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Impact of the density of states on the dynamical hopping conductivity

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We argue that the dynamical hopping conductivity at low frequencies can be strongly affected by the form of the density of states, if the density of states depends strongly on energy and the transport is not restricted to a small vicinity of the Fermi energy. To this end we focus on an exponential density of states. For such a density of states we show that the strong energy dependence of the density of states affects significantly the characteristic frequencies, governing the impact of the frequency dependence on the conductivity, the expected magnitude of the loss, and the characteristic exponents in the multiple hopping regime. These facts manifest also themselves in the transient current close to equilibrium.

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I. INTRODUCTION

A characteristic feature of many disordered materials is their strong frequency dependence for low frequencies. The frequency dependence manifests itself in an increase of the conductivity with increasing frequency. This sets disordered materials apart from ordered materials, which are only affected by the frequency of the electric field at very high frequencies. In contrast to disordered materials, an increase of the frequency always results in a decrease of the conductivity in ordered materials. A particular strong dependence of the dynamical conductivity on frequency at very low frequencies is observed in strongly localized systems, in which transport proceeds by hopping between localized states, such as impurity bands, Anderson insulators, glasses, and polymers (see, e.g., Refs. 1–7).

The physical reason for the strong frequency dependence can easily be understood in the language of percolation theory. In an ordered system the current between two electrodes flows along straight lines. The conductivity is constant along these lines. In a strongly localized, disordered system, however, the current between two electrodes in a constant electric field chooses the optimal path. The opportunity to choose the path results from the fact that the local conductivity changes from point to point. Thus the current writhes somehow through the sample. In doing so, it somewhere passes a critical resistor, the is that piece of the path with the highest resistance. If the distribution function for the resistors is wide enough the critical resistor exceeds all other resistors on the path by orders of magnitude, and thus, determines the resistivity of the whole sample. In an alternating electric field, however, the current lines do not have to be closed. A substantial part of the current can be formed by the displacement current as well. In an ordered system the displacement current only contributes to the imaginary part of the conductivity. In a disordered system, however, the displacement current can be used to eliminate the most resistive parts of the current path merely by increasing frequency. Since with increasing frequency the resistors are excluded according to their strength, at first the critical resistor and thereafter the next smaller ones, the resistance of the whole network decreases.

Since some of the resistors have been excluded from the percolation cluster by the alternations of the electric field, the current in an alternating electric field is restricted to finite clusters. At high frequencies these clusters reduce to pairs of sites.[8] In this limit, the Pollak-Geballe limit, the conductivity is dominated by resonant pairs, which have the property that the transition probability per unit time for jumps between them is of the order of the frequency of the external field. Within that range the frequency dependence of the conductivity is close to $\mathrm{Re}\,\sigma(\omega)\propto\omega^s$, where $s<1$ and frequency dependent. Since in many experiments a frequency dependence of this type is observed, it appears on first glance as if the frequency dependence of hopping systems can almost always been explained by the two-site model. A further investigation of the two-site model, however, reveals that its range of applicability is restricted to very high frequencies, much higher than those usually used in the experiments (see, e.g., Ref. 1). Furthermore, the two-site model predicts a decreases of s with increasing frequency. In many experiments, however, an increase of s with increasing frequency is observed.[1]

Below the range of applicability of the two-site model, the current is restricted to finite but very large clusters, as first pointed out in the Refs. 9 and 10. Within that regime the conductivity has been investigated by percolation theory[9–11] and by effective-medium methods (see, e.g., Refs. 12] and Ref. 7, and references therein). From the practical point of view the results of both methods agree with each other.[11] According to the results of the effective-medium theory the dynamical conductivity in the hopping regime below the range of applicability of the two-site model satisfies the equation

$$\frac{\sigma(s)}{\sigma(0)}\ln\frac{\sigma(s)}{\sigma(0)}=\frac{s}{\omega_0}. \tag{1}$$

Here $s=-i\omega$ is the frequency of the alternating electric field and ω_0 is a frequency of the order of the dc conductivity.

