right) \\ &= p_0\int_0^\infty \mathrm{d}E\ g(E)f_R(E,t)\Big/\left(N_R f_R(\infty,t) + \int_0^\infty \mathrm{d}E\ g(E)f_R(E,t)\right) \\ &= p_0\int_0^\infty \mathrm{d}E\ g(E)f(E,t)\Big/\left(N_R + \int_0^\infty \mathrm{d}E\ g(E)f(E,t)\right) \end{aligned} \tag{16}$$

where p_0 is the initial density of carriers, E_R the energy of recombination centres ($E_R \to \infty$), and δ is the Dirac delta function. Substituting equation (7) into equation (16) and integrating under the condition

$$\frac{\pi N_t}{6\gamma^3}\,[\ln(\nu_0 t)]^3 \gg 1$$

yields

$$p(t) = p_0\Big/\left(1 + \frac{\pi N_R}{6\gamma^3}\,[\ln(\nu_0 t)]^3\right). \tag{17}$$

One should remember that equation (17) describes relaxation of carriers excited at $t = 0$. Since monomolecular recombination represents a linear process, this equation can be considered as a Green function of the problem. Therefore, one could try to use equation (17) to obtain a solution for the steady-state carrier density p_{st} corresponding to the constant generation rate G_0:

$$p_{st} = \frac{G_0}{p_0}\int_0^t \mathrm{d}t'\ p(t-t') \qquad t \to \infty. \tag{18}$$

However, substituting equation (17) into equation (18) leads to a divergence in the integral and, concomitantly, to an infinitely high steady-state density of carriers. This problem is solved if one remembers that the slow decrease of $p(t)$ is due to carriers localized in relatively deep states which are separated from both nearest accessible hopping neighbours and nearest recombination centres by rather long distances. The total number of such states is limited and sooner or later they will be completely filled. The effect of trap filling prevents new carriers from being localized in isolated states and leads to a higher average recombination rate. On the other hand, this effect makes the problem non-linear and restricts the time domain within which equation (18) is valid. Thus, the filling of deep localized states must be necessarily taken into account when considering the steady-state photoconductivity and photoluminescence at low temperatures.

4. Field-assisted energetic relaxation and drift of carriers at low temperatures

The application of an electric field, F, changes the total density of localized states, $n(E,r)$, accessible for a downward carrier jump from a localized state with the energy E over a distance equal to or less than r:

$$n(E,r) = 2\pi \int_0^r \mathrm{d}r'\ r'^2 \int_0^\pi \mathrm{d}\vartheta\ \sin\vartheta \int_{E-eFr'\cos\vartheta}^\infty \mathrm{d}E'\ g(E') \tag{19}$$

where e is the elementary charge and ϑ the polar angle between the vectors $\boldsymbol{F}$ and $\boldsymbol{r}$. Substituting equation (19) into equation (5) yields the time-dependent energetic distribution function $f_F(E,t)$ of localized carriers in the presence of the electric field:

$$f_F(E,t) = \exp\left[-2\pi \int_0^{(1/2\gamma)\ln(\nu_0 t)} \mathrm{d}r\ r^2 \int_{-1}^1 \mathrm{d}x \int_{E-eFrx}^\infty \mathrm{d}E'\ g(E')\right]. \tag{20}$$

At weak electric fields the function $f_F(E,t)$ given by equation (20) reduces to the zero-field distribution function—see equation (7). For strong fields, jump distances close to r_j in the field direction give the principal contribution to the integrand in equation (20). The problem is then effectively reduced from three to one spatial dimension, such that one can write

$$f_F(E,t) = \exp\left[-\frac{\pi}{\gamma}\left(\frac{E_0}{eF}\right)^2 \ln(\nu_0 t) \int_{E-eFr_j(t)}^\infty \mathrm{d}E\ g(E)\right] \qquad \frac{eF}{2\gamma E_0}\ln(\nu_0 t) \gg 1 \tag{21}$$

where E_0 is the characteristic energy of the DOS distribution. An interesting feature of equation (21) is that the electric field F cannot be characterized as weak or strong irrespective of a time-scale since, in the most important terms, the field and time are linked in a universal parameter

$$\frac{eF}{2\gamma E_0}\ln(\nu_0 t).$$

In particular, any weak-field approximation for the distribution function is valid only within a restricted time domain.

