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Dispersive energy transport and relaxation in the hopping regime

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A method for investigating relaxation phenomena for charge-carrier hopping between localized tail states is developed. It allows us to consider both charge and energy *dispersive* transport. The method is based on the idea of quasielasticity: the typical energy loss during a hop is much less than all other characteristic energies. We investigate two models with different density-of-state energy dependencies with our method. In general, we find that the motion of a packet in energy space is affected by two competing tendencies. First, there is a packet broadening, i.e., dispersive energy transport. Second, there is a narrowing of the packet if the density of states is depleting with decreasing energy. It is the interplay of these two tendencies that determines the overall evolution. If the density of states is constant, only broadening exists. In this case a packet in energy space evolves into a Gaussian one, moving with a constant drift velocity and mean-square deviation increasing linearly in time. If the density of states depletes exponentially with decreasing energy, the motion of the packet slows down tremendously with time. For large times the mean-square deviation of the packet becomes constant, so that the motion of the packet is ''solitonlike.''

I. INTRODUCTION

In recent years much attention has been devoted to the study of relaxation processes of nonequilibrium charge carriers in strongly localized systems, where transport proceeds via phonon-assisted hopping, as with photoexcited charge carriers in band tails (see, e.g., Refs. 1–6) and Anderson insulators (see, e.g., Refs. 7–9). In such systems particularly small relaxation rates are observed. Often the smallness of the relaxation rate is attributed to interaction effects. However, even in strongly localized noninteracting electron systems, long-lasting relaxation processes are known to be the rule, and not the exception.[10]

A theoretical investigation of such relaxation processes is notoriously difficult, since the system is strongly disordered and always in a transient state. In most problems one is interested in time scales which are large compared to the time needed for a single hop. For such time scales all quantities depend strongly on frequency, even for very low frequencies, so that the consideration of dispersive transport is vital. On the other hand, in most problems of interest both spatial and energetic disorder exists. Due to the latter fact, transitions are inelastic. The inelastic character of transitions, the relaxation, leads to a flow of energy from the electron system to the phonon system. Due to disorder, this transport of energy is also dispersive. Therefore, dispersive energy transport and relaxation are intimately connected. Investigation of the relaxation process requires both a consideration of dispersive particle transport *and* a consideration of dispersive energy transport.

The intricate physical situation also manifests itself in equations that have to be considered. Since transport is inelastic even in the simplest approximation, the investigation requires solution of integral equations (see, e.g., Refs. 11–13). Due to the fact that the particle moves in an energy-dependent density of states, the kernel of these integral equations does not depend only on the difference between the site energies. The traditional method for handling the situation, percolation theory, is not applicable here. The effective-medium methods of Movaghar and co-workers, as pointed out by those authors, give incorrect results for systems at low temperature.[14,15] Furthermore, beyond the Markovian approximation the derivation of these integral equations itself represents an intricate problem. To our knowledge, in addition to the attempts by Movaghar and co-workers, numerical investigations (see, e.g., Refs. 1 and 2), and physical intuitive considerations (see, e.g., Ref. 1), mainly Markovian equations have been used (see, e.g., Ref. 12).

It is the aim of the present paper to fill this gap, and to provide a formalism that can be used for studying relaxation phenomena of strongly localized charge carriers far from equilibrium in the hopping regime, taking into account both dispersive particle transport and dispersive energy transport. To this end we focus, for the sake of definiteness, on relaxation of photoexcited, noninteracting, nonequilibrium charge carriers in band tails of, e.g., amorphous semiconductors, like amorphous Si:H. At first glance, this problem seems to be rather special. If the number of charge carriers excited is small, the Fermi correlation is negligible, so that one has to cope with linear rate equations. The linearity of the transport equations is, of course, the basic ingredient in solving the problem. A closer look at the problems of interest reveals, however, that most of the problems can be formulated in this way. Clearly, how to achieve linearization depends on the experiment chosen, but, provided the number of charge carriers excited is small, the smallness of the particle number can always be invoked. In this case, the number of particles at a site is certainly not small compared to its equilibrium value, but small compared to unity. After linearization the structure is, in principle, quite similar to the case considered here.

Below we present a derivation of a framework for the consideration of relaxation phenomena due to phonon-assisted hopping at zero temperature. We focus on the limit of strong localization, where dispersive effects are expected to be most pronounced. Here the disorder manifests itself in a strong dependence of transport coefficients on frequency for very low frequencies. Consequently, in this regime the diffusion propagator cannot be calculated from Markovian transport equations, such as those used, e.g., in Ref. 12. To simplify our integral equations we use the concept of quasielasticity: the particle changes its energy only by a small amount in one hop.

The method is applied to a study of dispersive energy transport. We find that two tendencies are always present as long as the density of states does not decrease with increasing energy. First, there is a widening of the packet due to statistical spreading, i.e., dispersive energy transport. Second, there is narrowing of the packet due to a decrease of the density of states with decreasing energy. The overall evolution is determined by the interplay of these two tendencies. We have studied both tendencies in two limiting cases: for a particle moving in a constant density of states, and for a particle moving in an exponential density of states. In the first case the impact of the density-of-state decrease is absent, so that only the statistical spreading is present. Here the packet evolves into a Gaussian packet moving with constant drift velocity in energy space. The packet width increases with time as $\sqrt{t}$. The other result is obtained for an exponential density of states. Here both tendencies are present. We find that the velocity of the packet slows down strongly with time. In this case the mean-square deviation of energy eventually becomes time independent. Consequently, the motion of the packet in energy space is ''solitonlike.'' Concrete results on the diffusion propagator, its time dependence, and its moments for exponential densities of states are of relevance for photoluminescence experiments on amorphous Si:H.

II. BASIC EQUATIONS

We consider photoexcited, localized charge carriers in band tails at zero temperature. After excitation the charge carriers lower their energy by phonon-assisted hops between localized states. Since $T=0$, only hops from higher to lower energy occur. In this situation the charge carriers are in strong nonequilibrium. We assume that the number of excited charge carriers is small, and that their energies are far from the Fermi level, so that it is very unlikely that an electron jumps to a site already occupied. Consequently, we can neglect the Fermi correlation. In this case electron transport can be described by the rate equation.[17,18]

$$\frac{dp_m}{dt}=\sum_{m'}\,(p_{m'}W_{m'm}-p_mW_{mm'}). \tag{1}$$

In calculating the transition probabilities, we assume that the electron-phonon coupling strength is weak, so that only one-phonon processes have to be taken into account. Then, at zero temperature, the transition probabilities are given by

$$W_{m'm}=\Theta(\omega-\varepsilon_{m'}+\varepsilon_m)\Theta(\varepsilon_{m'}-\varepsilon_m)W(|\mathbf{R}_{mm'}|), \tag{2}$$

where

$$W(|\mathbf{R}_{mm'}|t)=\nu e^{-2\alpha|\mathbf{R}_{mm'}|}. \tag{3}$$

Here α is the inverse of the localized state radius, and ν is the phonon frequency. The energy ω is the upper bound for the energy transferred in one hop. Note that in the materials of interest ω can be much smaller than the Debye frequency, since not all phonons can interact with localized electrons equally well. Short-wavelength phonons are ineffective since the electron-phonon coupling constant tends to zero for momenta q with $q/2\alpha\gg1$. Therefore, the effective upper phonon momentum is of the order 2α, and not of the order of the inverse lattice constant of the host material. Furthermore, in disordered systems the high-energy phonons are localized, and need not contribute to transport.

The first step function in front of the transition probabilities restricts transitions to those between sites separated by at most ω in energy space. Thus it decreases the energy relaxation rate. In impurity conduction, and in nearly all papers on relaxation of charge carriers in band tails, this step function is usually replaced by unity. In impurity conduction this is quite reasonable, since it is assumed that hops are restricted within a narrow strip near the Fermi level, which is small compared to the Debye energy. In the band-tail problem, however, we can see no reason for neglecting it in advance.

To calculate the transport quantities of interest, we have to calculate the diffusion propagator. In order to render the analytical calculations feasible, we introduce continuous coordinates. The change of representation is defined by

$$n(\rho)=\sum_m\;p_m\delta(\rho-\rho_m), \tag{4}$$

where $\rho=(\mathbf{R},\varepsilon)$ and $\rho_m=(\mathbf{R}_m,\varepsilon_m)$. In this representation rate equation (1) takes the form

$$\frac{dn(\rho)}{dt}=\int d\rho' n(\rho')V(\rho',\rho), \tag{5}$$

where V is determined by the equations

$$V(\rho',\rho)=\int d\rho_1\,\eta(\rho_1)w_{\rho_1}(\rho',\rho), \tag{6}$$

$$w_{\rho_1}(\rho',\rho)=W(\rho',\rho_1)[\delta(\rho_1-\rho)-\delta(\rho'-\rho)], \tag{7}$$

and

$$\eta(\rho)=\sum_m\;\delta(\rho-\rho_m). \tag{8}$$

The Laplace-transformed equation is

$$sn(\rho)-n_o(\rho)=\int d\rho' n(\rho')V(\rho',\rho), \tag{9}$$

where $n_o(\rho)=n(\rho,t=0)$ is the initial condition. We will assume that $p_m(t=0)$ is a function $p_0(\mathbf{R}_m,\varepsilon_m)$, so that $n_o(\rho)=p_0(\rho)\eta(\rho)$.