Surprisingly, Eq. (1) is found to also hold in many other disordered systems, which are not necessarily of hopping type (see, e.g., the recent review by Dyre and Schroder[7]), so that ac universality seems to exist. The only ingredient, which is shared by all of these systems, seems to be disorder.

Equation (1) leads to the following two conclusions for the ac conductivity. First, the characteristic frequency ω_0, which marks the beginning of the strong frequency dependence, is of the order of the dc conductivity. Second, the real and the imaginary part of the ac conductivity increases with frequency, and third, for $\omega \gg \omega_0$, i.e., in the multiple-hopping regime, the real and imaginary part of the conductivity can be approximated by $\mathrm{Re}\,\sigma(\omega)\propto\omega^{s'(\omega)}$ and $\mathrm{Im}\,\sigma(\omega)\propto\omega^{s''(\omega)}$, where both $s', s''<1$ and frequency dependent. With increasing frequency, s' and s'' increase, as observed in the experiments. The above-mentioned properties also reflect in the time dependence of the transient current, the Heaviside transform of the conductivity. They lead to anomalous diffusion for $t<1/\omega_0$, where $\boldsymbol{j}(t)\propto 1/t^{1-\alpha(t)}$ $[0<\alpha(t)<1]$, and the exponential decay of $\boldsymbol{j}(t)-\boldsymbol{j}(\infty)$ for $t>1/\omega_0$.

In all of the investigations on the frequency dependence of the dynamical conductivity below the Pollak-Geballe limit, however, the consideration has been restricted to that situation where the density of states varies only weakly with energy, and the main contribution to the conductivity results from the vicinity of the Fermi energy. In an important class of hopping systems, however, e.g., in band tails of amorphous semiconductors (see, e.g., Refs. 13 and 14) or in certain polymers (see, e.g., Refs. 15–17), this situation is usually not met. These systems have the property that their density of states varies strongly with energy. The conduction band tail of amorphous hydrogenated silicon, e.g., is widely believed to increase exponentially with increasing energy.[13,14] Furthermore, the Fermi energy is often not situated within the band. In this case the most important contributions to the dc conductivity do not result from the vicinity of the Fermi energy, but from the vicinity of a certain energy level, the transport energy.[18,19] This energy results from the competition between two contributions. On the one hand, the Boltzmann distribution functions favors those contributions to the conductivity, which result from small energies. On the other hand, the mobility favors high energies, since the density of states increases strongly with energy.

It is the purpose of the article to show that the frequency dependence can be strongly affected by the density of states, if the density of states depends strongly on energy. To this end we focus on an exponential density of states and assume that the Fermi energy is not contained in the band. Although this density of states is of much interest for the description of transport properties of many amorphous materials, e.g., for amorphous hydrogenated silicon, so far only investigations on the frequency dependence in the Pollak-Geballe limit have been published.[20] We show that for such a system the first and the third property of Eq. (1), as discussed above, are violated. In such a system the characteristic frequency, which governs the impact of the frequency on the conductivity is not given by the dc conductivity, and s' and s'' are one in the multiple hopping regime.

In order to perform the calculation we use the effective-medium theory by Gouchanour *et. al.*[21] To simplify the effective-medium equations we use the assumption that the energy transferred in one hop is small. Accordingly, the physical situation is, in principle, different from that usually considered in the band tail problem, e.g., in Refs. 18, 19, or 22–25. In our model the charge carrier cannot jump from the very bottom of the tail to the very top of the tail, or vice versa, in one jump. It turns our that this fact reflects itself in certain dimensionless parameters, as discussed further below.