To illustrate the field dependence of $f_F(E,t)$ we again employ an exponential DOS distribution. Substituting equation (9) into equation (21) and integrating yields

$$f_F(E,t) = \exp\left\{-\frac{\pi N_t}{\gamma}\left(\frac{E_0}{eF}\right)^2 \ln(\nu_0 t)\exp\left(\frac{eF}{2\gamma E_0}\ln(\nu_0 t)\right)\exp\left(-\frac{E}{E_0}\right)\right\}. \tag{22}$$

The distribution function given by equation (22) is also characterized by a demarcation energy $E_d(t)$ [8]:

$$E_d(t) = \frac{eF}{2\gamma}\ln(\nu_0 t) + E_0 \ln\left[\frac{\pi N_t}{\gamma}\left(\frac{E_0}{eF}\right)^2 \ln(\nu_0 t)\right] \cong \frac{eF}{2\gamma}\ln(\nu_0 t). \tag{23}$$

A logarithmic time dependence of the demarcation energy is well known for either trap-controlled [15–17] or hopping [12, 14] dispersive transport at weak electric fields and at finite temperatures T:

$$E_d(t) = kT\ln(\nu_0 t). \tag{24}$$

Comparing equations (23) and (24) shows that the value $eF/2\gamma$ plays the role of an effective temperature T_{eff} of the energetic distribution of localized carriers:

$$T_{eff} = \frac{eF}{2\gamma k}. \tag{25}$$

Under the dispersive transport regime, field-assisted jumps lead to higher jump rates and, correspondingly, to faster diffusion of localized carriers to deeper states independently of the particular form of the DOS function. It seems, however, an exceptional property of an exponential DOS distribution that this process can be described in terms of the effective temperature. Substituting other DOS functions into equation (21) yields field dependencies of the localized-carrier distribution function which cannot be simply described in terms of an effective temperature. Of course, any smoothly decreasing energetic distribution of states can be approximated by an appropriate exponential DOS function within a limited region of energies. Concomitantly, the effective-temperature approximation can be valid within some initial time domain of carrier kinetics even if the DOS is not exponential, but it then fails to describe the whole relaxation process [18]. Moreover, it was recently shown [11] that even where the concept works, different T_{eff}-functions are obtained from different experimental situations.

Within the initial time domain of the 'weak electric field'

$$\frac{eF}{2\gamma E_0}\ln(\nu_0 t) \ll 1 \tag{26}$$

the characteristic jump distance $\langle r_j\rangle$ cannot be significantly affected by the field, and average displacement along the field per jump $\langle x_j\rangle$ occurs due to a higher probability of finding the nearest neighbour in that direction. For a carrier jump from a localized state with the energy E, the value $\langle x_j\rangle(E,t)$ may be estimated as

$$\begin{aligned}\langle x_j\rangle(E,t) &= \langle r_j\rangle(t)\left[4\pi\int_E^\infty \mathrm{d}E'\ g(E')\right]^{-1} 2\pi\int_0^\pi \mathrm{d}\vartheta\ \sin\vartheta\cos\vartheta\int_{E-eF\langle r_j\rangle\cos\vartheta}^\infty \mathrm{d}E'\ g(E') \\ &\cong \langle r_j\rangle(t)\left[2\int_E^\infty \mathrm{d}E'\ g(E')\right]^{-1}\int_{-1}^1 \mathrm{d}x\ x\left[\int_E^\infty \mathrm{d}E'\ g(E') + eF\langle r_j\rangle(t)xg(E)\right] \\ &= \frac{1}{3}eF\left[\langle r_j\rangle(t)\right]^2 g(E)\left[\int_E^\infty \mathrm{d}E'\ g(E')\right]^{-1}. \end{aligned}\tag{27}$$

For an exponential DOS function, equation (27) gives the energy-independent average displacement

$$\langle x_j\rangle(t) = \frac{1}{12}\frac{eF}{E_0\gamma^2}\left[\ln(\nu_0 t)\right]^2. \tag{28}$$

The average carrier drift velocity $\langle v\rangle$ may be calculated from the average displacement and the jump rate as

$$\langle v\rangle(t) = \langle x_j\rangle(t)\langle \nu_j\rangle(t) = \frac{1}{12}\frac{eF}{E_0\gamma^2}\frac{1}{t}\left[\ln(\nu_0 t)\right]^2. \tag{29}$$

Integrating equation (29) yields the time dependence of the carrier packet mean position in the form

$$\langle x\rangle(t) = \frac{1}{36}\frac{eF}{E_0\gamma^2}\left[\ln(\nu_0 t)\right]^3. \tag{30}$$

The function $\langle x\rangle(t)$ given by equation (30) is proportional to the field F and has the same time dependence as the zero-field carrier packet dispersion $\sigma(t)$ defined by equation (13). In this case the ratio $\sigma F/\langle x\rangle$ can be considered as a generalization of the Einstein relation to the non-equilibrium transport regime. Dividing equation (13) by equation (30) yields

$$\frac{\sigma F}{\langle x\rangle} = \frac{E_0}{e}. \tag{31}$$

Equation (31) shows that the characteristic energy E_0 of the DOS function appears in the low-temperature non-equilibrium Einstein relation instead of the characteristic thermal energy kT [5–7]. In reference [7], the relation was derived from arguments in the energy domain. The following cautionary notes are relevant to equation (31).