Equation (9) can be solved using the Green's-function method. The solution is given by

$$n(\rho)=\int d\rho' n_o(\rho')\Phi(\rho',\rho). \tag{10}$$

The Green's function satisfies the equation

$$s\Phi(\rho',\rho)-\int d\rho_1\, V(\rho',\rho_1)\Phi(\rho_1,\rho)=\delta(\rho'-\rho). \tag{11}$$

Due to probability conservation, the Green's function Φ and the probability $w_{\tilde{\rho}}$ satisfy the relations

$$\int d\rho\, \Phi(\rho',\rho)=\frac{1}{s}, \tag{12}$$

$$\int d\rho\, w_{\tilde{\rho}}(\rho',\rho)=0. \tag{13}$$

III. CONFIGURATION AVERAGE

In order to calculate the configuration average, we assume that the sites are distributed homogeneously in space. The distribution of site energies is supposed to be given by some distribution function $p(\{\varepsilon_i\})$. Accordingly, the average of any quantity depending on the energy and positions of sites is given by

$$\langle A\rangle=\int \Pi_m \frac{d\mathbf{R}_m}{\mathcal{V}} d\varepsilon_m\, p(\{\varepsilon_m\})A(\{\mathbf{R}_m,\varepsilon_m\}), \tag{14}$$

where $\mathcal{V}$ is the volume of the system. The application of the averaging procedure to the structural factor η serves, in particular, as a definition for the density of states, i.e.,

$$\mathcal{N}(\varepsilon)=\langle \eta(\rho)\rangle. \tag{15}$$

Products of the structural factor η are averaged according to

$$\langle \eta(\rho_1)\cdots\eta(\rho_n)\rangle=\mathcal{N}(\rho_1)\,\delta(\rho_1-\rho_2)\cdots\delta(\rho_{n-1}-\rho_n). \tag{16}$$

Using these definitions, the configuration average of Eq.(10) can be calculated diagrammatically.[19,20] The diagrammatic method leads to the following set of equations for the calculation of the configuration average $\langle n(\rho)\rangle$ of the electron density $n(\rho)$ (Ref. 20):

$$\langle n(\rho)\rangle=\int d\rho_1\, d\rho_2\, p_0(\rho_1)S(\rho_1,\rho_2)F(\rho_2,\rho), \tag{17}$$

$$sF(\rho',\rho)=\delta(\rho'-\rho)+\int d\rho_1\, \Pi(\rho',\rho_1)F(\rho_1,\rho), \tag{18}$$

$$\Pi(\rho',\rho)=\int d\rho_1\, \mathcal{N}(\rho_1)\Pi_{\rho_1}(\rho',\rho), \tag{19}$$

$$\Pi_{\rho_1}(\rho',\rho)=w_{\rho_1}(\rho',\rho) +\int d\rho_2\, d\rho_3\, w_{\rho_1}(\rho',\rho_2)F(\rho_2,\rho_3)\Pi_{\rho_1}(\rho_3,\rho), \tag{20}$$

$$S(\rho',\rho)=\mathcal{N}(\varepsilon')\left[\delta(\rho'-\rho)+\int d\rho_1\, F(\rho',\rho_1)\Pi_{\rho'}(\rho_1,\rho)\right]. \tag{21}$$

Here $F(\rho',\rho)=\langle\Phi(\rho',\rho)\rangle$.

IV. EFFECTIVE MEDIUM

Given the system of integral equations (18)–(21), the main problem is to find an approximate self-consistent solution to it. The situation is quite similar to that of the calculation of the equilibrium conductivity in a disordered system.[19,20] There, an approximate solution of the system could be found by introducing a proper decomposition of the function F, the diffusion propagator, into short- and long-wavelength limits, according to

$$F(\rho',\rho)=f(s)C(\varepsilon)\,\delta(\rho'-\rho)+\tilde{F}(\rho',\rho), \tag{22}$$

where $f(s)$ was a frequency-dependent parameter that could be related to the critical hopping length R_c via the equation

$$f\nu=\exp(-2\alpha R_c), \tag{23}$$

and $C(\varepsilon)$ was an energy-dependent function determined by the principle of detailed balance. This decompostion originated from the notion that the integrals in the integral equations are governed by products of transition probabilities and diffusion propagators, and for strongly localized electrons the latter quantities are short-ranged functions compared to the transition probabilities. Using the decomposition (22), the effective-medium approximation reduces to the replacement of F by $fC(\varepsilon)\,\delta(\rho'-\rho)$ in the calculation of the effective transition probability Π [Eq. (20)] and of the irreducible block S [Eq. (21)].

Here in the band-tail problem we use the same philosophy. We first decompose the diffusion propagator into two parts, according to

$$F(\rho',\rho)=f(\varepsilon,s)\,\delta(\rho'-\rho)+\tilde{F}(\rho',\rho). \tag{24}$$

Then, to investigate the consequences of this decomposition, we insert Eq. (24) into Eq. (20). Performing a partial summation, we obtain

$$\Pi_{\tilde{\rho}}(\rho',\rho)=\tilde{w}_{\tilde{\rho}}(\rho',\rho)+\int d\rho_1\, d\rho_2\, \tilde{w}_{\tilde{\rho}}(\rho',\rho_1) \times\tilde{F}(\rho_1,\rho_2)\Pi_{\tilde{\rho}}(\rho_2,\rho), \tag{25}$$

where the renormalized transition probabilities $\tilde{w}_{\tilde{\rho}}(\rho',\rho)$ are given by Eq. (7), with W replaced by

$$\tilde{W}(\rho',\rho;s)=\frac{W(\rho',\rho)}{1+f(\varepsilon',s)W(\rho',\rho)}. \tag{26}$$

Note that the renormalized transition probability depends now on s via the function $f(\varepsilon,s)$.

At this point, introduction of the function $f(\varepsilon,s)$, the effective medium, still seems to be rather arbitrary. However, if we now choose the effective medium in such a way that the integrals over $\tilde{F}$ vanish, the renormalized transition probabilities yield the exact solution of the diffusion problem. In that case the function S turns into

$$S(\rho',\rho)=\mathcal{N}(\varepsilon')\,\delta(\rho'-\rho), \tag{27}$$

so that Eq. (17) can be cast in the form

$$\langle n(\rho;s)\rangle=\int d\rho'\langle n_o(\rho')\rangle F(\rho',\rho). \tag{28}$$

Therefore, the function $F(\rho',\rho)$ can be identified with the diffusion propagator.

Note that, in contrast to the calculation of the conductivity close to equilibrium, the critical hopping length is expected to depend somehow on the instantaneous position of the particle in the tail. Consequently, in the present situation f is a function of energy, and not a parameter, that has to be determined self-consistently.

V. DIFFUSION PROPAGATOR IN EFFECTIVE-MEDIUM APPROXIMATION

For the moment we put aside the question of the determination of the effective medium to elaborate further on the consequences of the renormalization of the transition probabilities. To this end we focus on the diffusion propagator.

The equation for the diffusion propagator is given by Eq. (14). In the effective-medium approximation, when calculating the irreducible part Π, only the lowest-order contribution to Π with respect to $\tilde{F}$ is taken into account, so that $\Pi_{\tilde{\rho}}=\tilde{w}_{\tilde{\rho}}$. In that approximation the equation for the diffusion propagator in the momentum representation reads

$$\begin{aligned}sF(\mathbf{q}|\varepsilon',\varepsilon)={}&\delta(\varepsilon'-\varepsilon)\\&+\int d\varepsilon_1\{F(\mathbf{q}|\varepsilon',\varepsilon_1)\tilde{W}(\mathbf{q}|\varepsilon_1,\varepsilon;s)\mathcal{N}(\varepsilon)\\&-F(\mathbf{q}|\varepsilon',\varepsilon)\tilde{W}(\mathbf{0}|\varepsilon,\varepsilon_1;s)\mathcal{N}(\varepsilon_1)\}.\end{aligned} \tag{29}$$

Of course, since we do not yet know what the effective medium looks like, and moreover, as the equation is a complicated integral equation, we cannot find a solution. The fact that the effective medium is a function of energy makes the problem much more complicated. Progress can only be achieved if we can find arguments to simplify the equation considerably. To this end we focus on the renormalized transition probability. According to Eqs. (2) and (26), this is given by

$$\begin{aligned}\tilde{W}(R|\varepsilon',\varepsilon;s)&=\Theta(\varepsilon'-\varepsilon)\Theta(\omega-\varepsilon'+\varepsilon)\frac{W(R)}{1+f(\varepsilon',s)W(R)}\\&=\Theta(\varepsilon'-\varepsilon)\Theta(\omega-\varepsilon'+\varepsilon)\tilde{W}(R|\varepsilon';s).\end{aligned} \tag{30}$$

From the explicit form of the transition probabilities [Eq. (3)], we conclude that for large $f(\epsilon)\nu$, that is, for large values of the parameter

$$\rho_c(\epsilon)=\ln[f(\epsilon)\nu], \tag{31}$$

$\tilde{W}$ acts like the function $\theta(\rho_c(\epsilon)-2\alpha R)/f(\epsilon)$, so that $\rho_c(\epsilon)$ is to be identified with the dimensionless critical hopping length.

Owing to the step functions in front of the transition probability, the energy integrations are restricted to intervals of length ω. Taking this fact into account, the integral equation for the calculation of the diffusion propagator takes the form

$$\begin{aligned}sF(\mathbf{q}|\varepsilon',\varepsilon)={}&\delta(\varepsilon'-\varepsilon)+\int_0^{\omega}d\varepsilon_1\{F(\mathbf{q}|\varepsilon',\varepsilon+\varepsilon_1)\\&\times\tilde{W}(\mathbf{q}|\varepsilon_1+\varepsilon;s)\mathcal{N}(\varepsilon)-F(\mathbf{q}|\varepsilon',\varepsilon)\\&\times\tilde{W}(0|\varepsilon;s)\mathcal{N}(-\varepsilon_1+\varepsilon)\}.\end{aligned} \tag{32}$$

Further, we assume that the diffusion propagator, the transition probability $\tilde{W}(R,\varepsilon;s)$, and the density of states $\mathcal{N}(\varepsilon)$ are slowly varying functions on intervals of length ω, so that the integrand can be expanded with respect to ε_1. Doing so, we obtain

$$\begin{aligned}sF(\mathbf{q}|\varepsilon',\varepsilon)={}&\delta(\varepsilon'-\varepsilon)+\omega\mathcal{N}(\varepsilon)[\tilde{W}(\mathbf{q}|\varepsilon;s)\\&-\tilde{W}(\mathbf{0}|\varepsilon;s)]F(\mathbf{q}|\varepsilon',\varepsilon)\\&+\frac{1}{2}\omega^2\frac{\partial}{\partial\varepsilon}[F(\mathbf{q}|\varepsilon',\varepsilon)\tilde{W}(\mathbf{q}|\varepsilon;s)\mathcal{N}(\varepsilon)].\end{aligned} \tag{33}$$

We terminate the expansion after the first derivative with respect to energy. This term describes the biased motion of the particle from sites of higher energy to sites of lower energy. For finite temperatures one would have to replace $F(\mathbf{q}|\varepsilon',\varepsilon)$ in the last term with $F(\mathbf{q}|\varepsilon',\varepsilon)+kT\partial F(\mathbf{q}|\varepsilon',\varepsilon)/\partial\varepsilon$ which would describe the energy diffusion current. At zero temperature there is no flow of energy from the phonon system to the particle. Therefore, we expect the additional term to be rather small, and neglect it.