II. BASIC EQUATIONS

We consider localized charge carriers far from the Fermi energy close to equilibrium. If we assume that the number of charge carriers is small we can neglect Fermi correlation. In this case, the motion of the charge carriers can be described by the simple rate equation

$$\frac{dn_m}{dt}=\sum_n [n_n W_{nm}-n_m W_{mn}]. \tag{2}$$

Here n_m is the number of particles on the site m with site energy ϵ_m and position vector $\boldsymbol{R}_m$,

$$W_{nm}=\theta(\kappa-|V_{nm}|)\,\nu\exp\left[-2\alpha\left|\boldsymbol{R}_{nm}\right|+\frac{\beta}{2}(V_{nm}-|V_{nm}|)\right] \tag{3}$$

is the transition probability for a hop from the site n to the site m, α^{-1} is the localization length, ν is the attempt-to-escape frequency, β is the inverse temperature, and κ is the maximal amount of energy transferable in one hop. Furthermore, $V_m=\epsilon_m-e[\boldsymbol{E}(t)\boldsymbol{R}_m]$, where $\boldsymbol{E}(t)$ is the electric field at time t, and e is the charge. Both the position vectors and the site energies are random quantities. We assume that the sites are uniformly and homogeneously distributed in space, and that the site energies $\{\epsilon_n\}$ are distributed randomly according to a distribution function proportional to the density of states $N(\epsilon)$.

To calculate the current, we restrict the consideration to weak electric fields, and linearize the rate equation (2) with respect to the electric field. To this end we decompose n_m into two parts, into the Boltzmann distribution function

$$f_m=C\exp(-\beta\epsilon_m) \tag{4}$$

and a small deviation $n_m^{(1)}$ due to the electric field, according to the relationship

$$n_m=f_m+n_m^{(1)}. \tag{5}$$

Doing so, we find that the quantity

$$U_m=n_m^{(1)}-\beta f_m e\boldsymbol{E}\boldsymbol{R}_m \tag{6}$$

satisfies the equation

$$s\{U_m+\beta f_m e[\boldsymbol{E}(s)\boldsymbol{R}_m]\}=\sum_n [U_n W_{nm}-U_m W_{mn}], \tag{7}$$

after a Laplace transformation with respect to time. Here s is the frequency which corresponds to the Laplace transformation with respect to time. If we solve Eq. (6) formally with the Green's function P, which satisfies the equation

$$sP_{m'm}=\delta_{m'm}+\sum_{n}\,[P_{m'n}W_{nm}-P_{m'm}W_{mn}], \tag{8}$$

we find that

$$n_m^{(1)}(s)=-s\beta\sum_{n}\,f_nP_{nm}(s)e(\boldsymbol{E}\boldsymbol{R}_{nm}). \tag{9}$$

Therefore, we obtain for the Ohm current the equation

$$\boldsymbol{j}(s)=\frac{e^2s^2\beta}{2\Omega}\sum_{nm}\,f_nP_{nm}(s)\boldsymbol{R}_{mn}(\boldsymbol{E}\boldsymbol{R}_{mn}). \tag{10}$$

Here Ω is the volume of the system.

III. THE DIFFUSION PROPAGATOR

Equation (10) yields a convenient starting point for the calculation of the configuration averaged current. If we introduce continuous coordinates, according to the definition

$$P(\boldsymbol{R}',\epsilon'|\boldsymbol{R},\epsilon)=\sum_{nm}\,\delta(\epsilon'-\epsilon_n)\,\delta(\boldsymbol{R}'-\boldsymbol{R}_n)\times P_{nm}\,\delta(\boldsymbol{R}-\boldsymbol{R}_m)\,\delta(\epsilon-\epsilon_m), \tag{11}$$

we find that the equation for the calculation of the configuration averaged current takes the form

$$\boldsymbol{j}(s)=\frac{e^2s^2\beta}{2}\int\,d\epsilon\,d\epsilon'\;d\boldsymbol{R}\,f_B(\epsilon)\boldsymbol{R}(\boldsymbol{E}\boldsymbol{R})\langle P(\boldsymbol{R},\epsilon'|\boldsymbol{0},\epsilon)\rangle. \tag{12}$$

Here the bracket indicates the configuration average, and $f_B(\epsilon)=f_m|_{\epsilon_m=\epsilon}$ is the Boltzmann distribution function. Thus the calculation of the configuration averaged current is reduced to the calculation of the configuration averaged Green's function.

