

**Energy relaxation in a tail at zero temperature in the hopping regime**

E. Haba, O. Bleibaum, and H. Böttger

*Institut für Theoretische Physik, Otto-von-Guericke Universität Magdeburg, PF 4120, 39016 Magdeburg, Germany*

V. V. Bryksin

*A. F. Ioffe Physico-Technical Institute, St. Petersburg, Politekhnicheskaya 26, 194021 St. Petersburg, Russia*

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We argue that energy relaxation of nonequilibrium charge carriers at zero temperature in the hopping regime is Miller-Abraham-like, that is, the charge carriers always jump to the next closest site. The Miller-Abraham-like motion of the charge carriers manifests itself in the moments of the energy distribution function. Due to the motion on Miller-Abraham paths the rate of energy relaxation is much smaller, as if the particles would relax in a percolation-like fashion. In contrast to percolation-like relaxation the dispersion of the transport coefficients in the Miller-Abraham regime depends truly on two parameters. A small rise of the temperature leads to a strong increase of the energy relaxation rate and the strength of the increase depends strongly on disorder.

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**I. INTRODUCTION**

Energy relaxation processes have been investigated in many disordered materials, in which transport proceeds by phonon-assisted hopping between localized states, e.g., in amorphous semiconductors and polymers (see, e.g., Refs. 1–5). In many experiments the excitations are produced either by illumination (see, e.g., Refs. 6 and 7) or by an electric field (see, e.g., Refs. 8 and 9). If the charge carriers are far from the Fermi energy and the temperatures are sufficiently low, jumps down in energy space are most important. In this case nearly every jump of a charge carrier is accompanied by a loss of energy, so that it seems to be reasonable to ignore jumps upward in energy space when modeling this situation. The energy-loss hopping model, which is based on this approximation, has proven to be successful in explaining the experimental data of time-resolved photoluminescence and transport experiments in many strongly disordered materials, including band tails of amorphous semiconductors, polycrystalline materials, and polymers (see, e.g., Refs. 1 and 10–15).

Despite the neglect of upward jumps the investigation of relaxation phenomena on the basis of the energy-loss hopping model is still very intricate. In this model every site is connected to a huge number of sites by transition probabilities ranging over many orders of magnitude. Since the configuration-averaged transition probabilities do not describe the situation adequately typical rates are often used.<sup>1</sup> The use of these rates is based on notions, which have been developed in the theory of hopping conduction by Mott in his investigation of the hopping conductivity in the variable-range hopping regime.<sup>16</sup>

In the theory of hopping conduction the assumptions underlying the introduction of typical rates have been clarified by Ambegaokar *et al.*,<sup>17</sup> by Shklovskii and Efros,<sup>18</sup> and by Pollak,<sup>19</sup> who have reformulated the problem in the language of percolation theory. While it was often pointed out that relaxation phenomena far from equilibrium might be different from conduction phenomena in the vicinity of the Fermi energy,<sup>1</sup> and that therefore the conventional procedures for

the investigation of transport problems in the vicinity of the Fermi energy might not be applicable to the nonequilibrium situation, the typical rates used in the investigations of relaxation processes were the same as those that could have also been obtained from percolation theory (see, e.g., the Refs. 1 and 14). That is, it was assumed that the characteristic hopping length is of the order of the average site distance, so that the typical transition probability for a jump of an electron from a site with site energy  $\epsilon$  down in energy space is proportional to  $\exp[-\rho_c(\epsilon)]$ , where  $\rho_c(\epsilon)$  is of the order of the ratio between the average spacing between accessible sites and the localization length, which in the hopping regime is assumed to be large. This notion has also been supported further by the results of our effective-medium theory.<sup>15</sup>

While at first glance the notion that the typical rates in the energy-loss hopping model agree with those from the percolation theory close to equilibrium seems to be plausible, its consequence—that the relaxation is determined by the same paths as those if the problem would be a percolation problem close to equilibrium—is surprising. Certainly, there is a substantial part of sites with neighbors within the average site spacing, but there is also a substantial part of distribution holes. Distribution holes are sites without accessible neighbors within the average site spacing. Since the transition probability is an exponentially small quantity with respect to the hopping length, we expect that a charge carrier prefers jumping to the next closest site. Doing so, it does not matter whether this site is a distribution hole or not. If the temperature is zero, as it is assumed to be in the energy-loss hopping model, the particle cannot return to its initial site. Accordingly, a particle, which has jumped into a distribution hole, can only leave the distribution hole by an extraordinarily hard jump, that is, a jump over a length which is large compared to the average site spacing. Therefore, we expect that the relaxation at large times is determined by jumps out of distribution holes.

In the percolation theory of the conductivity for small electric fields, such distribution holes are called dead ends. There they are dead since the resistors, which would lead out of them, are not taken into account in the percolation con-

struction, since they are large compared to the critical resistor. If the relaxation at large times is determined by jumps out of dead ends, as we suggest in our paper, then the relaxation paths are necessarily different from those which would be obtained if the conventional percolation theory for close-to-equilibrium problems were applied, since these paths are determined by jumps between pairs of sites not taken into account in the percolation construction for the conductivity close to equilibrium.

In the context of the hopping conductivity, dead ends were referred to in the paper by Miller and Abrahams<sup>20</sup> in their investigation of the hopping conductivity in the nearest-neighbor hopping regime. They argued that the electron would always jump to the next closest site. Doing so, its motion would inevitably be stopped in a dead end and thus the jump out of the dead end would determine the conductivity. Following this route they concluded that the conductivity

$$\sigma \propto \exp[-\rho_c^{d/(d-1)}]. \quad (1)$$

Here  $\rho_c$  is again the ratio between the average spacing between sites and the localization length, which is assumed to be large. Later it was recognized that Miller-Abraham paths are not the most effective current paths. More effective paths can be formed if dead ends are avoided. The most effective paths are those which avoid all dead ends. These are the percolation paths. A percolation path is different from a Miller-Abraham path since it does not contain jumps out of dead ends. The contributions from percolation paths to the conductivity by far exceed those from Miller-Abraham paths, since the contributions of the first are only proportional to  $\exp(-\rho_c)$  while the contributions of the latter are given by Eq. (1). Only in a very strong electric field, in which the charge carriers cannot return to their initial sites, have the Miller-Abraham assumptions been proven correct.<sup>18,21</sup> For energy relaxation the situation at zero temperature is very similar to that in a very strong electric field in that the motion is directed. On the other hand, in the case of the conduction in the presence of a very strong electric field the motion proceeds in position space, and in the case of energy relaxation in energy space. These two spaces are not the same. Therefore, it is not immediately clear whether for energy relaxation similar results as those for the conductivity in a very strong electric field hold.

It is the purpose of this paper to investigate whether the relaxation at zero temperature is Miller-Abraham-like or not, and to investigate its consequences. To this end we develop a simplified model for the investigation of Miller-Abrahams relaxation processes at zero temperature and compare the results of our theoretical calculations with results of numerical calculations, which do not use further assumptions. The impact of finite temperatures is studied numerically.

## II. BASIC EQUATIONS

We consider localized nonequilibrium charge carriers far from the Fermi energy. Such charge carriers can, e.g., be produced by illumination. We assume that the number of charge carriers is small, so that Fermi correlation is negli-

gible. The charge carriers lower their energy by jumps with phonon emission. Only jumps with phonon emission can occur, since the temperature is zero. In this case the transport can be described by the simple rate equation

$$\frac{dn_m(t)}{dt} = \sum_n [n_n(t)W_{nm} - n_m(t)W_{mn}], \quad (2)$$

where  $n_m(t)$  is the number of particles at site  $m$  with position vector  $\mathbf{R}_m$  and site energy  $\epsilon_m$  at an instant of time  $t$ , and

$$W_{nm} = \theta(\epsilon_m - \epsilon_n) \theta(\omega - \epsilon_m + \epsilon_n) \nu \exp(-2\alpha|\mathbf{R}_{nm}|) \quad (3)$$

is the transition probability for a hop from site  $n$  to site  $m$ . Here  $\alpha^{-1}$  is the localization radius,  $\nu$  is the attempt-to-escape frequency,  $\mathbf{R}_{nm} = \mathbf{R}_n - \mathbf{R}_m$ , and the energy axis is directed down in energy space. Both the positions of the sites and their site energies are random quantities. The sites are distributed randomly homogeneously in space, and the site energies are distributed statistically independently according to a distribution function proportional to the density of states.