To investigate transport properties, we restrict our attention to the long-wavelength limit. In this case, the elastic contribution yields the diffusion coefficient for particle diffusion. The term containing the derivative with respect to energy is finite for $q\rightarrow 0$. If we are interested only in the long-wavelength limit, here we can set $q=0$, since the remaining terms are of higher order with respect to q and $\omega\partial/\partial\varepsilon$. Since a nonzero momentum in this term couples particle diffusion to energy transport, this approximation corresponds to a decoupling of these two processes. Then, in the long-wavelength limit, we obtain

$$\begin{aligned}sF(\mathbf{q}|\varepsilon',\varepsilon)={}&\delta(\varepsilon'-\varepsilon)-D(\varepsilon,s)q^2F(\mathbf{q}|\varepsilon',\varepsilon)\\&+\frac{\partial}{\partial\varepsilon}[F(\mathbf{q}|\varepsilon',\varepsilon)v(\varepsilon,s)],\end{aligned} \tag{34}$$

where

$$D(\varepsilon,s)=-\frac{1}{2}\omega\boldsymbol{\nabla}_q^2\tilde{W}(q|\varepsilon;s)|_{\mathbf{q=0}} \tag{35}$$

and

$$v(\varepsilon,s)=\frac{1}{2}\omega^2\mathcal{N}(\varepsilon)\tilde{W}(0|\varepsilon;s). \tag{36}$$

An asymptotic calculation of the transport coefficients $D(\epsilon,s)$ and $v(\epsilon,s)$, for large $\rho_c(\epsilon,s)$, yields

$$v(\varepsilon,s)=\frac{1}{2}\frac{S(d)}{d}\omega^2\mathcal{N}(\varepsilon)\left[\frac{\rho_c(\varepsilon,s)}{2\alpha}\right]^d\nu\exp[-\rho_c(\varepsilon,s)] \tag{37}$$

and

$$D(\varepsilon,s)=\frac{1}{2}\frac{S(d)}{d(d+2)}\omega\mathcal{N}(\varepsilon)\left[\frac{\rho_c(\varepsilon,s)}{2\alpha}\right]^{d+2}\nu\exp[-\rho_c(\varepsilon,s)]. \tag{38}$$

Here $S(d)$ is the solid angle, and d is the spatial dimension. Thus $D(\varepsilon,s)$ and $v(\varepsilon,s)$ differ only in pre-exponential factors.

Equation (34) is easily solved. Its solution is

$$F(\mathbf{q}|\varepsilon',\varepsilon)=\frac{\Theta(\varepsilon'-\varepsilon)}{v(\varepsilon;s)}\exp\left[-\int_{\varepsilon}^{\varepsilon'}d\varepsilon_1\frac{s+D(\varepsilon_1;s)q^2}{v(\varepsilon_1;s)}\right]. \tag{39}$$

Now, at this stage, the validity of our quasielastic approximation requires that the second derivative terms are small compared to terms containing first derivatives with respect to energy. This requirement imposes the following restrictions on the transport coefficients v and D, the frequency s, and the momentum q:

$$\omega\left|\frac{1}{v}\frac{dv}{d\varepsilon}\right|\ll 1, \tag{40}$$

$$\omega\left|\frac{s}{v}\right|\ll 1, \tag{41}$$

$$q^2\omega\frac{D}{v}\ll 1. \tag{42}$$

The applicability of the quasielastic approximation was also discussed in Ref. 21. There it was concluded that this approximation should be inapplicable. To substantiate this statement, numerical calculations were invoked. However, in interpreting these data it has to be taken into account that they were obtained using a model that neglects a weighting of the transition probabilities according to the number of phonons emitted, so that hops between nearly isoenergetic sites were treated as likely as hops from the very top to the very bottom of the tail. Consequently the discussion in Ref. 21 applies only to systems with sufficiently strong electron-phonon interactions, but not to systems with weak electron-phonon interactions. For extraordinarily deep hops to contribute to the diffusion propagator they should be characteristic of an ensemble of electrons. This is, however, not expected and in the experimental data, e.g. on amorphous Si:H,[22] not observed. For these reasons the discussion in Ref. 21 does not apply to our model.

VI. INTERPRETATION OF THE TRANSPORT COEFFICIENTS

Before establishing a self-consistency equation, we first elaborate further on the physical content of our diffusion equation. Imagine that we have a particle initially located at $(\mathbf{R}_0,\varepsilon_0)$. Then the initial condition is $\langle n_o(\mathbf{R},\varepsilon)\rangle=\delta(\mathbf{R}-\mathbf{R}_0)\delta(\varepsilon-\varepsilon_0)$. According to our approximation, the motion of the charge carrier is composed of two contributions: particle diffusion between isoenergetic sites, and relaxation in energy space. Characteristics of these two processes are the mean-square displacement and the energy-relaxation rate. The mean-squared displacement is related to the coefficient $D(\epsilon,s)$, the diffusion coefficient for particle diffusion. The rate of energy relaxation is obtained from the derivative with respect to time of the mean energy, defined by

$$E(s)=\int d\rho'\varepsilon'\langle n(\rho',s)\rangle. \tag{43}$$

If we use our diffusion equation and perform an integration by parts, we obtain

$$sE(s)-\varepsilon_0=-\int d\varepsilon\, P_L(\varepsilon_0,\varepsilon;s)v(\varepsilon,s), \tag{44}$$

where

$$P_L(\varepsilon_0,\varepsilon;s)=F(q=0|\epsilon_0,\epsilon). \tag{45}$$

Therefore, in general, the time dependence of the mean energy is given by complicated integrals. These integrals simplify considerably in two limiting cases: in the absence of dispersive energy transport, and for an energy independent $v(\varepsilon,s)$. In the absence of dispersive energy transport, i.e., in the Markovian limit in which the transport coefficients are independent of s, the integrals can readily be calculated in the time representation. In this situation we simply obtain

$$P_L(\varepsilon_0,\varepsilon;t)=\Theta(\varepsilon_0-\varepsilon)\delta[\varepsilon_m(t)-\varepsilon], \tag{46}$$

where $\varepsilon_m(t)$ is defined by

$$t=\int_{\varepsilon_m}^{\varepsilon_0}\frac{d\varepsilon_1}{v(\varepsilon_1)}. \tag{47}$$

Therefore, we obtain

$$\frac{dE(t)}{dt}=\frac{d\varepsilon_m(t)}{dt}=-v[\varepsilon_m(t)]. \tag{48}$$

Consequently, v is the rate of energy relaxation, and $\varepsilon_m(t)$ is the instantaneous position of the particle in energy space. In general, however, in disordered systems the coefficient $v(\varepsilon,s)$ depends on s, so that *energy transport is dispersive*. In that case the integral in Eq. (44) can only be calculated easily if v is independent of energy. Then, owing to probability conservation,

$$\left(\frac{dE}{dt}\right)(s)=-\frac{v(s)}{s} \tag{49}$$

is obtained. Below we shall see that energy independent transport coefficients are obtained for constant density of states only.

VII. SELF-CONSISTENCY EQUATION

So far we have only investigated the consequences of the renormalization. In order to complete the approximation scheme, we still have to calculate the effective medium itself. Of course, we cannot calculate the effective medium exactly, since this would amount to finding an exact solution to the diffusion problem. Rather we shall try to calculate $f(\varepsilon,s)$ self-consistently.

The transport coefficients are properties of the Green's function F, the diffusion propagator. Thus, in order to find an equation for $f(\varepsilon,s)$, we should relate the transport coefficients to Π, the irreducible part of the diffusion propagator. The diffusion coefficient comprises only elastic contributions; however, in the relaxation problem a proper description of inelastic processes is vital. Therefore, in establishing a self-consistency equation, we focus on the energy relaxation rate v.

The equation for the diffusion propagator for $q=0$ is given by

$$sP_L(\varepsilon',\varepsilon;s)=\delta(\varepsilon'-\varepsilon)+P_L(\varepsilon',\varepsilon,s)\int d\varepsilon_1\,\Pi(\varepsilon_1,\varepsilon;s)+\frac{\partial P_L(\varepsilon',\varepsilon;s)}{\partial\varepsilon}\int d\varepsilon_1(\varepsilon_1-\varepsilon)\Pi(\varepsilon_1,\varepsilon;s). \tag{50}$$

If we compare Eq. (50) with Eq. (34) we deduce that

$$v(\varepsilon,s)=\int d\varepsilon_1(\varepsilon_1-\varepsilon)\Pi(\varepsilon_1,\varepsilon;s). \tag{51}$$

Now we decompose v, as defined in Eq. (51), into two parts, one part that contains only the effective-medium approximation (EMA) for Π,

$$v(\varepsilon,s)=\int d\varepsilon_1(\varepsilon_1-\varepsilon)\Pi|_{EMA}(\varepsilon_1,\varepsilon;s), \tag{52}$$

and a part $\delta v(\varepsilon,s)$, that contains the deviations $\tilde{F}$. Self-consistency requires that

$$\delta v(\varepsilon,s)=0. \tag{53}$$

$v(\varepsilon)$, as defined by Eq. (52), is in accordance with definition (36), taking into account Eq. (37) and the inequality $\omega f'(\varepsilon)/f(\varepsilon)\ll 1$ [Eq. (40)], owing to which contributions proportional to this parameter are negligible.

We now focus on the self-consistency equation (53). While Eq. (52) contains only the effective-medium contribution to the diffusion propagator, δv is a functional of the effective medium f and the deviation $\tilde{F}$. By construction, it is at least linear in $\tilde{F}$. If this equation could be solved exactly, an exact solution to the diffusion problem could be found within quasielastic accuracy. In practice this is not possible, and therefore we depend on further approximations. To simplify this equation, we take into account only the lowest-order contributions to this equation with respect to $\tilde{F}$, i.e., we linearize δv with respect to $\tilde{F}$ and require that the first-order contribution vanishes. This approach is quite close to the usual coherent-potential-approximation philosophy, in which vanishing of the t matrix is required in its lowest-order approximation. Using this procedure, we obtain the self-consistency equation

$$\frac{1}{2}\omega^2\mathcal{N}(\epsilon)f(\epsilon,s)\tilde{W}(0|\epsilon;s)=a\omega-\frac{sb}{\tilde{W}(0|\epsilon;s)\mathcal{N}(\epsilon)}, \tag{54}$$

where a and b are simply numbers. A detailed derivation of this equation is given in Appendix A.

In deriving the self-consistency equations, we have imposed further restrictions on the effective transition probabilities, which determine the range of its applicability. In terms of ρ_c, inequalities (40) and (41), used in the derivation, read (prime is derivative with respect to ε)

$$|\omega\rho_c'(\varepsilon,s)|\ll 1, \tag{55}$$

$$|\rho_c(\varepsilon,0)-\rho_c(\varepsilon,s)|\ll\rho_c(\varepsilon,0). \tag{56}$$

In addition, when calculating the integrals,

$$\rho_c(\varepsilon,s)\gg 1, \tag{57}$$

was used. A closed solution to the self-consistency equation can only be found in the limit $s=0$. In this limit, we obtain

$$\rho_c(\varepsilon,s=0)=\frac{2\alpha}{[\omega\mathcal{N}(\varepsilon)]^{1/d}}\left[\frac{2da}{S(d)}\right]^{1/d}, \tag{58}$$

where $S(d)$ is the solid angle in d dimensions.