To calculate the configuration averaged Green's function we use the effective medium approximation by Gouchanour, Anderson, and Fayer.[21] Originally, this technique has been developed for the investigation of hopping systems with topological disorder. However, it is a simple matter to check, that the extension of this method to systems with both topological and energetic disorder only amounts to a change of notation, so that we can directly use the results of Ref. 21. Doing so, we find that the configuration averaged Green's function satisfies the system of integral equations

$$\langle P(\boldsymbol{R},\epsilon'|\boldsymbol{0},\epsilon)\rangle=N(\epsilon')F(\boldsymbol{R}|\epsilon',\epsilon), \tag{13}$$

$$\begin{aligned}sF(\boldsymbol{R}|\epsilon',\epsilon)={}&\delta(\boldsymbol{R}'-\boldsymbol{R})\,\delta(\epsilon'-\epsilon)\\&+\int d\rho_1[F(|\boldsymbol{R}-\boldsymbol{R}_1||\epsilon',\epsilon_1)\tilde{W}(R_1|\epsilon_1,\epsilon)N(\epsilon)\\&-F(|\boldsymbol{R}||\epsilon',\epsilon)\tilde{W}(R_1|\epsilon',\epsilon_1)N(\epsilon_1)],\end{aligned} \tag{14}$$

$$\tilde{W}(R|\epsilon',\epsilon)=\frac{W(R|\epsilon',\epsilon)}{1+f(\epsilon)W(R|\epsilon,\epsilon')+f(\epsilon')W(R|\epsilon',\epsilon)}, \tag{15}$$

and

$$f^{-1}(\epsilon)=s+\int\,d\epsilon_1\,dR\,\tilde{W}(R|\epsilon,\epsilon_1)N(\epsilon_1). \tag{16}$$

Here $W(R|\epsilon',\epsilon)=W_{nm}|_{R_{nm}=R,\epsilon_n=\epsilon',\epsilon_m=\epsilon}$. Obviously, the function F has to be identified with the diffusion propagator, $\tilde{W}$ is the renormalized transition probability, and $f(\epsilon)$ is the effective medium. Due to detailed balance and probability conservation, the diffusion propagator satisfies the relationships

$$\int\,d\epsilon\,dR\,s\,F(R|\epsilon',\epsilon)=1 \tag{17}$$

and

$$\int\,d\epsilon'\,dR\,s\,N(\epsilon')f_B(\epsilon')F(R|\epsilon',\epsilon)=N(\epsilon)f_B(\epsilon). \tag{18}$$

IV. QUASIELASTIC APPROXIMATION

The equation of motion for the calculation of the diffusion propagator is an integral equation with respect to energy, which cannot simply be solved, if the density of states depends on energy. Only for a density of states that is independent of energy, that is, for a constant density of states, can this equation be reduced to an algebraic equation after Fourier transformation. Therefore, in order to simplify this system further, we use additional approximations. To render the calculations feasible we use the quasielastic approximation. This approximation relies on the notion that the energy exchanged in one hop is small, so that

$$\frac{\kappa}{f(\epsilon)}\frac{df(\epsilon)}{d\epsilon}\ll 1. \tag{19}$$

Furthermore, we assume that

$$\beta\kappa\ll 1. \tag{20}$$

In this case the renormalized transition probabilities take the form

$$\begin{aligned}\tilde{W}(R|\epsilon',\epsilon)={}&\theta(\epsilon'-\epsilon)\,\theta(\kappa-\epsilon'+\epsilon)\tilde{W}(R|\epsilon')\\&+\theta(\epsilon-\epsilon')\,\theta(\kappa-\epsilon+\epsilon')\tilde{W}(R|\epsilon)e^{-\beta(\epsilon-\epsilon')},\end{aligned} \tag{21}$$

where

$$\tilde{W}(R|\epsilon)=\frac{W(R)}{1+2f(\epsilon)W(R)}. \tag{22}$$

Here $W(R)=\nu\exp(-2\alpha R)$. If we use this approximation and the inequalities (19) and (20), then the self-consistency equation (16) simplifies considerably. For $s=0$ the solution of this equation is given by

$$\rho_c(\epsilon,0)=\frac{2\alpha}{[\kappa N(\epsilon)]^{1/d}}\left(\frac{d}{S_d}\right)^{1/d}, \tag{23}$$

and for small s, that is, for s satisfying $|\rho_c(\epsilon,0)-\rho_c(\epsilon,s)|\ll\rho_c(\epsilon,0)$, we obtain the equation

$$[\rho_c(\epsilon,0)-\rho_c(\epsilon,s)]\exp[\rho_c(\epsilon,0)-\rho_c(\epsilon,s)]=\frac{s}{\omega_0(\epsilon)}. \tag{24}$$