In writing the expression (3) we have assumed that only a small amount of energy  $\omega$  is transferred from the electron to the phonon system in one hop. This fact manifests itself in the second step function in Eq. (3). If the electron-phonon coupling is treated as a constant this step function arises from the integration over all possible intermediate phonon states in the calculation of the transition probability. Since a phonon can maximally have an energy of the order of the Debye energy the energy  $\omega$  is equal to the Debye energy in this case, if only one process is taken into account. In an Anderson insulator, however, the assumption that the electron-phonon coupling constant is independent of the energy transferred in one jump, that is, independent of phonon momentum, does not hold. The electron-phonon coupling constant is proportional to the overlap integral between the electron wave functions and a phonon wave function, and to the Fourier-transformed potential, which couples the electron and the phonon wave electromagnetically. The overlap integral decreases rapidly if the magnitude  $q$  of the phonon wave vector becomes larger than twice the inverse localization radius, and thus renders phonons with high energy ineffective. The Fourier-transformed potential is usually treated as a constant, in line with the deformation-potential approximation. However, this approximation assumes that the electrons can move quickly, so that that they can keep the system neutral. In an Anderson insulator, however, there are no electrons that could move fast enough to screen out the electric field of the phonon wave on a time scale of the order of the inverse phonon frequency for most phonon frequencies. Only very long-wavelength phonons can be screened out in this way. Thus the electro-magnetic potential, which yields the coupling between the electron and the phonon wave, is of long range for most phonon frequencies, so that its Fourier transform rapidly drops to zero. Accordingly we conclude that the electron-phonon coupling constant rapidly drops to zero for phonons with an energy larger than a characteristic energy  $\omega$ . Therefore, the sum over the intermediate phonon states is effectively restricted to modes with energy small compared to the Debye energy so that the Debye energy is replaced by  $\omega$ .

One might argue that the high-energetic part of the phonon spectrum is localized, and that the coupling between the localized phonon modes and the electron is very strong, if the electron is placed on a site which carries a localized mode. However, we argue that the number of sites which do not carry localized modes is large compared to the number of sites with localized modes, so that it is unlikely that the electron can be found on such a site.

Due to the reasons discussed above we expect that the energy scale  $\omega$  is small so that an electron can change its energy by a multiple of the energy  $\omega$  during its relaxation. To assess whether this approximation is in conflict with the experimental data we refer the reader to the application of this approximation to amorphous hydrogenated silicon in Sec. VI. In the main part of our paper, however, we focus on relaxation in an unbounded constant density of states, in which the density of accessible sites only becomes finite due to the finiteness of  $\omega$ , and equal to  $\omega N$ . We believe that investigations on relaxation processes in such a density of states are relevant for many modern Anderson insulators, for which field effect measurements indicate that the density of states depends only weakly on energy (see, e.g., Refs. 22 and 23). We would like to stress that the use of Eq. (2) for the investigation of photoluminescence experiments presumes the existence of an observable low excitation generation regime, in which the kinetic of the excitations is independent of their density, that is, of the generation rate. Such a regime has been observed in numerous photoluminescence experiments (see, e.g., Refs. 24–26). The existence of the low rate excitation generation regime is thus experimentally verified.

In order to investigate the relaxation we focus on the moments of the energy distribution function  $F(\epsilon_0, \epsilon|t)$ , which is the probability to find an electron at a site with energy  $\epsilon$  at time  $t$ , if it was at a site with energy  $\epsilon_0$  at  $t=0$ . Of particular interest is the rate of energy relaxation

$$v(t) = \left\langle \sum_n \epsilon_n \frac{dn_n}{dt} \right\rangle, \quad (4)$$

which coincides with the derivative of the first moment of the energy distribution  $F$  function with respect to time, that is,

$$v(t) = \frac{d}{dt} \int_{\epsilon_0}^{\infty} d\epsilon \epsilon F(\epsilon_0, \epsilon|t), \quad (5)$$

the mean-squared deviation of the distribution function, and their asymmetry. In writing Eq. (5) we have assumed that the energy axis is directed down in energy space, so that the energy of the particles increases in the relaxation.

### III. THE ENERGY DISTRIBUTION FUNCTION

Since we expect that the transport is Miller-Abraham-like the question arises as to how the transport can be modeled analytically. That percolation theory cannot handle this situation is obvious. However, also, the effective-medium theories are not suitable to describe this situation. The reason for this is simply that in all of these methods it is assumed that the consideration can be restricted to ladderlike diagrams in

the calculation of the true transition probability, that is, to diagrams with one averaging point but arbitrarily many legs. In the language of standard effective-medium theory these are all contributions to the effective transition probability which result from one bond,<sup>27</sup> in the language of the method by Gouhanour, Andersen, and Fayer these are the two circle diagrams,<sup>28</sup> and in our method these are the ladder diagrams.<sup>29</sup> The diagrams, which correspond to the Movaghar-Schirmacher method, are equivalent to the Gouhanour-Andersen-Fayer diagrams.<sup>30</sup> However, since from the physical point of view these diagrams describe only the oscillations between the initial and the final site, such diagrams are completely unimportant in the Miller-Abraham hopping regime. The only approximation which is known to always yield a Miller-Abraham-like answer is the CTRW method by Scher and Lax.<sup>31</sup>

As a simple alternative to this method, which turns out to contain all of the essential physics, we suggest the consideration of a simplified model. The predictions of this model are in complete agreement with those which we obtain if we use a generalized CRTW method to describe this situation. The introduction of this model can be explained as follows. Suppose that the electron is sitting at time  $t=0$  on the site  $m$  with site energy  $\epsilon_m$ . According to Eq. (2) the probability to find the site  $m$  still occupied at time  $t$  is given by

$$n_m(t) = \exp(-tW_m), \quad (6)$$

where

$$W_m = \sum_n W_{mn}. \quad (7)$$

Due to Eq. (3) only sites within the slice  $\epsilon_m < \epsilon_n < \epsilon_m + \omega$  contribute to the sum. Thus the electron has effectively left its site after the time  $t = W_m^{-1}$ . Most likely it has jumped to a close site, but due to the dispersion of the transition probabilities there is also a small probability of finding the electron on any site within the above-mentioned slice. Thus, the energy distribution function has already started to spread. Since  $\omega$  is small this spreading is also small. To model this system we suggest ignoring this spreading. That is, we imagine that all sites within the above-mentioned slice have the same energy. Then  $W_m$  is simply the probability to jump from a site with energy  $\epsilon_m$  to any site with energy  $\epsilon_{m+1}$ . All sites accessible from a site with site energy  $\epsilon_m$  are different from all sites accessible from a site with site energy  $\epsilon_{m+1}$ . It is important to realize that  $W_m$  is not the probability of jumping from a site with site energy  $m$  to a particular site, but to any site in the slice of width  $\omega$ . To make this point clear we now use indices  $M, N, \dots$  to distinguish between a particular site and the subsystem with energy  $\epsilon_M$ . Thus, the index  $M$  does not refer to a particular site, but to a site with energy level  $\epsilon_M$ . Every energy level  $M$  contains  $\omega N \Omega$  randomly distributed sites, where  $\Omega$  is the volume of the system. The distance between the energy levels is the random quantity. In this case the probability  $\rho_M$  to find the electron on a site of the subsystem with site energy  $\epsilon_M$  simply satisfies the equation

$$\frac{d\rho_M}{dt} = \rho_{M-1}W_{M-1} - \rho_M W_M. \quad (8)$$

Consequently, the Laplace transform of the probability to find an electron on a site of the subsystem with energy  $\epsilon_M$ , if it was on a site of the subsystem with energy  $\epsilon_0$  at  $t=0$ , is given by

$$\rho_M(s) = f_M \prod_{L=0}^{M-1} (1 - sf_L) \quad (9)$$

for  $M > 0$ , and by  $\rho_0 = f_0$  for  $M = 0$ , where

$$f_M = \frac{1}{s + W_M}. \quad (10)$$

Here  $s$  is the frequency which corresponds to a Laplace transformation with respect to time. By definition the energy distribution function satisfies the equation

$$F(\epsilon_0, \epsilon; s) = \sum_{N=1}^{\infty} \langle \delta(\epsilon - \epsilon_0 - \delta\epsilon_1 - \dots - \delta\epsilon_N) \rho_N(s) \rangle + \delta(\epsilon - \epsilon_0) \langle \rho_0 \rangle. \quad (11)$$