For s satisfying Eq. (56), Eq. (54) can be cast in the form

$$[\rho_c(\varepsilon,0)-\rho_c(\varepsilon,s)]\exp[\rho_c(\varepsilon,0)-\rho_c(\varepsilon,s)]=\frac{s}{\Omega(\varepsilon)}, \tag{59}$$

where

$$\Omega(\varepsilon)=\frac{2da}{b\omega\rho_c(\varepsilon,0)}v(\varepsilon,0). \tag{60}$$

According to Eq. (59), the critical hopping length decreases with increasing frequency. Note that the structure of Eq. (59) for calculation of the dispersion of the critical hopping length obtained here agrees completely with that obtained for the critical hopping length in calculating the equilibrium conductivity.[17,20]

The dispersion of the transport coefficients is determined completely by the dispersion of the critical hopping length. For small s the frequency dependent pre-exponential factors can be ignored, so that from Eq. (59) explicit equations for the transport coefficients can be obtained. They are given by

$$\frac{D(\varepsilon,s)}{D(\varepsilon,0)}\ln\frac{D(\varepsilon,s)}{D(\varepsilon,0)}=\frac{s}{\Omega(\varepsilon)}, \tag{61}$$

$$\frac{v(\varepsilon,s)}{v(\varepsilon,0)}\ln\frac{v(\varepsilon,s)}{v(\varepsilon,0)}=\frac{s}{\Omega(\varepsilon)}. \tag{62}$$

The formal solution of these equations is given by Lambert's W function $\mathcal{W}(z)$, defined by the equation $z=\mathcal{W}(z)\exp\mathcal{W}(z)$. Using Lambert's function, we can write

$$D(\varepsilon,s)=D(\varepsilon,0)\exp\mathcal{W}[s/\Omega(\varepsilon)], \tag{63}$$

$$v(\varepsilon,s)=v(\varepsilon,0)\exp\mathcal{W}[s/\Omega(\varepsilon)]. \tag{64}$$

VIII. CONSTANT DENSITY OF STATES

A constant density of states, although of not much physical relevance, gives us a unique opportunity to study purely dispersive energy transport. Here both $\Omega(\varepsilon)$ and $v_0=v(\varepsilon,0)$ are independent of energy. Consequently, the energy distribution function $P_L(\epsilon_0,\epsilon;s)$ [Eq. (45)], can readily be calculated. The calculation yields

$$P_L(\varepsilon_0-\varepsilon,s)=\frac{\Theta(\varepsilon_0-\varepsilon)}{v(s)}\exp\left[-\frac{s(\varepsilon_0-\varepsilon)}{v(s)}\right]. \tag{65}$$

The time dependence of this function can be obtained by inverse Laplace transformation. Using Eq. (62) to change the integration variable from s to $y=v/v_0$, the inverse Laplace transform of Eq. (65) may be written

$$P_L(\varepsilon_0-\varepsilon,t)=\frac{\Omega}{v_0}\Theta(\varepsilon_0-\varepsilon)\int_C\frac{dy}{2\pi i}\,\frac{\ln y+1}{y}\times\exp\left[\Omega t y\ln y-\frac{\Omega}{v_0}(\varepsilon_0-\varepsilon)\ln y\right], \tag{66}$$

with a properly chosen integration contour C. At large enough t, that is for $\Omega t\gg 1$, this expression, using the saddle-point method, simply gives the Gaussian packet

$$P_L(\varepsilon_0-\varepsilon,t)\approx\Theta(\varepsilon_0-\varepsilon)\frac{\Omega}{v_0}\,\frac{1}{2\sqrt{\pi\Omega t}}\times\exp\left[-\frac{(\varepsilon_0-\varepsilon-v_0t)^2}{4v_0^2t/\Omega}\right]. \tag{67}$$

According to Eq. (49), the energy-relaxation rate is

$$\frac{dE(t)}{dt}=-\int_C\frac{ds}{2\pi i}\,\frac{v(s)}{s}e^{st}. \tag{68}$$

Again, the time dependence of the energy-relaxation rate can be calculated using asymptotics. Doing so, we obtain

$$\frac{dE(t)}{dt}\approx -v_0\begin{cases}1+\sqrt{\dfrac{e}{2\pi}}\dfrac{1}{(\Omega t)^{3/2}}\exp(-\Omega t/e) & \text{as } \Omega t\gg 1\\[2ex] \dfrac{\sqrt{2\pi}}{e\Omega t}\{\ln[e/(\Omega t)]\}^{-2} & \text{as } \Omega t\ll 1.\end{cases} \tag{69}$$

Note that the problem of energy relaxation in the case of constant density of states [$v(\varepsilon,s)$ is independent of ε] is completely equivalent to one of the non-Markovian charge transport in strong electric fields E, when the diffusion (described by the second derivative with respect to the coordinates) is totally neglected. One has only to replace $v(s)\rightarrow u(s)E$, $u(s)$ being the mobility.[16,17] In the context of dispersive particle transport, the regimes $\Omega t\ll 1$ and $\Omega t\gg 1$ are the regimes of anomalous and normal ''diffusion,'' respectively. The main difference between them is that while the sites are usually distributed homogeneously in space, the density of states is usually an increasing function of energy.

IX. EXPONENTIAL DENSITY OF STATES

A. The saddle-point approximation and its breakdown for large times

Calculation of the time dependence of the energy distribution function for an arbitrary density of states on the basis of Eqs. (34)–(36), (61), and (62) turns out to be a quite intricate problem. A tool that can be utilized in tackling the problem is the saddle-point approximation. How to apply the saddle-point approximation for the calculation of the quantities of interest for an arbitrary density of states is shown in Appendix B. Below we focus on the exponential density of states, which is relevant, e.g., to amorphous Si:H.

We assume that the density of states is given by

$$\mathcal{N}(\varepsilon)=\mathcal{N}_0\exp\left(3\frac{\varepsilon}{\Delta}\right). \tag{70}$$

To simplify the notations, we use the abbreviations

$$\overline{\omega}=\frac{b\omega}{2da}, \tag{71}$$

$$\overline{\nu}=\frac{2da^2}{b}\nu. \tag{72}$$

Then Eqs. (37), (58), and (60) can be cast into the forms

$$v(\varepsilon,0)\equiv v(\varepsilon)=\overline{\omega\nu}\exp[-\rho(\varepsilon)], \tag{73}$$

$$\Omega(\varepsilon)=\frac{\overline{\nu}}{\rho(\varepsilon)}\exp[-\rho(\varepsilon)], \tag{74}$$

$$\rho(\varepsilon)=\rho_c(\varepsilon,0)=A\exp\left(-\frac{\varepsilon}{\Delta}\right), \tag{75}$$

where

$$A=\frac{2\alpha}{[\omega\mathcal{N}(0)]^{1/d}}\left[\frac{2da}{S(d)}\right]^{1/d}. \tag{76}$$

Using these equations it follows from the formulas derived in Appendix B that the time dependence of the mean-squared deviation and the time dependence of the mean energy are given by

$$\sigma^2(\varepsilon_m,\varepsilon_0)\equiv\sigma^2(t,\varepsilon_0)\simeq\overline{\omega}\Delta\left[1-\frac{t_0^2}{(t+t_0)^2}\right], \tag{77}$$

$$\varepsilon_m(t) \simeq -\Delta \ln\left[\frac{1}{A}\ln\left(\frac{\bar{\omega}}{\Delta}\bar{\nu}(t+t_0)\right)\right], \tag{78}$$

where

$$t_0(\varepsilon_0) = \frac{\Delta}{\bar{\nu}\rho(\varepsilon_0)v(\varepsilon_0)}. \tag{79}$$

Note that, according to the saddle-point approximation, the distribution is Gaussian. Furthermore, the dispersion is constant for $t \gg t_0$. Consequently, for time scales in line with the applicability of the saddle-point approximation, the motion of the energy packet is ''solitonlike,'' that is, the packet moves without distortion.

The applicability condition for the saddle point [Eq. (B13)] requires that

$$\frac{\bar{\omega}}{\Delta}\rho^2(\varepsilon_m) \ll 1. \tag{80}$$

Because $\rho(\varepsilon_m)$ grows with t, the saddle-point method, and consequently all the results of this section, becomes invalid for sufficiently large t. The physical meaning of condition (80) is the following: at $t \gg t_0$ the width of the electron's energies distribution is $\delta\varepsilon = \sqrt{\bar{\omega}\Delta}$. Because $\rho \sim \exp(-\varepsilon/\Delta)$, the variation of ρ is $\delta\rho/\rho = \sqrt{\bar{\omega}/\Delta}$, and because $v \sim \exp(-\rho)$, we have $\delta v/v = \delta\rho = \sqrt{\bar{\omega}\rho^2/\Delta}$. Thus condition (80) is just that the variation of the electron's ''velocity'' across the distribution is smaller than the velocity itself.

B. Form of the distribution for large times

Even if the initial conditions are such that condition (80) is fulfilled, and a Gaussian distribution is formed, at some moment of time $\rho_m \equiv \rho(\varepsilon_m)$ becomes of the order of $\sqrt{\Delta/\bar{\omega}}$, which definitely should result in some deviation of the distribution function from its symmetric, Gaussian form. For the exponential density of states, the distribution function in the Laplace representation, using Eqs. (64) and (73)–(75), may be written

$$P_L(\varepsilon_0,\varepsilon;s) = -\frac{1}{s}\rho\frac{\partial}{\partial\rho}\exp\left[-\frac{1}{\bar{\omega}}\int_{\rho_0}^{\rho}dx\frac{\mathcal{W}(sxe^x)}{x^2}\right], \tag{81}$$

where we have set $\bar{\nu}=1$ and $\Delta=1$ by appropriate choice of time and energy units. Under the same conditions which were used in the derivation of the above formula, the following approximation is valid:

$$\int_{\rho_0}^{\rho}dx\frac{\mathcal{W}(sxe^x)}{x^2} \equiv G(s,\rho)-G(s,\rho_0);$$

$$G(s,\rho) = \frac{1}{\rho^2}\mathcal{W}(s\rho e^\rho)\left[1+\frac{1}{2}\mathcal{W}(s\rho e^\rho)\right]+O\left(\frac{\mathcal{W}}{\rho^3},\frac{\mathcal{W}^3}{\rho^3}\right). \tag{82}$$

When calculating the distribution function at sufficiently large times, the characteristic values of s, giving the main contribution in the inverse Laplace integral, become so small that one can suppose $|s|\rho_0\exp(\rho_0) \ll 1$ and $G(s,\rho_0) \approx s\rho_0^{-1}\exp(\rho_0)$. Under this condition the distribution function approaches initial conditions independence of shape: $P(\varepsilon_0,\varepsilon;t) \to \phi[\varepsilon, t+t_0(\varepsilon_0)]$, $t_0(\varepsilon_0) = (\bar{\omega}\rho_0)^{-1}\exp\rho_0$,

$$\begin{aligned}\phi(\varepsilon,t) &= \int_{-i\infty}^{+i\infty}\frac{ds}{2\pi i}\phi_L(\varepsilon,s)e^{st}\\ &= -\int_{-i\infty}^{+i\infty}\frac{ds}{2\pi is}\rho\frac{\partial}{\partial\rho}\\ &\quad\times\exp\left\{st-\frac{1}{\bar{\omega}\rho^2}\mathcal{W}(s\rho e^\rho)\left[1+\frac{1}{2}\mathcal{W}(s\rho e^\rho)\right]\right\}.\end{aligned} \tag{83}$$

Now we consider the moments of the distribution at large times. In the Laplace-representation the moments are defined by the equation

$$\begin{aligned}\chi_{nL}(s) &= \int_0^\infty dt\, e^{-st}\langle\varepsilon^n\rangle(t) = \int_{\rho_0}^\infty\frac{d\rho}{\rho}(-\ln\rho)^n P_L(\varepsilon_0,\varepsilon,s)\\ &\to e^{st_0}\int\frac{d\rho}{\rho}(-\ln\rho)^n\phi_L(\varepsilon,s).\end{aligned} \tag{84}$$

We omit the irrelevant time shift multiple e^{st_0} in further discussions.