Here

$$\rho_c(\epsilon,s)=\ln[2f(\epsilon,s)\nu] \tag{25}$$

is the characteristic hopping length,

$$\omega_0(\epsilon)=\frac{2d\nu}{\rho_c(\epsilon,0)}\exp[-\rho_c(\epsilon,0)], \tag{26}$$

and S_d is the solid angle in d dimensions ($S_2=2\pi$, $S_3=4\pi$). For large s, that is, for $s\gg\omega_1(\epsilon)$, where

$$\omega_1(\epsilon)\approx 2\nu\exp[-\rho_c(\epsilon,0)], \tag{27}$$

the self-consistency equation (16) simply yields the exact high-frequency result, which is

$$\rho_c(\epsilon,s)=\ln\frac{2\nu}{s}. \tag{28}$$

Now, where the effective transition probability is known, we can insert the Eqs. (13), (14), and (21) into Eq. (12) and calculate the current. Doing so, we find that

$$\boldsymbol{j}(s)=\boldsymbol{E}\int d\epsilon\,\sigma(\epsilon,s), \tag{29}$$

where the spectral conductivity satisfies the equation

$$\sigma(\epsilon,s)=eN(\epsilon)f_B(\epsilon)\mu(\epsilon,s), \tag{30}$$

and the mobility $\mu(\epsilon,s)$ is related to the diffusion coefficient by the Einstein relationship

$$\mu(\epsilon,s)=\frac{e}{kT}D(\epsilon,s). \tag{31}$$

The diffusion coefficient

$$D(\epsilon)=\frac{1}{2d}\int d\boldsymbol{R}\,d\epsilon_1\,R^2\tilde{W}(R|\epsilon,\epsilon_1)N(\epsilon_1) \tag{32}$$

is given by

$$D(\epsilon,s)=\frac{1}{d+2}\,\frac{\nu}{(2\alpha)^2}\,\frac{\rho_c^{d+2}(\epsilon,s)}{\rho_c^d(\epsilon,0)}\exp[-\rho_c(\epsilon,s)]. \tag{33}$$

Equations (29)–(33) yield the starting point for our investigation of conduction processes in model densities of states. To calculate the dc current we only have to set $s=0$ in Eq. (29). In order to calculate the ac conductivity we put $s=-i\omega$ in Eq. (29), where ω is the frequency of the applied external electric field. Furthermore, we investigate the transient current. To this end we assume that the system is in equilibrium for $t<0$ and that a constant electric field is switched on suddenly at time $t=0$. Then

$$\boldsymbol{j}(t)=\frac{1}{2\pi i}\int_c\frac{ds}{s}e^{st}\boldsymbol{j}(s), \tag{34}$$

where c is the appropriate chosen contour in the complex plane.

V. TRANSPORT IN AN EXPONENTIAL DENSITY OF STATES NEAR EQUILIBRIUM

In this section we consider the exponential density of states

$$N(\epsilon)=N_0\exp(3\epsilon/\Delta)\,\theta(\epsilon) \tag{35}$$

at low temperatures, that is in the limit $kT/\Delta\ll 1$, in three dimensions, so that $d=3$. Here $\theta(x)$ is the step function. In the literature, this type of density of states is traditionally used for the description of conduction processes in the conduction band tail of amorphous hydrogenated silicon (see, e.g., Refs. 13, 18, and 19).