Here  $\delta\epsilon_N$  is the energy transferred in the  $N$ th jump,  $\epsilon_N = \epsilon_0 + \delta\epsilon_1 + \dots + \delta\epsilon_N$ , and the brackets  $\langle \dots \rangle$  indicate the configuration average. By construction, the quantities  $\delta\epsilon_M$  are random quantities, which are uniformly distributed in the range  $(0, \omega)$ . Since the temperature is zero we neglect any statistical correlation between the quantities  $W_N$  for different indices  $N$ , and any statistical correlation between the quantities  $\delta\epsilon_N$  and  $W_N$ . The reason for this is simply that no site can be visited twice, since only hops down in energy can occur. Furthermore, due to translation invariance in energy space, we can put  $\epsilon_0 = 0$  and use the Fourier transformation to perform the calculation. Then we find that the energy distribution function takes the form

$$F(0, \epsilon; s) = \frac{1}{2\pi i} \int_{-i\infty}^{i\infty} dx \frac{f(s)e^{\epsilon x}}{1 - [1 - sf(s)]h(\omega x)}, \quad (12)$$

where

$$h(\omega x) = \frac{1 - e^{-\omega x}}{\omega x}, \quad (13)$$

and

$$f(s) = \left\langle \frac{1}{s + W_N} \right\rangle. \quad (14)$$

The integral (12) is determined by the singularities of the integrand, that is, by the solutions of the equation

$$0 = 1 - (1 - sf)h(\omega x). \quad (15)$$

In calculating the integral with respect to  $x$  we take into account that we are only interested in investigating the situation for large times, where the frequency  $s$  is already very

small. In this case we can restrict the consideration to that singularity which has the smallest real part in the limit of small  $s$  if  $\epsilon/\omega \gg 1$ . For

$$sf(s) \ll 1, \quad (16)$$

this singularity is situated at

$$x_0(s) = -\frac{2sf(s)}{\omega}, \quad (17)$$

and yields a simple pole. Consequently, we obtain

$$F(0, \epsilon; s) = \frac{1}{v(s)} \exp\left[-\frac{\epsilon s}{v(s)}\right], \quad (18)$$

where

$$v(s) = \frac{\omega}{2f(s)}. \quad (19)$$

Thus, the energy distribution function satisfies the equation

$$sF(0, \epsilon; s) = \delta(\epsilon) - \frac{d}{d\epsilon}(F(0, \epsilon; s)v(s)). \quad (20)$$

Thus,  $v(s)$  is the energy relaxation rate,<sup>15</sup> and the requirement (16), which we have used in the calculation of the position of the pole, is equivalent to the requirement that the transitions are quasielastic in the limit  $\epsilon/\omega \gg 1$ . We would like to note that our effective-medium theory,<sup>15</sup> which is based on the assumption that the transport is percolationlike, yields the same equation for the energy distribution function in the quasielastic approximation, but a different expression for the energy relaxation rate  $v(s)$ . This point reflects itself in the moments of the energy distribution function, as discussed below.

#### IV. THE MOMENTS

If we use Eqs. (5), (14), and (19) we find that the first derivative of the first moment with respect to time, the energy relaxation rate, is given by

$$v(t) = \frac{1}{2\pi i} \left(\frac{\omega}{2}\right) \int ds \frac{e^{st}}{sf(s)}. \quad (21)$$

In calculating this integral we restrict the consideration to large times. Doing so, we find that

$$v(t) = v_{\infty} \left\{ 1 + 2 \frac{v_{\infty}}{\omega v'} \frac{\rho_c^d}{d \ln^{d-1}(v't)} v' t \exp\left[-\frac{\ln^d(v't)}{\rho_c^d}\right] \right\}, \quad (22)$$

where

$$\rho_c = \frac{2\alpha}{(\omega N)^{1/d}} \left(\frac{d}{S_d}\right)^{1/d}, \quad (23)$$

$d$  is the dimension of the system,  $S_d$  is the solid angle,  $v' = v \exp(\gamma)$  ( $\gamma$  is the Euler constant), and

$$v_\infty = \frac{\omega v'}{2I(\rho_c)}. \quad (24)$$

Since

$$I(\rho_c) = \sqrt{\frac{2\pi}{(d-1)d^{1/(d-1)}}} \sqrt{\rho_c^{d/(d-1)}} \times \exp[(d-1)(\rho_c/d)^{d/(d-1)}], \quad (25)$$

we find that

$$v_\infty \propto \exp\left[-(d-1)\left(\frac{\rho_c}{d}\right)^{d/(d-1)}\right]. \quad (26)$$

Equation (22) is applicable, provided  $d \ln^{d-1}(v't)/\rho_c^d \gg 1$ . The details of the calculation leading to Eq. (22) are discussed in the Appendix.

Equation (22) has to be contrasted with the result which would have been obtained if the transport were percolation-like (see, e.g., Ref. 15). It differs from that in two ways. First, if the transport were percolationlike we would have obtained  $v_\infty \propto \exp(-\kappa\rho_c)$ , where  $\kappa$  is a number. Second, the system would approach the stationary energy relaxation rate  $v_\infty$  exponentially, if the time was large compared to the inverse characteristic hopping frequency. Equation (22), however, leads to extremely slow, nonanalytic relaxation at very large times.

A further investigation of the situation reveals that there are further differences between Miller-Abraham relaxation and percolationlike relaxation. These differences manifest themselves in the higher moments of the distribution function. Since

$$\langle \epsilon^n \rangle(s) = \int_0^\infty d\epsilon F(0, \epsilon; s) \epsilon^n = n! \frac{v^n(s)}{s^{n+1}} \quad (27)$$

the higher moments of the distribution function are determined by integrals of the type

$$\langle \epsilon^n \rangle(t) = \frac{n!}{2\pi i} \left(\frac{\omega}{2}\right)^n \int_c ds \frac{e^{st}}{s^{n+1} f^n(s)}. \quad (28)$$

Thus, the moments are determined by integrals over the function  $f(s)$  [see Eqs. (14) and (A7)]. The function  $f(s)$  depends truly on both the parameter  $s/\nu$  and the parameter  $\rho_c$ , and the integral (A7) which can be used for its calculation cannot be reduced to a simple integral which depends only on one parameter at small  $s$ . This sets the situation apart from that in percolation theory where the frequency dependence of the transport coefficients at low frequencies depends truly only on one parameter,  $s/\omega_0(\rho_c)$ , where  $\omega_0(\rho_c)$  is a characteristic frequency of the order of the dc conductivity. This peculiarity reflects itself in the moments of the energy distribution function. To calculate the moments we expand the function  $f(s)$  with respect to  $s$ , and use this expansion to generate a Laurent expansion of the integrand with respect to powers of  $s$ . Since the  $n$ th derivative of  $f(s)$  is

$$f^{(n)}(s=0) = (-1)^n n! \left\langle \frac{1}{(W_m)^{n+1}} \right\rangle \approx \frac{(-1)^n}{v'^{n+1}(n+1)} I[(n+1)\rho_c] \quad (29)$$

we obtain for the first moment

$$\langle \epsilon \rangle(t) = v(s=0)t + \frac{\omega}{4} \frac{I(2\rho_c)}{[I(\rho_c)]^2}, \quad (30)$$

for the dispersion  $\sigma^2(t) = \langle \epsilon^2 \rangle(t) - [\langle \epsilon \rangle(t)]^2$

$$\sigma^2(t) = \frac{\omega^2}{4} \frac{I(2\rho_c)}{[I(\rho_c)]^3} v't - \frac{\omega^2}{6} \frac{I(3\rho_c)}{[I(\rho_c)]^3}, \quad (31)$$

and for the asymmetry  $\chi(t) = \langle [\epsilon - \langle \epsilon \rangle(t)]^3 \rangle$

$$\chi(t) = -5 \left(\frac{\omega}{2}\right)^3 v't \frac{I(3\rho_c)}{[I(\rho_c)]^4} + \frac{3}{4} \left(\frac{\omega}{2}\right)^3 \frac{I(4\rho_c)}{[I(\rho_c)]^4}. \quad (32)$$

By construction the leading term in these asymptotic expansions is the term with the highest power with respect to  $t$ . The remaining terms are the next corrections. The requirement that the corrections are small compared to the leading term yields the range of validity of our approximation. If we use this philosophy we obtain from the first moment the requirement

$$vt \gg \frac{I(2\rho_c)}{I(\rho_c)}, \quad (33)$$

from the second moment the requirement

$$vt \gg \frac{I(3\rho_c)}{I(2\rho_c)}, \quad (34)$$

and from the third moment the requirement

$$vt \gg \frac{I(4\rho_c)}{I(3\rho_c)}. \quad (35)$$

Thus, since  $I(2\rho_c)/I(\rho_c) \ll I(3\rho_c)/I(2\rho_c) \ll I(4\rho_c)/I(3\rho_c) \ll \dots$  there is no simple parameter for the applicability of the small  $s$  expansion of the integrand. Every moment needs a new scale. The larger the moment the larger the smallest time to which the expansion applies. We would like to stress that this conclusion also holds for all higher moments. Here the fact that the dispersion depends truly on two parameters manifests itself.