For $\bar{\omega}\rho^2 \gg 1$, moments can be calculated as an expansion in powers of the small parameter $1/(\sqrt{\bar{\omega}}\rho)$. Doing so, we obtain for the first moment, the mean energy,

$$\begin{aligned}\chi_1(t) = \langle\varepsilon\rangle(t) &\approx -\Delta\ln\frac{1}{A}\ln(\tilde{b}\sqrt{\bar{\omega}}t)-\sqrt{\frac{\pi\bar{\omega}}{2}}\\ &\quad+\sqrt{\frac{\pi}{8\bar{\omega}}}\frac{1}{\ln^2(\tilde{b}\sqrt{\bar{\omega}}t)}+\cdots,\end{aligned} \tag{85}$$

where $\tilde{b} = \sqrt{2}e^{-1+\gamma/2}$, and γ is Eulers's constant. For the distribution's dispersion $\sigma^2(t) = \chi_2(t)-\chi_1^2(t)$,

$$\sigma^2(t) = \left(2-\frac{\pi}{2}\right)\bar{\omega}+(1+\ln 2)\frac{\sqrt{2\pi\bar{\omega}}}{\ln(\sqrt{\bar{\omega}}t)}+\cdots, \tag{86}$$

is obtained, and for the third central moment $\mu_3(t) = \langle[\varepsilon-\langle\varepsilon\rangle(t)]^3\rangle$ we have

$$\mu_3(t) = -(\pi-3)\sqrt{\frac{\pi}{2}}\bar{\omega}^{3/2}+3[\pi(1-\ln 2)-1]\frac{\bar{\omega}}{\ln(\sqrt{\bar{\omega}}t)}. \tag{87}$$

Figures 1, 2, and 3 show the mean-energy, the mean square deviation, and the dimensionless coefficient of asymmetry.

Moving along the same line the whole distribution function can be reconstructed at large times from all of its moments. Doing so, we find that at large times and small $\bar{\omega}$ the distribution function approaches

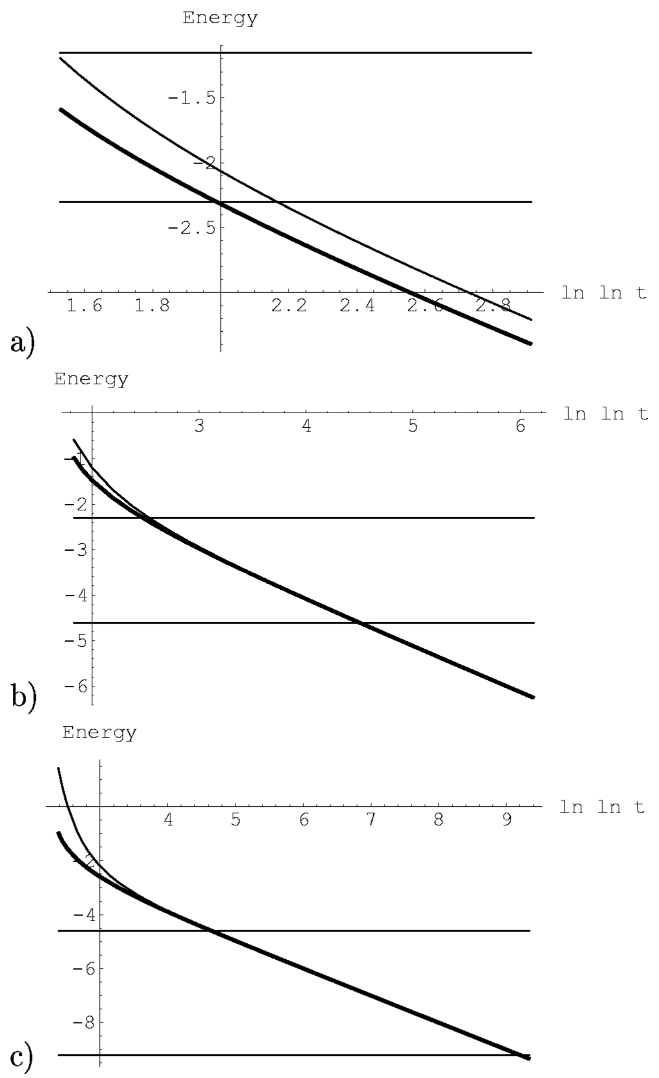

FIG. 1. Mean energy plotted as a function of $\ln\ln t$ for (a) $\overline{\omega}/\Delta=10^{-1}$, $100<t<10^8$; (b) $\overline{\omega}/\Delta=10^{-2}$, $5\times10^2<t<10^{200}$; and (c) $\overline{\omega}/\Delta=10^{-4}$, $5\times10^4<t<10^{5000}$. (Thick line, numerical calculation of the integrals; thin line, asymptotical calculation of the integrals.)

$$\phi(\varepsilon,t)=\frac{\partial}{\partial\varepsilon}\begin{cases}\exp\left[-\frac{(\varepsilon-\varepsilon_t)^2}{2\overline{\omega}}\right], & \varepsilon<\varepsilon_t\\ 1, & \varepsilon>\varepsilon_t,\end{cases}\qquad \varepsilon_t=\ln\ln t. \tag{88}$$

In Fig. 4 some plots of the distribution function (83), compared with its asymptotic form [Eq. (88)], are shown.

C. Initial stage of the evolution

What happens at the initial stage of the evolution depends much on the initial condition. At $t=0$ it was supposed that $P(\varepsilon,\varepsilon_0;t=0)=\delta(\varepsilon-\varepsilon_0)$, so that all charge carriers have the same energy ϵ_0. To gain some insight into the initial stage of the evolution, we study the behavior of the derivative

$$P_1(\epsilon_0,t)=\left.\frac{\partial P_L(\epsilon_0,\epsilon;t)}{\partial\epsilon}\right|_{\epsilon=\epsilon_0}. \tag{89}$$

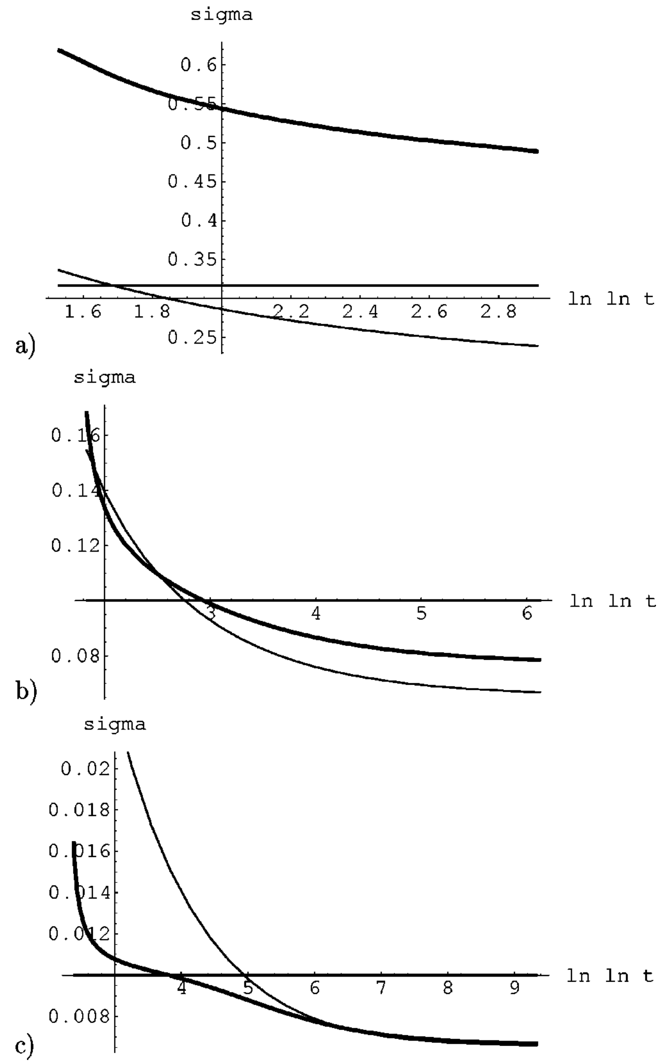

FIG. 2. Mean-square deviation σ plotted as a function of $\ln\ln t$. The values of the parameters are the same as in Fig. 1.

P_1 is expected to change sign in the process of unsticking. For small times P_1 should be positive, so that the distribution still sticks at $\epsilon=\epsilon_0$. In the course of time, the distribution separates from its initial condition. If P_1 is negative the distribution is unstuck, so that the instant of unsticking is related to the zero of P_1. Using formulas (B1), (64), and (73)–(75), we obtain

$$P_1(\rho_0,t)\approx\frac{1}{(\overline{\omega}\rho_0)^2}\int_C\frac{dz}{2\pi i}\left[z(1+z)-\frac{\overline{\omega}}{\Delta}\rho_0^2\right]\exp t(\tau z e^z). \tag{90}$$

Here $\rho_0=A\exp(-\varepsilon_0/\Delta)$ and $\tau=(\overline{\nu}t/\rho_0)e^{-\rho_0}$. In Eq. (90) the new integration variable $z=\mathcal{W}[(s/\overline{\nu})\rho_0e^{\rho_0}]$ was introduced for s, and terms proportional to $(\overline{\omega}/\Delta)\rho\ll1$ were neglected.