(i) This equation is obtained for an exponential DOS function. Using another energetic distribution of localized states leads to different time dependences of the functions $\sigma(t)$ and $\langle x\rangle(t)$, even within the 'weak-field' time domain, which makes any generalization of the Einstein relation for low-temperature conditions impossible.

(ii) Validity of the linear-field approximation for the low-temperature carrier drift is restricted by equation (26) which, therefore, also restricts the time domain within which equation (31) can be used.

A sufficiently strong electric field enhances carrier jumps to rather shallow localized states. In this case a carrier, making a jump from a 'currently metastable state', will on average execute a relatively long series of jumps before it finds another state which is also still metastable at time t. Since carrier jumps along the field represent the dominant mode of carrier transport at strong fields, the total distance r_F covered by a carrier in the course of such 'multiple hopping' between metastable states can be found from the condition

$$2\pi\rho^2 r_F \int_{E_d(t)}^{\infty} \mathrm{d}E\ g(E) = 1 \tag{32}$$

where ρ is the effective radius of a cylinder within which a carrier executes a random walk, mainly in the field direction. To estimate the radius ρ, it is assumed that the field is not strong enough to excite carriers to energies above the distribution of localized states. Under these conditions the average jump distance during the multiple hopping, r_{mh}, can be estimated from the balance between carrier jumps to shallower and to deeper states. Such an estimation yields $r_{mh} = E_0/eF$, where E_0 is the characteristic energy of the DOS function. Substituting $\rho = r_{mh}$ into equation (32) gives

$$r_F(t) = \frac{1}{2\pi}\left(\frac{eF}{E_0}\right)^2 \left[\int_{E_d(t)}^{\infty} \mathrm{d}E\ g(E)\right]^{-1}. \tag{33}$$

For an exponential DOS function, equations (23) and (33) give

$$r_F(t) = \frac{\ln(\nu_0 t)}{2\gamma}(\nu_0 t)^{eF/2\gamma E_0}. \tag{34}$$

The characteristic frequency of multiple jumps still should be estimated as $\langle \nu_j\rangle \cong \nu_0 \exp(-2\gamma r_j) = 1/t$ since after the first jump from a currently metastable state over the distance r_j, a carrier executes a series of shorter jumps with higher jump frequencies. The carrier drift velocity can then be written in the form

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle x\rangle(t) = \frac{\nu_0 \ln(\nu_0 t)}{2\gamma}(\nu_0 t)^{eF/2\gamma E_0 - 1}. \tag{35}$$

Integrating equation (35) yields the average carrier drift displacement

$$\langle x\rangle(t) = \frac{E_0 \ln(\nu_0 t)}{eF}(\nu_0 t)^{eF/2\gamma E_0} \tag{36}$$

which has the form $\langle x\rangle \propto t^{\alpha}$ typical for the dispersive transport regime. For both trap-controlled and hopping modes of the carrier drift in weak electric fields at finite temperatures in materials with exponential DOS distributions the exponent α is given by $\alpha = kT/E_0$. This again leads to the conclusion that the strong-field carrier drift in hopping systems with

an exponential DOS function can be described by the effective temperature determined by equation (25). And again one should note that this is an exceptional property of exponential DOS distributions.

5. Conclusions

Non-equilibrium carrier transport at low temperatures cannot be characterized as of either weak-field or strong-field type without reference to the time-scale considered. At sufficiently long times, carrier drift reveals characteristic features of the strong-field transport for arbitrarily weak applied fields. For materials with exponential DOS distributions, and within an initial time region of 'weak-field transport', a non-equilibrium analogue of the Einstein relation between carrier mobility and diffusivity can be introduced with the characteristic energy of the distribution, E_0, playing the role of the thermal energy kT.

For strong electric fields, both the energetic distribution of localized carriers and the carrier drift mobility exhibit strong field dependences. For materials with exponential DOS functions the both dependences are described by the effective, field-dependent temperature

$$T_{eff} = \frac{eF}{2\gamma k}.$$

As long as the DOS distribution in a particular material can be approximated by an exponential function the concept of the effective temperature is applicable within some restricted time interval of carrier relaxation.

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