Furthermore, from Eq. (32) and Fig. 6 below we see that for small times the asymmetry is positive and for large times negative. This fact reflects that more and more particles are captured by distribution holes when time elapses. The asymmetry of the distribution function at large times is absolutely large, so that also the deviation of the first moment of the center of the distribution function is absolutely large. However, despite this fact the distribution function becomes more

Gaussian when time elapses, since the asymmetry becomes small compared to the dispersion, that is,  $|\chi(t)|/|\sigma^2(t)|^{-3/2} \rightarrow 0$  for  $t \rightarrow \infty$ .

We would like to note that the dispersion

$$\sigma^2(t) \propto \omega^2 \nu' t \exp\left(\left[(d-1)\left(\frac{\rho_c}{d}\right)^{d/(d-1)}\right] [2^{d/(d-1)} - 3]\right) \quad (36)$$

depends strongly on the dimension. While for  $d=3$ , the main objective of our paper,  $\sigma^2(t)/(\nu t)$  is an exponentially small quantity with respect to the parameter  $\rho_c^{d/(d-1)}$ , it is exponentially large for  $d=2$ . The reason for this fact is not entirely clear. It might be caused by the nonanalytic dependence of the function  $f(s)$  at small  $s$  and large  $\rho_c$ , and thus reflects that in two-dimensional systems it is much harder to avoid the distribution hole than in three-dimensional systems.

## V. NUMERICAL RESULTS

To model the system numerically we have investigated the transport Eq. (2) both with Runge-Kutta integration and with Monte Carlo simulations. Basically we have used a  $d+1$ -dimensional cube with side length  $L$  and energy depth  $E$ , where the unit of length was chosen in units of  $\alpha^{-1}$  and the unit of energy was measured in units of  $\omega$ . To populate the cube, the spatial positions of  $NEL^d$  sites and their site energies were generated with a random number generator in such a way that the sites could be considered as uniformly distributed in our cube with volume  $L^d \times E$ . Then the cube was continued periodically in space. To model the dynamics first one site of the cube was chosen as the initial site  $i$ . Then a jump from site  $i$  to another site  $f$  was performed. To determine the site  $f$  an interval of length

$$\frac{W_{ik}}{\sum_n W_{in}}, \quad (37)$$

in the interval  $[0,1]$  was assigned to every site. Thereafter, a random number  $x$  was generated from a random number generator, which yields uniformly distributed numbers in the interval  $[0,1]$ , and then that site was chosen, the interval of which contains the number  $x$ . To determine the time  $t_{if}$  required for the jump a second random number  $y$  was generated with the same random number generator, and then  $t_{if}$  was calculated from the equation

$$t_{if} = - \frac{1}{\sum_n W_{in}} \ln y. \quad (38)$$

Then the process was repeated. To obtain the final results we averaged the results for test particles in 20 000 different configurations. To minimize the impact of the parameter  $L$  the results were also studied for different values of  $L$ . The impact of this parameter turned out to be negligible, if it was chosen in such a way that  $L > 3\rho_c$ . The parameter  $E$  was chosen in such a way that the results for the packet's shape had the

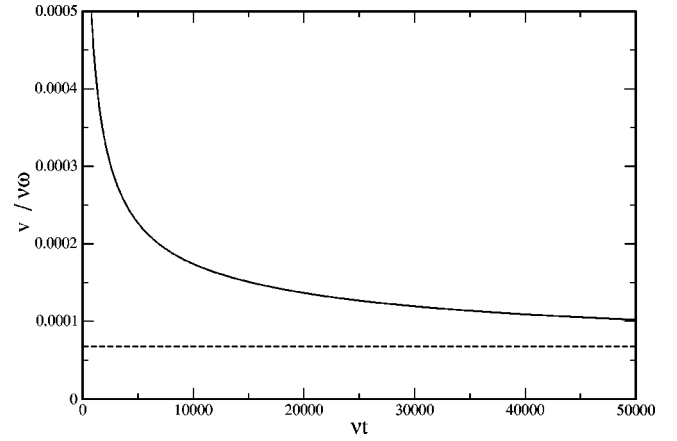


FIG. 1. Energy relaxation rate  $v(t)/(\omega\nu)$  versus time for  $d=3$  and  $N\omega/\alpha^3=0.005$ .

following property: the packet, including its width, was always well contained in the interval  $[0,E]$ , that is, in such a way that  $\epsilon(t) + 4\sigma(t) < E\omega$ . This turned out to be the case for the value  $E=20\,000$  chosen in our simulations.

In Fig. 1 we have depicted the energy relaxation rate for a three-dimensional system with  $N\omega/\alpha^3=0.005$ . According to this picture energy relaxation is fast at small times [ $v(t=0)/(\omega\nu)=7.3 \times 10^{-3}$ ], and slow at long times. A short, strong, initial decay to some value close to the stationary value of the energy relaxation rate is followed by a very slow approach to  $v_\infty$  ( $v_\infty/\omega\nu \approx 6.7 \times 10^{-5}$ ).

From the preceding section it is clear that the character of the transport is determined by the dependence of the energy relaxation rate on the parameter  $\rho_c$ . To investigate this dependence we have measured the energy relaxation rate for various values of the parameter  $\rho_c$ , and then, at first, have tried to fit our results to a dependence of the type

$$v_\infty \propto \exp(-\kappa\rho_c), \quad (39)$$

as suggested by percolation theory. Here  $\kappa$  is a number. Figure 2 shows a plot of our data for  $\ln[v_\infty/(\omega\nu)]$  versus  $\rho_c$  for three-dimensional systems. In this picture the full line is a guide for the eye of the numerical results and the dashed line is a simple straight line, drawn for comparison. If the transport were percolationlike it should be possible to fit the results to a straight line. The strong deviation of the numerical results from the straight line, in particular, at large  $\rho_c$ , shows that it is difficult to obtain a good fit. A much better result is obtained if we plot our result for the energy relaxation rate versus  $\rho_c^{3/2}$  and assume that

$$v_\infty = \omega\nu C_0 \rho_c^{-3/4} \exp(-C_1 \rho_c^{3/2}). \quad (40)$$

Such a plot (full line) is depicted in Fig. 3. If we fit the data for the full line in Fig. 3 to Eq. (40) we obtain  $C_1=0.40$  and  $C_0=0.758$ . Our Eqs. (24) and (25) yield  $C_1=0.385$  and  $C_0=0.661$ . The very good quantitative agreement between the results of our theoretical investigations and the results of the numerical calculations is also demonstrated by the dashed line in Fig. 3, which is a plot of Eqs. (24) and (25)

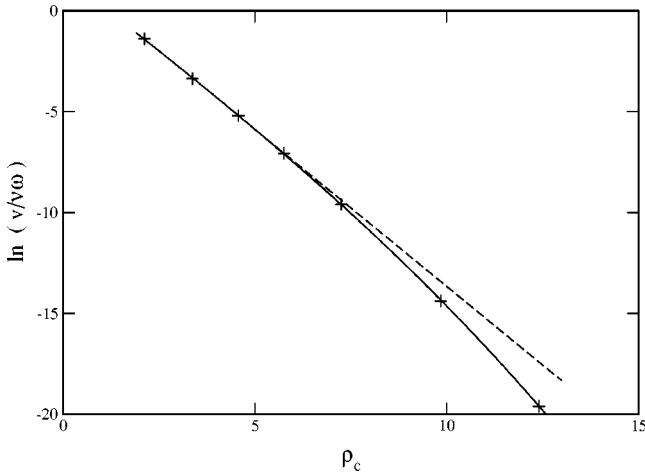


FIG. 2. Energy relaxation rate  $v_\infty/(\nu\omega)$  versus  $\rho_c$  for  $d=3$ . The full line is a guide for the eye for the numerical results, and the dashed line a straight line drawn for comparison.

and thus does not contain any adjusted parameter. Thus our calculations confirm the notion that the jumps are Miller-Abraham-like.

For two-dimensional systems the situation turns out to be same. If we try to fit our data to an expression of the type

$$v_\infty = \nu\omega C_0 \rho_c^{-1} \exp(-C_1 \rho_c^2) \quad (41)$$

we find that  $C_0=0.759$  and  $C_1=0.275$ . Equation (24) yields  $C_0=0.502$  and  $C_1=0.25$ . A plot of this fit is given in Fig. 4.