For $\tau\gg1$ this integral may be readily evaluated in the saddle-point approximation, which yields

$$P_1(\rho,t)\approx-\frac{1}{\overline{\omega}\Delta}\sqrt{\frac{e}{2\pi\tau}}\exp\left(-\frac{\tau}{e}\right). \tag{91}$$

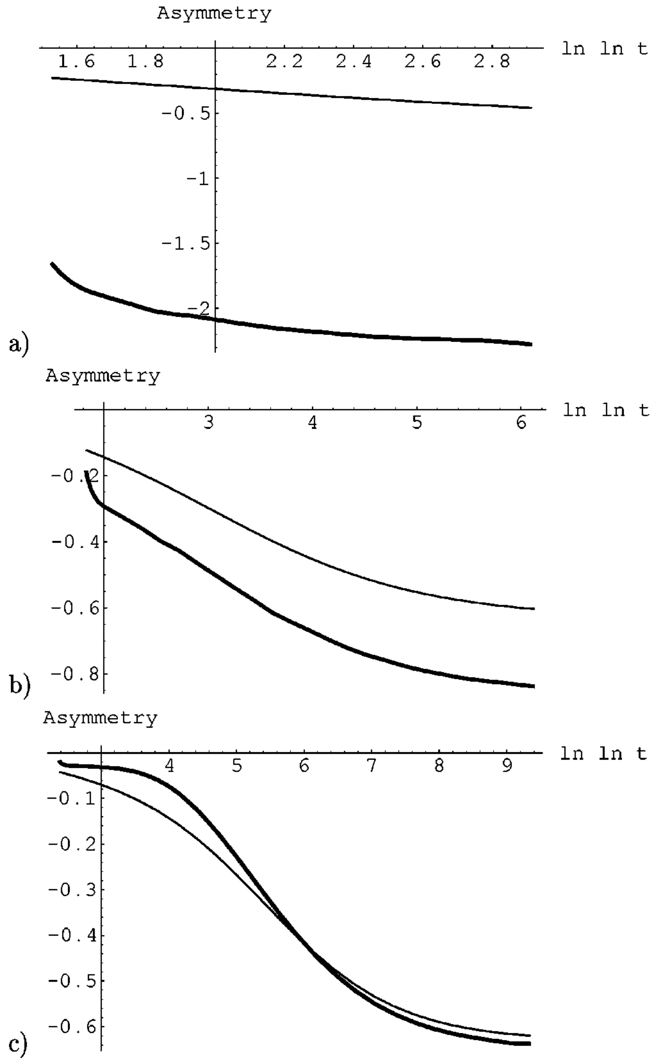

FIG. 3. Coefficient of asymmetry μ_3/σ^3 plotted as a function of $\ln\ln t$. The values of the parameters are the same as in Fig. 1.

When $\tau\ll 1$ one has to perform an integration by parts first, which gives

$$P_1(\rho,t)=\frac{1}{\bar{\omega}^2\rho^2\tau}\int_C\frac{dz}{2\pi i}\times\left[z-1-\frac{\bar{\omega}\rho^2}{\Delta(1+z)}-\frac{\bar{\omega}\rho^2}{\Delta(1+z)^2}\right]\exp(\tau z e^z-z).$$

Then the saddle point-approximation yields

$$P_1(\rho,t)\approx\frac{e}{\sqrt{2\pi\bar{\omega}^2}\rho^2}(z_c^2-1-\bar{\omega}\rho^2),\quad z_c\approx\ln\frac{1}{\tau}-\ln\ln\frac{1}{\tau}. \tag{92}$$

At sufficiently small t the above expression is positive, which means that the distribution is ''stuck'' near $\varepsilon=\varepsilon_0$, that is, it monotonously decreases as $\varepsilon_0-\varepsilon>0$ increases. At some $\tau=\tau_0$, P_1 changes sign. If $(\bar{\omega}/\Delta)\rho_0^2\gg 1$, this corresponds to $z_{c0}\approx\ln(1/\tau_0)\approx\sqrt{\bar{\omega}/\Delta}\rho_0$, otherwise $\tau_0\sim 1$. This corresponds just to the instant of time when the distribution separates from the initial point—its maximum is at $\varepsilon<\varepsilon_0$. For $\tau\ll\tau_0$ the width of the distribution on the energy scale may be estimated as

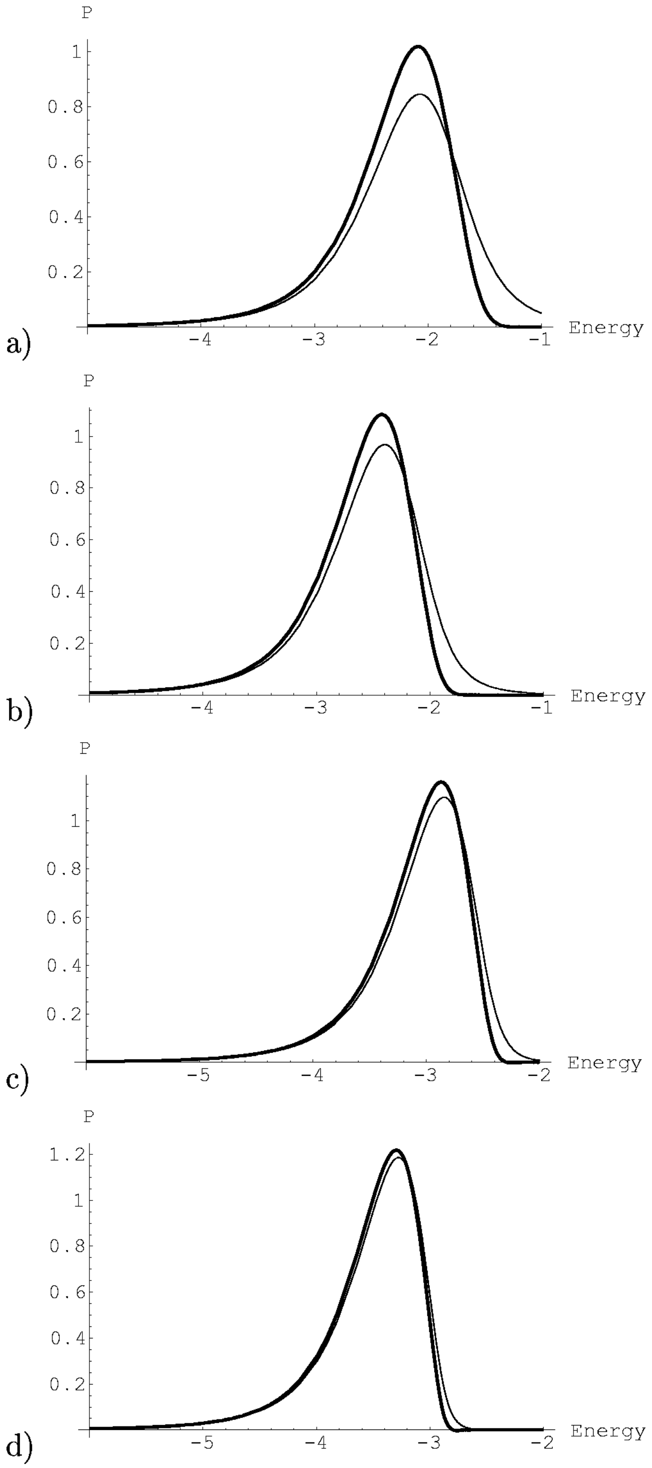

FIG. 4. Plots of the energy distribution function at $\bar{\omega}/\Delta=0.1$ and (a) $t=10^3$, (b) $t=10^4$, (c) $t=10^6$, and (d) $t=10^9$. (Thick line, numerical calculation of the integrals; thin line, asymptotical calculation of the integrals).

$$\Delta\varepsilon(t)\sim\frac{P_0(\rho,t)}{P_1(\rho,t)}, \tag{93}$$

where $P_0(\rho_0,t)=P_L(\epsilon_0,\epsilon_0;t)$. Using formulas (B1), (64), and (73)–(75), we obtain

$$P_0(\varepsilon_0,t)=\frac{1}{\overline{\omega}\rho_0\tau}\int_C\frac{dz}{2\pi i}\exp(\tau z e^z-z). \tag{94}$$

The integral may be evaluated using the saddle-point method. The saddle-point equation, $f'(z)=\tau(z+1)e^z-1=0$, gives us the saddle-point value $z_c=\mathcal{W}(e/\tau)-1$, that is $z_c\approx\ln(1/\tau)-\ln\ln(1/\tau)$ at $\tau\ll 1$, and $z_c\approx e/\tau-1$ as $\tau\gg 1$. To ensure the correctness of the saddle-point approximation, one should require the parameter $|f'''(z_c)|^2/|f''(z_c)|^3=|(3+z_c)^2/(2+z_c)^3|$ to be small. While this is true at $\tau\gg 1$, this parameter appears to be ≈ 1 at $\tau\ll 1$. Therefore, the result for $\tau\ll 1$ is correct up to a multiple of order 1 only. In the saddle-point approximation we have

$$P_0(\rho,t)\approx\begin{cases}\dfrac{1}{\sqrt{2\pi\overline{\omega}\rho}}\left(\dfrac{e}{\tau}\right)^{3/2}\exp\left(-\dfrac{\tau}{e}\right) & \text{as } \tau\gg 1\\[2ex] \dfrac{e}{\sqrt{2\pi\overline{\omega}\rho}}\left(\ln\dfrac{e}{\tau}-\ln\ln\dfrac{e}{\tau}\right) & \text{as } \tau\ll 1.\end{cases} \tag{95}$$

Consequently,

$$\Delta\epsilon(t)\approx\frac{\overline{\omega}\rho_0}{z_c}\approx\frac{\overline{\omega}\rho}{\ln(1/\tau)}. \tag{96}$$

This formula is valid if $\ln(1/\tau)\ll\rho$, up to times corresponding to either $\tau\sim 1$ if $(\overline{\omega}/\Delta)\rho_0^2\ll 1$, or $\ln(1/\tau)\approx\sqrt{\overline{\omega}/\Delta}\,\rho_0$ otherwise. In the former case the distribution width becomes of the order of $\overline{\omega}\rho$ just before it separates from the initial point. After this, the distribution, as shown in a previous subsection, widens to $\sqrt{\overline{\omega}\Delta}$, and the Gaussian packet moves downward, until its center reaches the value corresponding to $\rho\sim\sqrt{\Delta/\overline{\omega}}$. In the latter case the packet's width is $\sim\sqrt{\overline{\omega}\Delta}\ll\overline{\omega}\rho$ at the very moment of its ''unsticking.'' In both cases, to consider subsequent evolution, one has to use some other approximation instead of the saddle-point one.