Since

$$\rho_c(\epsilon,0)=\rho_0 e^{-\epsilon/\Delta}, \tag{36}$$

where $\rho_0^3=3(2\alpha)^3/(\kappa N_0 4\pi)$, we can use the characteristic hopping length at $s=0$ to parametrize the density of states. We chose the parametrization in such a way, that ρ_0 is the largest characteristic hopping length in the tail. Accordingly, $\epsilon=0$ for a particle at the bottom of the tail. For definiteness we assume that the smallest hopping length in the tail is 1, so that the largest attainable energy is $\epsilon=\Delta\ln\rho_0$, although this not essential, since the results are independent of the upper bound at low T.

To characterize our system completely we still have to adjust the normalization constant C in Eq. (4). To adjust C we assume that the density of charge carriers at temperature T is given by $n(T)$. Here the temperature dependence of the charge-carrier density reflects the origin of the charge carriers in the tail. In general there can be two situations; either the number of charge carriers in the tail is constant or the position of the chemical potential is independent of temperature. The latter situation arises, e.g., if the chemical potential is situated below the band, so that the charge carriers in the tail result from the tails of the Fermi distribution function, that is, from thermal activation from a lower band, such as in amorphous hydrogenated silicon.[19] In this case the temperature dependence of $n(T)$ also manifests itself in the conductivity. On the other hand, if the number of charge carriers in the tail is constant, the Fermi energy is situated within the band. Since we have already neglected Fermi correlation, the consideration of this situation is only justified if the main contributions to the conductivity result from a region that is far from the Fermi energy, which in this case has to be checked if the formulas derived here are applied to a real system. Our formulas apply, if $\epsilon_F < 0$ and $|\epsilon_F/kT| \gg 1$. If we take into account these remarks we can calculate C from the equation

$$n(T) = \int_0^{\Delta \ln \rho_0} d\epsilon \, N(\epsilon) f_B(\epsilon), \tag{37}$$

so that we find

$$C \approx \frac{n(T)}{kTN_0} \tag{38}$$

in the limit $\Delta/kT \gg 1$.

A. dc conductivity

If we now apply Eqs. (29), (31), and (33) to the calculation of the dc conductivity, and use $\rho_c(\epsilon,0)$ instead of ϵ as an integration variable, we find that the dc conductivity is given by the integral

$$\sigma(0) = \frac{e^2}{kT} \frac{N_0 C \Delta}{5(2\alpha)^2} \frac{\nu}{\rho_0^{\Delta/kT-3}} \int_1^{\rho_0} d\rho \exp\left[\ln(\rho)\left(\frac{\Delta}{kT} - 2\right) - \rho\right]. \tag{39}$$

The exponent of the integrand has a maximum at

$$\rho_t = \frac{\Delta}{kT} - 2, \tag{40}$$

at energy

$$\epsilon_t = \Delta \ln \frac{\rho_t}{\rho_0}. \tag{41}$$

The latter energy has already been introduced by Grünewald and Thomas[18] and Shapiro and Adler[19] and is called transport energy.

If we check the saddle-point criterion we find that the saddle-point approximation is applicable provided $\rho_t \gg 1$, which is the case of interest here. The saddle-point approximation yields

$$\sigma(0) = \frac{\sqrt{2\pi}}{5} \frac{e^2 n(T)}{kT} \frac{\nu}{(2\alpha)^2} \rho_0 \rho_t^{3/2} \times \exp\{-\rho_t[1 + \ln(\rho_0/\rho_t)]\}. \tag{42}$$

Thus, since $\rho_t \propto \Delta/kT$, the conductivity appears to be thermally activated, although thermal activation into the conduction band (the top of the tail) is absent. The temperature dependence reflects entirely on the temperature dependence of the transport energy, which results from the competition between the mobility and the Boltzmann function. On the one hand, the mobility of particles at higher energy is much larger than the mobility of particles with lower energy. On the other hand, there are much more particles at lower energy than at higher energy, so that the product of the distribution function and the mobility has a maximum somewhere. However, we stress that despite the existence of the transport energy most of the particles are sitting at the bottom of the tail. Similar results for the dc conductivity have also been obtained earlier by Grünewald and Thomas[18] and Shapiro and Adler.[19]