Unfortunately it turns out to be difficult to compare the predictions of our model for the time dependence of the energy relaxation rate at large times quantitatively with the numerical calculations, since for large  $\rho_c$  the corrections to the energy relaxation rate  $v_\infty$  are already so small within the range of applicability of Eq. (22), that they cannot reliably be

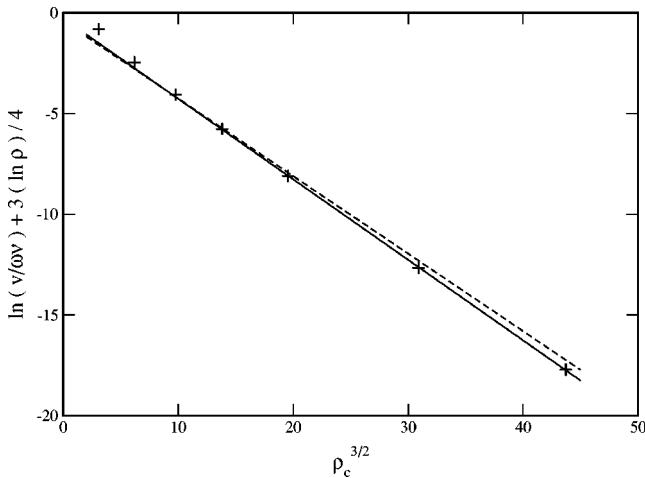


FIG. 3. Energy relaxation rate  $\ln(v_\infty) + 3 \ln(\rho_c)/4$  versus  $\rho_c^{3/2}$  for  $d=3$  ( $v_\infty$  is measured in units of  $\omega\nu$ ). The crosses are the results of the numerical calculations; the full line is the approximation of the result by a straight line. The dashed line is a plot of the result of our analytical calculation and does not contain any adjusted parameters. If we use the data for the full line we find that  $v_\infty = 0.758 \rho_c^{-3/4} \exp(-0.3998 \rho_c^{3/2})$ .

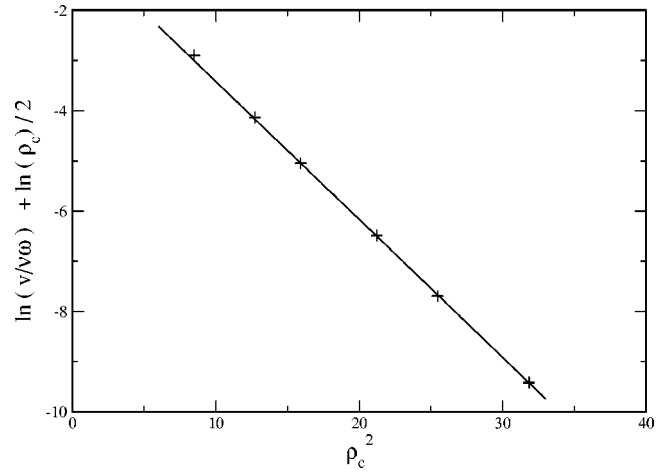


FIG. 4. Energy relaxation rate  $\ln(v_\infty) + \ln(\rho_c^2)/2$  versus  $\rho_c^2$  for  $d=2$  ( $v_\infty$  is measured in units of  $\omega\nu$ ). The crosses are the results of the numerical calculations. The straight line is the approximation of the results. If we use this approximation, we find that  $v_\infty = 0.759 \exp(-0.275 \rho_c^2)/\rho_c$ .

measured in our numerical simulations. For the second and third moments the situation is similar for large  $\rho_c$ . In order to compare the results of our theoretical calculations for these moments with the numerical calculations we have to satisfy the criteria (34) and (35), which was not possible in our numerical calculations for large  $\rho_c$ . For the mean-squared deviation our numerical calculations have only met the criterion (34) for small  $\rho_c$ , that is, for  $5 > \rho_c > 1$ . For such  $\rho_c$  we have found very good quantitative agreement between the theoretical results and the numerical ones. Nevertheless, we would like to stress that despite this fact the qualitative agreement between our theoretical and numerical results for large  $\rho_c$  is quite good. In Figs. 5 and 6 we have plotted our results for the mean-squared deviation and for the asymmetry for three-dimensional systems with  $N\omega/\alpha^3 = 0.005$  ( $\rho_c=7.25$ ) versus time. According to Eq. (31) the mean-squared deviation at large times increases linearly with time, and the first correction to the linear increase is constant and negative. If we look at Fig. 5 we see that the numerical

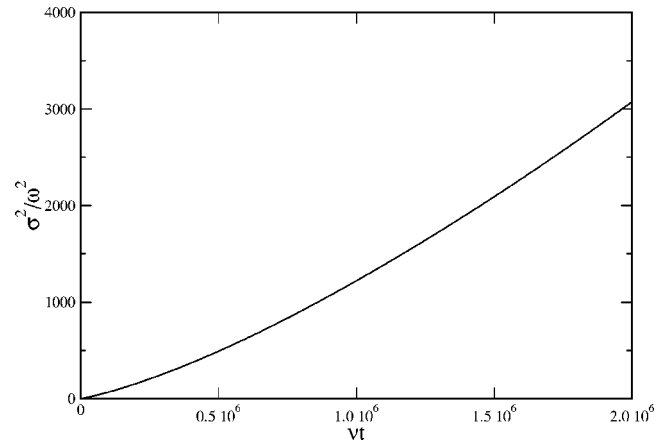


FIG. 5. Mean-squared deviation versus time for a three-dimensional system with  $N\omega/\alpha^3=0.005$ .

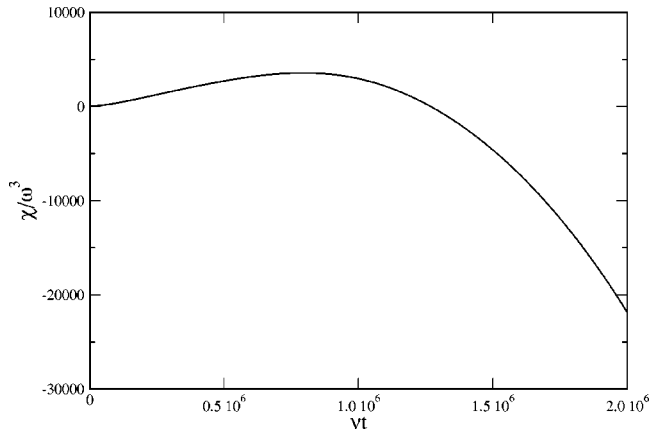


FIG. 6. Asymmetry versus time for a three-dimensional system with  $N\omega/\alpha^3 = 0.005$ .

calculations confirm that the mean-squared deviation increases with time, although we can only see the transition to the linear regime at large times. If we approximate the result for large times by a straight line, then the next correction is obviously negative, in agreement with Eq. (31).

In Fig. 6 we have depicted some numerical results for the asymmetry. According to Fig. 6 the asymmetry is positive at small times and becomes negative at large times. Furthermore, if we approximate the time dependence of the asymmetry in Fig. 6 at large times by a linear function, then the next correction is obviously positive, in agreement with Eq. (32). This fact reflects that more and more particles jump into distribution holes when time elapses, as discussed above.

Since the peculiarities of the relaxation at zero temperature result from the fact that a particle, which has jumped into a distribution hole, can only escape by an extraordinarily hard jump, we expect that the relaxation properties are strongly affected by temperature. If the temperature is finite the particle can, in principle, return to its initial site or simply jump to another site with higher energy by phonon absorption, and then choose to jump to a more effective site. In this way the distribution hole can, in principle, be avoided. Consequently, a small increase of the temperature results in an increase of the energy relaxation rate, since it eliminates the hard jumps from the possible relaxation paths. A further increase of the temperature leads to elimination of distribution holes, until all distribution holes have been cut off from the relaxation path. Thereafter, the relaxation is percolationlike. Miller-Abraham paths are no longer important. Despite this fact the energy relaxation rate increases further with increasing temperature, since the critical hopping length decreases with increasing temperature. However, since in our model a charge carrier can exchange at most an energy of the order of  $\omega$  the hopping length reaches its minimal value at a temperature  $kT \propto \omega$ . Thereafter, the situation becomes nearest-neighbor hoppinglike. Accordingly, the energy relaxation rate has a maximum as a function of temperature at an energy  $\omega \approx kT$ .