X. CONCLUSIONS

In this paper we have presented an effective method which permits an investigation of relaxation phenomena of localized charge carriers far from equilibrium due to phonon-assisted hopping at zero temperature. From the point of view of the formalism, the main equations are those for the calculation of the diffusion propagator [Eqs. (34), (35), and (36)], and Eqs. (60), (61), and (62), which determine the dispersion of the transport coefficients. These equations show that both particle transport and energy transport are dispersive. Equations (61) and (62) lead to a strong dependence of the diffusion constant $D(\epsilon;s)$ and the rate of energy relaxation $v(\epsilon;s)$ on frequency s even for low frequencies. To our knowledge, these equations have not been derived in the literature so far, for systems far from equilibrium. In fact, in the literature, mainly frequency-independent transport coefficients can be found (see, e.g., Ref. 21 and references therein). The strong dependence of the transport coefficients on s results in a non-Markovian equation for the calculation of the diffusion propagator [Eq. (34)]. This distinguishes our equations from the Markovian integral equations used in Refs. 11–13. The latter equations can, in principle, be obtained from our leading Eq. (11) by neglecting statistical correlation, so as to average every factor independently.

Using our effective-medium method we have investigated the relaxation of charge carriers in band tails at $T=0$. According to our results, energy relaxation is connected with dispersive transport. Even if we have no real diffusion in the system, since the temperature is zero, we have some spreading of the energy distribution with time. For a constant density of states the situation is completely equivalent to that of an electron's motion in a disordered system, subjected to an electric field.[16] The main difference between the above-mentioned problem and the energy relaxation is that in the former case the sites are distributed homogeneously in space, while in the energy-relaxation problem the density of states is a decaying function of energy for most physical systems. In this situation we arrive at a picture in which particles of a packet with lower energies move slower than particles with higher energy. This leads to the opposite tendency: at first, there is a slowing of the packet spreading. The time dependence of the dispersion, being linear at the first stage of the evolution, slows down later. Later on, different possibilities exist, depending on the particular energy dependence of the density of states.

We performed a detailed investigation for the exponential density of states for two time regimes. If the variation of the rate v in energy space across the distribution is smaller than the rate itself, that is, if $\omega\rho^2/\Delta\ll 1$, we arrive at the situation of a packet, moving steadily down along the energy axis without deformation. Dispersion becomes time independent [see Eq. (77)]. However, since the particles are sinking down this condition is violated as time goes by. The parameter $\omega\rho^2/\Delta$, being small at the first stage of the evolution, becomes large. When this parameter is larger than 1, the steady motion condition is again violated. The packet, previously of Gaussian form, undergoes some restructuring to another non-Gaussian stable form, with its width lower than before by some numerical factor of order 1 [Eq. (86)]. In Figs. 3 and 4 it is clearly seen that the packet becomes non-Gaussian. This result remains valid until the very moment that the quasielasticity conditions break down $(\omega\rho/\Delta\approx 1)$.

For the exponential density of states we have found the packet to move as $\ln\ln t$, i.e., its motion is strongly slowing down with time; roughly speaking, the packet almost stops. This type of behavior may be called ''glassy,'' because the overall time scale for packet evolution (governed by the exponential function of the large parameter Δ/ω), becomes huge.

The main simplification used in our paper is the quasielastic approximation. This approximation relies on the smallness of the upper bound for energy transferred to the phonon system in one hop. For localized electrons, this upper bound can be much smaller than the Debye energy of the host material, since not all phonons can interact with localized electrons equally well. For localized electrons the electron-phonon coupling constant approaches zero for phonons with a wave vector $q>2\alpha$. Thus highly energetic phonons are less effective. Only phonons with energies $\omega<\omega_D 2\alpha a$, where a is the lattice constant of the host material, are effective. Furthermore, in disordered systems the highly energetic acoustical phonons are localized, and thus need not contribute to transport. Nevertheless, the question of whether the quasielastic approximation is applicable depends very much on the material of interest. If, however, we compare our result for the time dependence of the mean energy [Eq. (78)] in an exponential density of states with those existing in the literature,[1] we also find agreement for $\omega/\Delta\approx 1$, which indicates that our results are, at least qualitatively, of wider validity. Unfortunately, to our knowledge, in the literature there are no other results on the width of the energy distribution available with which to compare.

From our point of view the main open question remaining is how the results obtained for the exponential density of states may be generalized for other types of energy dependencies. One can imagine, e.g., that for densities of states decaying with a decreasing energy that is slower than the exponential energy, the dispersion grows, according to some sublinear law. Also, in the opposite case, for a density of states decaying faster than the exponential energy, we would possibly have a dispersion tending to zero for large times.

However, it should also be mentioned that it is not completely clear under which situation zero-temperature results can be applied to systems at finite temperature. In our approach, we assumed that $T=0$. However, in a real system, when the carriers are sinking down, the criterion for the temperature to be treated as zero is violated at every finite value of T at some moment of time, since the contribution of hops to sites with higher-energy values becomes more and more comparable with the contribution of one of the downward hops. Therefore, the consideration of temperature becomes vital. Work in this direction is in progress.

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APPENDIX A: DERIVATION OF SELF-CONSISTENCY EQUATION

Here we derive the self-consistency equation (54). In order to work out the first-order contribution to the self-consistency equation (53) explicitly, we first-need the first-order correction to Π. This is given by

$$\Pi^{(1)}(\rho',\rho;s)=\int d\rho_1\,d\rho_2\,d\rho_3\,\mathcal{N}(\varepsilon_3)\tilde{w}_{\rho_3}(\rho',\rho_1;s)\times\tilde{F}(\rho_1,\rho_2)\tilde{w}_{\rho_3}(\rho_2,\rho;s). \tag{A1}$$

If we insert the expression for $\tilde{w}_{\rho_3}$, we obtain

$$\begin{aligned}\Pi^{(1)}(\rho',\rho;s)=\int d\rho_1\{&\mathcal{N}(\varepsilon)\tilde{W}(\rho',\rho;s)\\&\times[\tilde{F}(\rho,\rho_1)-\tilde{F}(\rho',\rho_1)]\tilde{W}(\rho_1,\rho;s)\\&-\mathcal{N}(\varepsilon_1)\tilde{W}(\rho',\rho_1;s)\\&\times[\tilde{F}(\rho_1,\rho)-\tilde{F}(\rho',\rho)]\tilde{W}(\rho,\rho_1;s)\}.\end{aligned} \tag{A2}$$

Thus, at this stage, the self-consistency equation takes the form

$$\begin{aligned}0=\int d\mathbf{q}\,d\varepsilon'\,d\varepsilon_1(\varepsilon'-\varepsilon)&\times\{\mathcal{N}(\varepsilon)\tilde{W}(0|\varepsilon',\varepsilon;s)\tilde{F}(q|\varepsilon,\varepsilon_1)\tilde{W}(q|\varepsilon_1,\varepsilon;s)\\&-\mathcal{N}(\varepsilon)\tilde{W}(q|\varepsilon',\varepsilon;s)\tilde{F}(q|\varepsilon',\varepsilon_1)\tilde{W}(q|\varepsilon_1,\varepsilon;s)\\&-\mathcal{N}(\varepsilon_1)\tilde{W}(0|\varepsilon',\varepsilon_1;s)\tilde{F}(q|\varepsilon_1,\varepsilon)\tilde{W}(q|\varepsilon,\varepsilon_1;s)\\&+\mathcal{N}(\varepsilon_1)\tilde{W}(q|\varepsilon',\varepsilon_1;s)\tilde{F}(q|\varepsilon',\varepsilon)\tilde{W}(q|\varepsilon,\varepsilon_1;s)\}.\end{aligned} \tag{A3}$$

In order to eliminate $\tilde{F}$, we have to replace $\tilde{F}(q|\varepsilon',\varepsilon)=F(q|\varepsilon',\varepsilon)-f(\varepsilon,s)\delta(\varepsilon'-\varepsilon)$, where F is the effective-medium approximation of the diffusion propagator. Let us first work out the local contribution. Replacing $\tilde{F}$ by $f\times\delta$, the first and the third terms of Eq. (A3) cancel each other. The fourth term is zero, taking into account that the δ function of the effective medium is multiplied by its argument. Thus, when $\tilde{F}$ is replaced by $f(\varepsilon,s)\delta(\varepsilon'-\varepsilon)$, only the second term of Eq. (A3) survives. To simplify this term we take into account that

$$\tilde{W}(q|\epsilon;s)=\tilde{W}(0|\epsilon,s)\,\phi\left(\frac{q\rho_c(\epsilon,s)}{2\alpha}\right), \tag{A4}$$

where ϕ is a dimensionless function. Consequently, we obtain

$$\begin{aligned}&-\int d\mathbf{q}\,d\varepsilon'(\varepsilon'-\varepsilon)\mathcal{N}(\varepsilon)f(\varepsilon',s)[\tilde{W}(q|\varepsilon',\varepsilon;s)]^2\\&\approx-\int d\mathbf{q}[\tilde{W}(\mathbf{q}|\varepsilon,s)]^2\frac{\omega^2}{2}f(\varepsilon,s)\mathcal{N}(\varepsilon)\\&=-\frac{1}{2}\omega^2\left(\frac{2\alpha}{\rho_c(\epsilon,s)}\right)^d\mathcal{N}(\epsilon)f(\epsilon,s)\tilde{W}^2(0|\epsilon,s)\\&\quad\times\int d^dx\,\phi^2(x).\end{aligned} \tag{A5}$$

Here terms proportional to $\omega f^{-1}(\varepsilon,s)df(\varepsilon,s)/d\varepsilon\ll 1$ have been neglected.

Let us now focus on the contribution of the regular part of the diffusion propagator to the self-consistency equation. Owing to the step functions in F and $\tilde{W}$, the first and third terms in Eq. (A3) are zero, when $\tilde{F}$ is replaced by F. Thus we are left with

$$\begin{aligned}\int d\mathbf{q}\,d\varepsilon'\,d\varepsilon_1(\varepsilon'-\varepsilon)&\times\{-\mathcal{N}(\varepsilon)\tilde{W}(q|\varepsilon',\varepsilon;s)F(q|\varepsilon',\varepsilon_1)\tilde{W}(q|\varepsilon_1,\varepsilon;s)\\&+\mathcal{N}(\varepsilon_1)\tilde{W}(q|\varepsilon',\varepsilon_1;s)F(q|\varepsilon',\varepsilon)\tilde{W}(q|\varepsilon,\varepsilon_1;s)\}.\end{aligned} \tag{A6}$$

The range of integration in Eq. (A6) is determined by the step functions in F and $\tilde{W}(q|\varepsilon',\varepsilon;s)=\theta(\varepsilon'-\varepsilon)\theta(\omega-\varepsilon'+\varepsilon)\tilde{W}(q|\varepsilon';s)$. Owing to these step functions, the energy integrations extend at most over intervals of length ω, so that the quasielastic approximation can again be applied to the effective-transition probabilities and the density of states.