B. Calculation of $\sigma(s)$

In the preceding section we have seen that the main contribution to the dc conductivity results from a vicinity of the transport energy. Below we are going to show that for every finite s the situation is different. For every finite s the main contribution to the quantity $\sigma(s) - \sigma(0)$ results from the bottom of the tail. The reason for this is that the above-mentioned competition is affected. Due to Eqs. (24) and (28), the hopping length at high frequencies is lower than the hopping length at low frequencies. Furthermore, since both $\omega_0(\epsilon)$ and $\omega_1(\epsilon)$ at low energies are much smaller than at high energies, a frequency s, which acts like a small frequency in the vicinity of, e.g., the transport energy, is a very large frequency at the bottom of the tail. Thus the hopping length at the bottom of the tail is much reduced.

To investigate the change of the conductivity we first focus on small s, with $s \ll \omega_1(0)$. In this case we can use Eq. (24) in the whole range of integration. Then the change of the conductivity takes the form

$$\sigma(s) - \sigma(0) = \frac{1}{5} \frac{e^2 n(T)}{(kT)^2} \frac{\nu\Delta}{(2\alpha)^2} \frac{1}{\rho_0^{\rho_t - 1}} \int_1^{\rho_0} d\rho \exp(\rho_t \ln\rho - \rho) \times \left(\exp\left\{W\left[\frac{s}{6\nu}\rho \exp(\rho)\right]\right\} - 1\right). \tag{43}$$

Here $W(x)$, with $W(x)\exp[W(x)] = x$, is the Lambert function. The integrand of Eq. (43) is a monotonously increasing function of ρ. Consequently, the main contribution results from the vicinity of $\rho = \rho_0$. If we calculate the integral (see the Appendix), using the inequality $\rho_t W_0/\rho_0 \ll 1$, where $W_0 = W[s\rho_0 \exp(\rho_0)/(6\nu)]$, we find that

$$\sigma(s) - \sigma(0) = \sigma(0,s) kT \left\{ W_0 - \frac{\rho_t}{\rho_0}[(1 + W_0^2 - W_0) - \exp(-W_0)] \right\}. \tag{44}$$

Here $\sigma(0,s)$ is the spectral conductivity at the bottom of the tail. Accordingly, the frequency dependence of the conductivity at low frequencies is governed by $\omega_0(0)$, the lower limiting frequency at the bottom of the tail.

If $s \gg \omega_1(0)$, the electrons at the bottom of the tail move in the high-frequency limit. In this case

$$\sigma(s) = \frac{e^2 n(T)}{(kT)^2} \frac{\nu}{5(2\alpha)^2} \int_0^{\epsilon_1(s)} d\epsilon \frac{\ln^5(2\nu/s)}{\rho_c^3(\epsilon,0)} \times \exp\left[\frac{3\epsilon}{\Delta} - \frac{\epsilon}{kT} - \ln(2\nu/s)\right] + \sigma_1(s). \tag{45}$$

Here $\epsilon_1(s)$ is upper bound for the high-frequency limit, that is, electrons with $\epsilon < \epsilon_1(s)$ are in the high-frequency limit, and electrons with $\epsilon > \epsilon_1(s)$ are either in the multiple hopping regime or at lowest frequencies. The latter electrons contribute to the ac conductivity with $\sigma_1(s)$. Since the integrand decreases monotonously with increasing energy the main contribution to the integral results again from the vicinity of $\epsilon=0$, so that $\sigma_1(s)$ is negligible, and $\epsilon_1(s)$ irrelevant. If we take into account this fact, we obtain for $s \gg \omega_1(0)$ the simple equation