To investigate the impact of small but finite temperatures numerically we use the following transition probabilities in our Monte Carlo simulation:

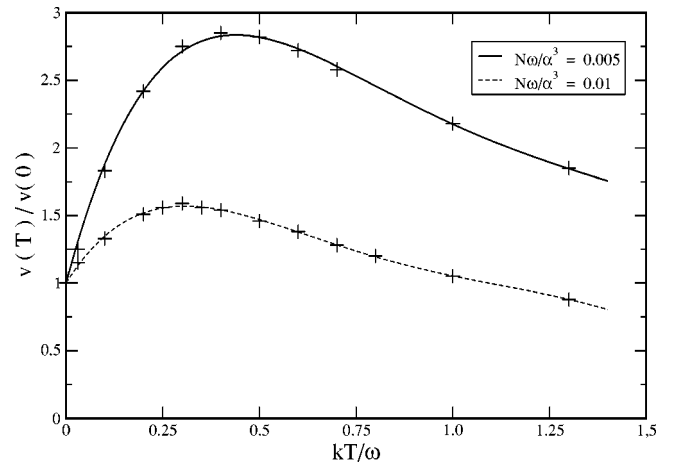


FIG. 7. Temperature dependence of the energy relaxation rate  $v_\infty$  for a two three-dimensional systems with  $N\omega/\alpha^3 = 0.005$  (solid line) and  $N\omega/\alpha^3 = 0.01$  (dashed line).

$$W_{nm} = \theta(\omega - |\epsilon_n - \epsilon_m|) \nu \exp \left[ -2\alpha |\mathbf{R}_{nm}| + \frac{\beta}{2} (\epsilon_m - \epsilon_n - |\epsilon_n - \epsilon_m|) \right], \quad (42)$$

where  $\beta = 1/(kT)$ ,  $T$  is the temperature, and  $k$  the Boltzmann constant. Results for two systems at low temperatures are depicted in Fig. 7. The maximum depends strongly on the magnitude of the parameter  $\rho_c$ . The larger the  $\rho_c$  the stronger the rise of the energy relaxation rate. Clearly, since the energy relaxation rate in the Miller-Abrahams regime is much smaller than the energy relaxation rate in the percolation regime a substantial part of this rise has to be attributed to the difference between both transport regimes. In our calculations we always find the maximum at  $kT < \omega < 1$ , although both quantities are of the same order of magnitude. If we interpret the temperature  $kT \approx \omega$  as the transition temperature from the variable-range hopping to the nearest-neighbor hopping regime then we note that there is obviously a smooth crossover between the Miller-Abraham behavior and the percolationlike variable-range hopping behavior. Accordingly, it is difficult to discriminate between both transport regimes, if we merely investigate the temperature dependence of the relaxation rate. Unfortunately, at present there is no theory which can describe the rise of the relaxation rate as a function of temperature in the Miller-Abrahams regime.

## VI. GENERALIZATION TO ENERGY-DEPENDENT DENSITY OF STATES

In the preceding sections we have seen that the relaxation at zero temperature is not percolationlike, but Miller-Abraham-like. Therefore, the question arises as to why this point was not noticed before. To answer this question we would like to note that in most of the investigations the authors have focused either on an exponential density of states, with



$$N(\epsilon) = N_0 \exp(-d\epsilon/\Delta) \quad (43)$$

( $N_0$  and  $\Delta$  are parameters), or a Gaussian density of states. A quantity that has been measured in time-resolved photoluminescence experiments is the instantaneous position of the mean energy. The arguments, which are based on the assumption that the transport is percolationlike, lead to a mean energy of the form (see, e.g., Refs. 1 and 15)

$$\langle \epsilon \rangle(t) = \Delta \ln \ln(\nu_0 t) \quad (44)$$

for an exponential density of states. The frequency  $\nu_0$  in this equation is equal to  $\nu$  if  $\omega = \infty$  (Ref. 1) and equal to  $\nu\omega/(2\Delta)$  in the quasielastic approximation.<sup>15</sup> The same double logarithmic time dependence was observed in many experiments, e.g., in the experiments in amorphous hydrogenated silicon of Ref. 32, which seemed to confirm the correctness of the assumptions used in the theoretical investigations. The parameter  $\nu_0$ , as measured in the experiments, was of the order of  $10^{12}$  Hz, thus of the order of a typical phonon frequency, and the parameter  $\Delta$  was of the order of the tailing parameter of the density of states, as expected. Thus, at first glance the agreement between those calculations which were based on the assumption that the transport is percolationlike and that  $\omega = \infty$  with the results of the experiments was quite good. On the other hand the transport measurements lead to a much larger value of the parameter  $\nu$  ( $\approx 10^{18}$  Hz) which indicates that the transitions are quasielastic.

To generalize Eq. (20) to energy-dependent densities of states we focus on the quasielastic limit. Doing so, we assume that  $\omega N'(\epsilon)/N(\epsilon) \ll 1$ , where the prime (') indicates the derivative with respect to  $\epsilon$ . Then, if the electron is sitting on a site with site energy  $\epsilon_m$ , the density of accessible sites is given by  $\omega N(\epsilon_m)$ . To calculate the energy distribution function we again use Eqs. (9)–(11). However, since now the quantity  $f_m$  (10) depends on the instantaneous position of the particle in energy space we cannot invoke translation invariance in energy space when calculating the energy distribution function, and cannot use the Fourier transformation. Therefore, we first obtain the set of equations

$$F(\epsilon_0, \epsilon; s) = f(\epsilon, s) G(\epsilon_0, \epsilon; s), \quad (45)$$

$$G(\epsilon_0, \epsilon; s) = \delta(\epsilon_0 - \epsilon) + \frac{1}{\omega} \int_0^\omega d\epsilon_1 G(\epsilon_0, \epsilon - \epsilon_1; s) \times [1 - sf(\epsilon - \epsilon_1, s)]. \quad (46)$$

Here  $f(\epsilon, s) = f_m(s)|_{\epsilon_m = \epsilon}$ . Now we use the notion that  $\omega$  is small to expand the integral with respect to  $\omega$ . Doing so, we obtain the equation

$$G(\epsilon_0, \epsilon; s) = \delta(\epsilon - \epsilon_0) + G(\epsilon_0, \epsilon; s) [1 - sf(\epsilon, s)] - \frac{\omega}{2} \frac{d}{d\epsilon} \{G(\epsilon_0, \epsilon; s) [1 - sf(\epsilon, s)]\}. \quad (47)$$

If we use this equation for the calculation of the energy distribution function we obtain

$$F(\epsilon_0, \epsilon; s) = \frac{1}{v(\epsilon, s)} \exp \left[ - \int_{\epsilon_0}^{\epsilon} d\tilde{\epsilon} \frac{s}{v(\tilde{\epsilon}, s)} \right], \quad (48)$$

where  $v(\epsilon, s)$  is again given by Eq. (19) with  $f(s)$  replaced by  $f(\epsilon, s)$ . Consequently, the energy distribution function satisfies Eq. (20) also if the density of states depends on energy, if  $v(s)$  is replaced by  $v(\epsilon, s)$ .

In deriving Eq. (47) we have restricted the expansion to the first derivative of the energy distribution function with respect to energy. The next term of the expansion, the second derivative with respect to  $\epsilon$ , would describe the energy diffusion current. We expect that this term is negligible since the temperature is zero. Note that in this approximation the width of the energy distribution function results entirely from the dispersion of the energy relaxation rate.<sup>15</sup> If we compare the terms ignored with the terms kept we find that this approximation is justified if the following inequalities are satisfied:

$$\left| \frac{\omega s}{v(\epsilon, s)} \right| \ll 1, \quad (49)$$

$$\left| \frac{\omega v'(\epsilon, s)}{v(\epsilon, s)} \right| \ll 1. \quad (50)$$

Here the prime again indicates the derivative with respect to  $\epsilon$ . Note that the inequalities (16) and (49) are identical.

We now focus on the first moment of the energy distribution function for the exponential density of states. If we are only interested in the time dependence of the first moment for large times we can put  $s = 0$  in  $v$ . Doing so, we find that at large times the first moment satisfies the equation

$$\frac{d\langle \epsilon \rangle(t)}{dt} = v[\langle \epsilon \rangle(t), s = 0]. \quad (51)$$

Accordingly, we find that at large times

$$\langle \epsilon \rangle(t) \approx \Delta \frac{d-1}{d} \ln \ln(\tilde{\nu} t). \quad (52)$$

Here we have used the abbreviations

$$\rho_c(\epsilon) = \rho_0 \exp(\epsilon/\Delta), \quad (53)$$

$$\tilde{\nu} = \frac{d\kappa_d}{(d-1)\lambda_d} \frac{\omega\nu'}{2\Delta}, \quad (54)$$

$\lambda_d = \sqrt{2\pi}/[(d-1)d^{1/(d-1)}]$ , and  $\kappa_d = (d-1)/d^{d/(d-1)}$ . Thus Eq. (52) differs from Eq. (44) only by the factor  $(d-1)/d$ , a factor of order unity, in front of the expression. If we investigate the reason for the closeness of both results we further note that Eq. (51) holds in both cases, if the relaxation is percolationlike and if the relaxation is Miller-Abraham-like. The only difference is that in the percolationlike version  $v(\epsilon) \propto \exp[-\rho_c(\epsilon)]$ , and in the Miller-Abraham version  $v(\epsilon) \propto \exp[-\rho_c(\epsilon)^{d/(d-1)}]$ . Therefore, the energy dependence of the energy relaxation rate in the Miller-Abraham version differs from that in the percolationlike version only in numbers, if the density of states has an exponential dependence

on energy, as is case in the exponential density of states or the Gaussian density of states. The agreement between the result for the mean energy in the Miller-Abraham version and the percolationlike version in such densities of states is therefore purely by chance, and would not occur if there was no exponential dependence of the density of states on energy, e.g., in a density of states which depends algebraically on energy.