The diffusion propagator entering Eq. (A6), however, cannot be dealt with in this way, since for arbitrary q the derivatives of the diffusion propagator are not small compared to the diffusion propagator itself. Therefore, another procedure is needed. To simplify this expression further we consider Eq. (29). If terms small with respect to $\omega N'/N$ and $\omega\tilde{W}'(q|\epsilon,s)/\tilde{W}(q|\epsilon,s)$ are neglected, the function

$$\Phi(x|y',y)=\omega^2F\left(x\frac{2\alpha}{\rho_c(\omega y,s)}\middle|\omega y',\omega y\right)\tilde{W}(\omega y,s)\mathcal{N}(\omega y), \tag{A7}$$

can be introduced, that satisfies the equation

$$\frac{s}{\tilde{W}(\omega y,s)\mathcal{N}(\omega y)\omega}\Phi(x|y',y) =\delta(y'-y)+\int_0^1 dy_1[\Phi(x|y',y_1+y)\phi(x) -\Phi(x|y',y)\phi(0)]. \tag{A8}$$

Then expression (A6) can be cast into the form

$$\omega\tilde{W}(\omega y)\left(\frac{2\alpha}{\rho_c(\omega y,s)}\right)^d\int d^dx\;\phi^2(x) \times\left[-\int_0^1 dy'\int_0^{y'}dy_1\,y'\Phi\left(x|y'+\frac{\epsilon}{\omega},y_1+\frac{\epsilon}{\omega}\right) +\int_0^1\int_0^{1-y_1}dy'\,y'\Phi\left(x|y'+\frac{\epsilon}{\omega},\frac{\epsilon}{\omega}\right)\right]. \tag{A9}$$

For $s=0$, Eq. (A8) contains no physical parameter. It only leads to a determination of the function

$$\Phi_0(x|y',y)=\theta(y'-y)\Phi_0(x|y'-y),$$

which satisfies the equation

$$0=\delta(y'-y)+\int_0^1 dy_1[\Phi_0(x|y',y_1+y)\phi(x) -\Phi_0(x|y',y)\phi(0)]. \tag{A10}$$

Provided we restrict our consideration to small frequencies in deriving the self-consistency equation, we only need to take into account the linear contribution of the function Φ with respect to $s/(\tilde{W}(\omega y)\mathcal{N}(\omega y)\omega)$. Then, again using the smallness of the variation of the effective transition probabilities and the density of states with respect to changes of energy over intervals of length ω, Φ can be approximated as

$$\Phi(x|y',y)=\Phi_0(x|y'-y)-\frac{s}{\tilde{W}(\omega y)\mathcal{N}(\omega y)\omega} \times\int_y^{y'}dy_1\,\Phi_0(x|y-y_1)\Phi_0(x|y_1-y). \tag{A11}$$

Using this expression the self-consistency equation takes the form of Eq. (54), where the coefficients a and b are given by

$$a=\frac{1}{2}\,\frac{\displaystyle\int d^dx\;\phi^2(x)\int_0^1(1-y)^2\Phi_0(x|y)}{\displaystyle\int d^dx\;\phi^2(x)}, \tag{A12}$$

$$b=\frac{1}{2}\,\frac{\displaystyle\int d^dx\,\phi^2(x)\int_0^1 dy(1-y)^2\int_0^y dy_1\,\Phi_0(x|y-y_1)\Phi_0(x|y_1)}{\displaystyle\int d^dx\;\phi^2(x)}. \tag{A13}$$

APPENDIX B: SADDLE-POINT APPROXIMATION

For energy-dependent densities of states, it turns out to be difficult to obtain explicit expressions for the time dependence of the energy distribution function. Here, according to Eq. (45), the time dependence has to be calculated from the equation

$$P(\varepsilon,\varepsilon_0,t)=\int_{-i\infty}^{+i\infty}\frac{ds}{2\pi i v(\varepsilon,s)}\exp\left[st-s\int_\varepsilon^{\varepsilon_0}\frac{d\varepsilon'}{v(\varepsilon',s)}\right], \tag{B1}$$

where $v(\varepsilon,s)$ is given by Eq. (62) or (64). Let us try to integrate over s, using the saddle-point approximation. The saddle-point position $s_0(\varepsilon,\varepsilon_0,t)$ may be obtained from the equation

$$t=\int_\varepsilon^{\varepsilon_0}\frac{d\varepsilon'}{v(\varepsilon')g(\varepsilon',s_0)}-s_0\int_\varepsilon^{\varepsilon_0}\frac{d\varepsilon'}{v(\varepsilon')g^2(\varepsilon',s_0)}\,\frac{\partial g(\varepsilon,s_0)}{\partial s_0}, \tag{B2}$$

where $g=v(\varepsilon,s)/v(\varepsilon,0)=\exp\mathcal{W}[s/\Omega(\varepsilon)]$ was introduced. The expression for the diffusion propagator in the saddle-point approximation may be written as

$$P_L(\varepsilon,\varepsilon_0,t)=\frac{1}{\sqrt{4\pi D(\varepsilon,\varepsilon_0,s_0)}}\,\frac{1}{v_0(\varepsilon)g(\varepsilon,s_0)}\times\exp\left[-s_0^2\int_{\varepsilon}^{\varepsilon_0}\frac{d\varepsilon'}{v_0(\varepsilon')g^2(\varepsilon',s_0)}\,\frac{\partial g(\varepsilon,s_0)}{\partial s_0}\right], \tag{B3}$$

where

$$D(\varepsilon,\varepsilon_0,s_0)=-\frac{\partial^2}{\partial s_0^2}\int_{\varepsilon}^{\varepsilon_0}\frac{d\varepsilon'\,\Omega(\varepsilon')\ln g(\varepsilon',s_0)}{2v_0(\varepsilon')}>0. \tag{B4}$$

Assuming $s_0/\Omega\ll 1$, we have $g(\varepsilon,s_0)\simeq 1+s_0/\Omega-s_0^2/2\Omega^2$,

$$s_0=\frac{t-T(\varepsilon,\varepsilon_0)}{2D(\varepsilon,\varepsilon_0)},\quad D(\varepsilon,\varepsilon_0)=\int_{\varepsilon}^{\varepsilon_0}\frac{d\varepsilon'}{v_0(\varepsilon')\Omega(\varepsilon')},$$

$$T(\varepsilon,\varepsilon_0)=\int_{\varepsilon}^{\varepsilon_0}\frac{d\varepsilon'}{v_0(\varepsilon')}, \tag{B5}$$

and, finally, the diffusion propagator

$$P_L(\varepsilon,\varepsilon_0,t)\simeq\frac{1}{\sqrt{4\pi D(\varepsilon,\varepsilon_0)}}\,\frac{1}{v(\varepsilon)}\exp\left[-\frac{[t-T(\varepsilon,\varepsilon_0)]^2}{4D(\varepsilon,\varepsilon_0)}\right]. \tag{B6}$$

At a given time t the exponent in the above distribution function is maximal at $\varepsilon=\varepsilon_m$, where ε_m is given by the condition $t=T(\varepsilon_m,\varepsilon_0)$, or

$$t=\int_{\varepsilon_m}^{\varepsilon_0}\frac{d\varepsilon'}{v_0(\varepsilon')}. \tag{B7}$$

Expanding expression (B6) around $\varepsilon=\varepsilon_m$, we have the Gaussian distribution

$$P_L(\varepsilon,\varepsilon_0,t)\simeq\frac{1}{\sigma(\varepsilon_m,\varepsilon_0)\sqrt{2\pi}}\exp\left[-\frac{[\varepsilon-\varepsilon_m(\varepsilon_0,t)]^2}{2\sigma^2(\varepsilon_m,\varepsilon_0)}\right], \tag{B8}$$

with dispersion

$$\sigma^2(\varepsilon_m,\varepsilon_0)=2v_0^2(\varepsilon_m)D(\varepsilon_m,\varepsilon_0)=2v^2(\varepsilon_m)\int_{\varepsilon_m}^{\varepsilon_0}\frac{d\varepsilon'}{v_0(\varepsilon')\Omega(\varepsilon')}. \tag{B9}$$

Note that, according to Eq. (B7), the dispersion is time dependent. If the density of states $\mathcal{N}(\varepsilon)$, and consequently $\Omega(\varepsilon)$ and $v_0(\varepsilon)$ are strongly varying functions of ε, the integrals can be further simplified. In this case we have

$$\int_{\varepsilon_m}^{\varepsilon_0}\frac{d\varepsilon'}{v(\varepsilon')}\simeq\left[\frac{dv(\varepsilon_0)}{d\varepsilon_0}\right]^{-1}-\left[\frac{dv(\varepsilon_m)}{d\varepsilon_m}\right]^{-1}, \tag{B10}$$

$$\int_{\varepsilon_m}^{\varepsilon_0}\frac{d\varepsilon'}{v(\varepsilon')\Omega(\varepsilon')}\simeq\left[\frac{d[v(\varepsilon_0)\Omega(\varepsilon_0)]}{d\varepsilon_0}\right]^{-1}-\left[\frac{d[v(\varepsilon_m)\Omega(\varepsilon_m)]}{d\varepsilon_m}\right]^{-1}, \tag{B11}$$

$$\int_{\varepsilon_m}^{\varepsilon_0}\frac{d\varepsilon'}{v(\varepsilon')\Omega(\varepsilon')}\simeq\left[\frac{dv(\varepsilon_0)\Omega(\varepsilon_0)}{d\varepsilon_0}\right]^{-1}-\left[\frac{dv(\varepsilon_m)\Omega(\varepsilon_m)}{d\varepsilon_m}\right]^{-1}. \tag{B12}$$

The applicability condition for the saddle-point method is

$$D^3(\varepsilon,\varepsilon_0,s_0)\gg\left[\frac{\partial^3}{\partial s_0^3}\int_{\varepsilon}^{\varepsilon_0}d\varepsilon'\frac{\Omega(\varepsilon')\ln g(\varepsilon',s_0)}{2v(\varepsilon')}\right]^2. \tag{B13}$$

Note that this condition restricts the applicability of the saddle-point approximation to times that are not too large. Also, from Eqs. (B5) one can see that this approximation is invalid when ε is close enough to the initial point ε_0; here $|s_0|$ becomes large, and it is not possible to expand over s_0. Thus the initial stage of the evolution, when the entire distribution is concentrated near ε_0, also has to be investigated separately.

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