The fact that nearly the same result is obtained in both cases raises the question of whether these two relaxation mechanisms can be distinguished in an experiment on energy relaxation in a material with an exponential density of states of the type (43). It turns out that the investigation of the second moment offers this chance. If the relaxation is percolationlike then the second moment becomes constant at large times, and of the order<sup>15</sup>

$$\sigma^2(t) \approx \omega \Delta. \quad (55)$$

Thus, in this case the motion of the packet in energy space at large times becomes solitonlike. On the other hand, if we use for the investigation of Eq. (48) the saddle-point method of Ref. 15 we find that in the Miller-Abraham version

$$\sigma^2(t) \propto \omega \Delta (\tilde{\nu} t)^{2^{1/(d-1)} - 2} \left( \frac{\ln \tilde{\nu} t}{\kappa_d} \right)^{2^{1/(d-1)} - 5/2}. \quad (56)$$

Accordingly, the mean-squared deviation in an exponential density of states in the Miller-Abraham version increases with time, and thus offers a way to distinguish experimentally between both cases.

We would like to note that for a two-dimensional system Eq. (56) predicts a much faster increase of the dispersion for particle packets moving in an exponential density of states than for particles moving in a constant density of states. Obviously, this is odd, and might reflect that the simplified model is not well suited for studying the situation in two dimensions, as already noted before.

## VII. CONCLUSIONS

Our numerical and theoretical investigations on energy relaxation of charge carriers at zero temperature lead to the conclusion that the relaxation at zero temperature and low excitation densities is Miller-Abraham-like. In this mode of transport the charge carriers almost always jump to the next closest site. Doing so, their motion inevitably ends up in a distribution hole or dead end, that is, on a site without close neighbors within the average site spacing. Since the charge carriers cannot return, their relaxation is determined by hard jumps out of dead ends at long times.

To model the relaxation we have developed a simplified model. In its spirit this model differs from the original Miller-Abraham model in that the jumps are not restricted to jump to the next-nearest site, but additionally jumps to remote sites have been taken into account. We believe that the consideration of remote sites is important for those quantities, which are related to the dispersion of the transport coefficients. The remote sites have, however, little impact on the first moment of the energy distribution function. If we

would restrict the consideration to jumps to the next closest site, we would obtain nearly the same energy relaxation rate at large times. The only difference would be that  $\nu'$  is replaced by  $\nu$ . We would like to note that a model, which in its structure is close to our model, has also been developed in Ref. 11. We believe that with that model it should have also already been possible to note that the transport at zero temperature is fundamentally different from the percolationlike descriptions used in the literature. However, since in this paper the authors have only focused on energy relaxation in densities of states with exponential dependence on energy, it was difficult to notice this fact, as explained in detail in Sec. VI.

According to the predictions of our simplified model the motion of a particle packet in energy space in the Miller-Abraham mode differs strongly from that in the percolationlike version. Already the energy relaxation rate is much smaller in the Miller-Abraham version than in the percolationlike version. Furthermore, there are strong differences between those quantities which are related to the dispersion of the transport coefficients, such as the mean-squared deviation of the packet in energy space or their asymmetry. While for a constant density of states the packet becomes Gaussian at sufficiently large times in the percolationlike version, in the Miller-Abraham mode the asymmetry is large and negative at large times. This fact indicates that in the Miller-Abraham mode the packet tends to stick. Another way to state this fact is that the memory of the initial state is kept for a very long time.

If we compare the predictions of our simplified model with the results of numerical calculations, in which do not use any assumptions, we find excellent quantitative agreement between the theoretical and numerical results for the first moment of the energy distribution function. For the higher moments of the energy distribution function we only find qualitative agreement. We attribute this fact to the complicated dispersion of the energy relaxation rate. In contrast to the percolationlike version, in which the impact of the dispersion on the transport coefficients truly depends on only one parameter, the dispersion of the energy relaxation rate in the Miller-Abraham regime truly depends on two parameters. This fact manifests itself in the range that determines the validity of Eqs. (22), (29), and (32). The lower limiting time, which is needed in order to establish Eq. (29), is much larger than the lower limiting time for the applicability of Eq. (22), but small compared to the lower limit of Eq. (32). It can be checked that the times used for the numerical calculations do not satisfy the criteria for the applicability of Eqs. (29) and (32), but they do for the criterion of applicability of Eq. (22). Therefore, we believe that the remaining discrepancies between the numerical and the theoretical calculations of the second and the third moments result from the fact that the times, which could be realized in the numerical calculations, were too small to achieve quantitative agreement.

The peculiarities of the relaxation at zero temperature result from the fact that the particle loses the connection to its starting site after a jump. However, at any finite temperature the particle can, in principle, return to its initial site, and then jump to a more favorable site, rather than to the dead end.

Therefore, the relaxation rate is strongly affected by a rise of the temperature. To investigate the impact of the temperature we have used Monte Carlo simulations. We find that the impact of the temperature is governed by the parameter  $\rho_c$  and by the parameter  $\omega/kT$ , the ratio between the maximal amount of energy transferable in one jump and the thermal energy. For very low temperatures a weak rise of the temperature results in a strong increase of the relaxation rate. The strength of the increase depends on the magnitude of the parameter  $\rho_c$ . The larger the  $\rho_c$  the stronger the increase. This increase is due to the fact that the relaxation path changes with increasing temperature. Since the magnitude of the relaxation rate at zero temperature is much smaller in the Miller-Abraham regime than that suggested by percolation theory a significant part of the rise has to be attributed to the fact that dead ends are gradually switched off from the relaxation path. The energy relaxation rate reaches a maximum at a temperature  $kT$ , which is of the same order of magnitude as  $\omega$ , but satisfies  $kT/\omega < 1$ . Beyond the maximum the relaxation is nearest-neighbor hoppinglike. Since for  $\omega/kT \gg 1$  all dead ends have been switched off we expect that the transport is percolationlike in this limit.

A very interesting question is how the transport mechanism is affected by an increase of the number of excitations. To some extent energy relaxation at zero temperature is similar to hopping transport in a strong electric field, as already stressed in the Introduction. In Refs. 33 and 34 arguments have been put forward leading to the conclusion that the hopping current in a strong electric field in a narrow impurity band, which is of a Miller-Abraham type for very small charge-carrier concentrations, becomes eventually percolationlike by increasing the charge-carrier concentration, if the thermal energy is large compared to the bandwidth of the impurity band. The reason for this is simply that a part of the charge carriers can be used to fill the Miller-Abraham traps, so that the remaining charges carriers can pass the sample without seeing traps at all. In the case of the current in an impurity band this argument leads to a strong dependence of the nonohmic current on the charge-carrier concentration. Photoluminescence experiments, however, indicate that this argument affects the relaxation at least less. According to experiments on amorphous hydrogenated silicon (see, e.g., Refs. 24–26) there is a low excitation generation regime in which the kinetics of the charge carriers is independent of the generation rate and in contradiction to the existing theoretical predictions. One of the reasons for this might be that in contrast to the electric current in the impurity band in the limit in question, which is determined by the most effective current path, the energy relaxation rate is determined by the position of the maximum of the energy distribution function. Those paths, which are most effective, and thus would be most important for the current, do not determine the maximum of the energy distribution function at low excitation generation rates, but only its outer tails, if the density of the charge carriers passing them is not too high. Obviously, further numerical simulations have to be performed to determine the density, which governs the impact of the charge-carrier concentration on the relaxation rate.

Finally the question remains as to which materials are

most suitable for observing the Miller-Abraham relaxation. Clearly, the steady-state Miller-Abraham relaxation rate can only be observed if the number of Miller-Abraham traps is sufficiently large. This requirement is always satisfied if the density of states is not bounded from below, as in this paper. However, in all relevant physical systems the density of states for charge carriers far from equilibrium is bounded from below by the Fermi surface. Due to the presence of the Fermi surface a charge carrier far from equilibrium can at most make  $n_{max}$  jumps, where  $n_{max}$  is of the order of  $(\epsilon_0 - \epsilon_F)/\omega$  ( $\epsilon_0$  is the initial energy,  $\epsilon_F$  the final energy). If all of those jumps were performed with the steady-state energy relaxation rate then the relevant hopping length would be of the order of  $r_c \propto \alpha^{1/(d-1)} n_{max}^{1/(d-1)} l^{d/(d-1)}$ , where  $l$  is determined by the number of sites  $Z_{\epsilon_0}$  below  $\epsilon_0$  according to the relationship  $l^d(\epsilon_F - \epsilon_0)N = Z_{\epsilon_0}$ . The probability that in none of  $n$  subsequent jumps there is a site within a sphere of radius  $r_c$  is of the order of

$$1 - \exp(-n\{\exp[-r_c^d/(n_{max}l^d)]\}). \quad (57)$$

In order that the steady-state Miller-Abraham energy relaxation rate can be reached the exponent in the expression (57) necessarily has to be large compared to unity. If we use the expression for  $r_c$  above we therefore conclude that the steady Miller-Abraham relaxation rate can only be reached in systems in which the situation  $\alpha l < 1$  can be realized, since  $n < n_{max}$ . Accordingly, either the number of states above the Fermi energy has to be sufficiently large, or  $\omega$  must be sufficiently small. Thus, at first glance it seems that band tails of amorphous semiconductors, being particularly wide, would be the most promising candidates. However, in Sec. VI we have shown that in such materials, which typically have a density of states with exponential energy dependence, there is not much difference between the Miller-Abraham energy relaxation rate and the percolation energy relaxation rate. For such densities of states the energy relaxation rates differ only in numbers. While this fact gives some support to the present theory, since it shows that the results of the present theory are in line with the experiments, it also makes it hard to distinguish between both transport regimes experimentally. For such materials the only way to distinguish between the two transport mechanisms experimentally is in the investigation of the second and the third moments of the energy distribution function. To our knowledge such measurements have not been performed thus far.

Therefore it seems that the most promising candidates for the investigation of Miller-Abraham relaxation processes are two-dimensional Anderson insulators such as those used, e.g., in Refs. 9 and 35–38, with weak dependence of the density of states on energy, which, for our purpose, can be considered as materials with a density of states unbounded from above. For such densities of states the differences between the Miller-Abraham relaxation and percolationlike relaxation are particularly strong, as shown in this paper. In such materials a particular slow decay of the relaxation current is observed at low temperatures. If, e.g., we look at the curve for the decay of the excess current in Ref. 35 (Fig. 5 of Ref. 35) then we see a slow decay of the excess current for

times  $t > 1$  s, while the main decay of the excess current is quick and occurs for  $t < 1$  s. If we look at the above-mentioned experimental results then we see that it is neither qualitatively nor quantitatively possible to reconcile the experimental results with percolation theory. In the case of percolation theory the time scale which governs the approach to the steady relaxation rate is of the order (see, e.g., Ref. 15)

$$t_{perc} \approx \rho_c \exp(\rho_c) / \nu, \quad (58)$$

where  $\rho_c$  is the dimensionless critical hopping length. A typical value for  $\nu$  is  $\nu \approx 10^{12}$  Hz. Thus, in order to produce a relaxation time of the order of 1 s, as observed in the experiment in Ref. 35, one would need a hopping length of the order of  $\rho_c = 25$ , which is unreasonably large. Moreover, it would be hard to explain why the relaxation rate should increase so strongly with temperature as observed in the experiments, since the distribution holes would not be relevant. Due to the consideration of the distribution holes our theory leads to a much stronger increase of the relaxation rate with temperature such as the percolationlike theory. We admit that it is not clear whether the temperatures in the experiments are low enough to observe Miller-Abraham-like behavior. Since there is no transition temperature between the Miller-Abrahams regime and the percolationlike variable-range hopping regime the question as to whether the temperatures are low enough to observe Miller-Abraham behavior simply cannot be answered by investigating the temperature dependence of the relaxation rate alone. If, however, despite this fact we use our theory then we can try to understand the emergence of glassy dynamics in the above-mentioned materials as a transition between two types of different relaxation modes, that is, a transition between percolationlike relaxation and Miller-Abraham relaxation. Both transport regimes are separated by a characteristic temperature, the glass temperature, which is given by the maximum of the relaxation rate as a function of temperature. In our case, the characteristic time, which sets the fast initial decay apart from the low approach to equilibrium is given by the Miller-Abraham time

$$\frac{d \ln^{d-1}(\nu t_{MA})}{\rho_c^d} = 1. \quad (59)$$

If we use the Miller-Abraham time scale to produce a relaxation time of the order of 1 s, as observed in the experiment in Ref. 35, we only need a hopping length of  $\rho_c = 7.4$  for  $d = 2$ . This is perhaps larger than expected, but not unrealistically large. If we take this point of view then we conclude that obviously those particles that have passed the samples on percolation paths, since the Miller-Abraham traps on their path have already been occupied by other particles as discussed above, are not important at all for the investigation of slow relaxation processes in Anderson insulators.

We would like to point out that in the above-mentioned experiments a strong dependence of the relaxation rate on the electron concentration has been observed. This has led the authors of Refs. 9, and 37–39 to the conclusion that interaction effects are responsible for the smallness of the relaxation

rate. However, the dependence of the relaxation rate on the electron concentration could also reflect a dependence of the parameters  $\omega$  and  $\alpha$  on the charge-carrier concentration. If this were true, the explanation proposed here could be an alternative to the explanation given in the papers referred to above.

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## APPENDIX: CALCULATION OF THE ENERGY RELAXATION RATE FOR LARGE TIMES

To calculate the integral (21) we restrict the consideration to large times. Doing so, we can use the approximation

$$\frac{1}{f(s)} = \frac{1}{f(0) - (f(0) - f(s))} \approx \frac{1}{f(0)} \left( 1 + \frac{f(0) - f(s)}{f(0)} \right). \quad (A1)$$

If we use this approximation in Eq. (21) we find that

$$v(t) = v_\infty \left\{ 1 + \frac{1}{2\pi i} \int ds \frac{\exp(st)}{s} \left[ 1 + \frac{f(0) - f(s)}{f(0)} \right] \right\}, \quad (A2)$$

where

$$v_\infty = \frac{\omega}{2} \frac{1}{f(0)}. \quad (A3)$$

Accordingly,

$$\frac{dv(t)}{dt} = -v_\infty \frac{f(t)}{f(0)}, \quad (A4)$$

where

$$f(t) = \frac{1}{2\pi i} \int ds f(s) e^{st}, \quad (A5)$$

so that

$$v(t) = v_\infty \left[ 1 + \frac{2v_\infty}{\omega} \int_t^\infty dt' f(t') \right]. \quad (A6)$$

To calculate the integrals further we have to be familiar with the function  $f(s)$  [Eq. (14)]. The calculation of the configuration average in Eq. (14) leads to<sup>39</sup>

$$f(s) = \int_0^\infty dt \exp\left(-st - \frac{d}{\rho_c^d} \int_0^\infty dx x^{d-1} \times \{1 - \exp[-\nu t \exp(-x)]\}\right). \quad (A7)$$

Since the main contribution to the integral (A7) for small  $s$  and large  $\rho_c$  results from large  $\nu t$  we use the approximation

$$\exp\left(-\frac{d}{\rho_c^d}\int_0^\infty dx x^{d-1}\{1-\exp[-\nu t \exp(-x)]\}\right) \approx \exp\left[-\frac{\ln^d(\nu' t)}{\rho_c^d}\right]. \quad (\text{A8})$$

Then the saddle-point approximation yields

$$f(0) = \frac{I(\rho_c)}{\nu'}, \quad (\text{A9})$$

where  $I(\rho_c)$  is given by Eq. (25). Thus, the problem remaining is the calculation of the integral

$$J(\nu' t, \rho_c) = \int_t^\infty dt' f(t') \approx \int_t^\infty dt' \exp\left[-\frac{\ln^d(\nu' t)}{\rho_c^d}\right]. \quad (\text{A10})$$

If we change the integration variables we obtain

$$J(\nu' t, \rho_c) = \frac{1}{\nu'} \int_{\ln(\nu' t)}^\infty dx \exp\left(x - \frac{x^d}{\rho_c^d}\right). \quad (\text{A11})$$

To calculate this integral in the limit  $d \ln^{d-1}(\nu' t)/\rho_c^d \gg 1$  we expand the exponent into a power series with respect to  $x$  at  $x = \ln(\nu' t)$ . Under the condition  $d \ln^{d-1}(\nu' t)/\rho_c^d \gg 1$  the first derivative of the exponent has a negative sign, and the contribution from the second derivative is negligible. Therefore, we obtain

$$\begin{aligned} J(\nu' t, \rho_c) &\approx \frac{\exp\left[\ln(\nu' t) - \frac{\ln^d(\nu' t)}{\rho_c^d}\right]}{\nu'} \int_{\ln(\nu' t)}^\infty dx \\ &\times \exp\left\{-d \frac{\ln^{d-1}(\nu' t)}{\rho_c^d} [x - \ln(\nu' t)]\right\} \\ &= \frac{\exp\left[\ln(\nu' t) - \frac{\ln^d(\nu' t)}{\rho_c^d}\right]}{\nu'} \frac{\rho_c^d}{d \ln^{d-1}(\nu' t)}, \end{aligned} \quad (\text{A12})$$

which together with Eqs. (A6) and (A9) yields Eq. (22).

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