$$\sigma(s)=\sigma(0,s)kT. \tag{46}$$

C. Frequency dependence

For low frequencies of the applied external electric field, that is, for $\omega \ll \omega_1(0)$, the frequency dependence of the conductivity is governed by the lower limiting frequency $\omega_0(0)$, the lower limiting frequency at the bottom of the tail. This sets the situation in the exponential tail apart from that in the conventional theory on ac hopping conductivity, where the frequency dependence at low frequencies is governed by a characteristic frequency of the order of the dc conductivity. Since $\omega_0(0) \ll \omega_0(\epsilon_t)$ the conductivity depends on frequency at much lower frequencies than in the conventional theory. For $\omega \ll \omega_0(0)$ this dependence is weak. In this case Eq. (44) can be expanded with respect to powers of ω/ω_0. The expansion has the form

$$\sigma(s)-\sigma(0)=\sigma(0,0)kT\left[\frac{-i\omega}{\omega_0(0)}-\frac{1}{2}\frac{\rho_t}{\rho_0}\left(\frac{-i\omega}{\omega_0(0)}\right)^2+\frac{1}{3}\frac{\rho_t}{\rho_0}\left(\frac{-i\omega}{\omega_0(0)}\right)^3-\cdots\right]. \tag{47}$$

For $\omega \gg \omega_0(0)$ Eq. (44) takes the form

$$\sigma(\omega)-\sigma(0)\approx\sigma(0,0)kT\left(\frac{-i\omega}{\omega_0(0)}\right)\left(1-\frac{\rho_t}{\rho_0}W_0\Big|_{s=-i\omega}\right). \tag{48}$$

To obtain explicit expressions for the real and imaginary part of the dynamical conductivity we restrict the consideration to the leading term in the asymptotic expansion of $W(x)$ for $x \gg 1$. In this approximation $W(x)\approx \ln(x)$. If we use this approximation we obtain

$$\mathrm{Re}[\sigma(\omega)-\sigma(0)]=\sigma(0,0)kT\frac{\pi}{2}\frac{\rho_t}{\rho_0}\frac{\omega}{\omega_0(0)}, \tag{49}$$

$$\mathrm{Im}\,\sigma(\omega)=-\sigma(0,0)kT\frac{\omega}{\omega_0(0)}. \tag{50}$$

This result differs from the conventional result for the ac hopping conductivity in the multiple hopping regime (for a text book treatment see, e.g., Ref. 1) in two ways. First, if we write the real and the imaginary part of the conductivity in the form

$$\mathrm{Re}[\sigma(\omega)-\sigma(0)]\propto\omega^{s'(\omega)} \tag{51}$$

and

$$\mathrm{Im}\,\sigma(\omega)\propto\omega^{s''(\omega)}, \tag{52}$$

respectively, we find that in the exponential tail $s'(\omega)=s''(\omega)=1$, and independent of ω, for frequencies in the range of applicability of Eqs. (51) and (52). Second, while for a weakly energy dependent density of states the real part of the conductivity differs from the imaginary part only by a factor $\pi/\{2\ln[\pi\omega/(2\tilde{\omega}_0)]\}$,[1] where $\tilde{\omega}_0$ is a frequency of the order of the dc conductivity, the real part of the ac conductivity in the exponential density of states is smaller than the imaginary part by a factor of $\pi\rho_t/(2\rho_0)$. Accordingly, the loss is extremely small.

Since ρ_0 is very large the frequency dependence of the conductivity is already determined by the Pollak-Geballe limit at relatively low frequencies. If $\omega \gg \omega_1(0)$ jumps between pairs of sites prevail. In this case we find

$$\mathrm{Re}\,\sigma(\omega)=\frac{e^2 n(T)}{kT}\frac{\nu}{(2\alpha)^2\rho_0^3}\frac{\pi}{2}\frac{\omega}{2\nu}\ln^4(2\nu/\omega) \tag{53}$$

and

$$\mathrm{Im}\,\sigma(\omega)=-\frac{e^2 n(T)}{kT}\frac{\nu}{5(2\alpha)^2\rho_0^3}\frac{\omega}{2\nu}\ln^5(2\nu/\omega). \tag{54}$$

Accordingly, in this limit, the frequency dependence is the same as the frequency dependence of the ordinary hopping conductivity in the same limit (see, e.g., Ref. 1), but the temperature dependence is different. This difference is caused by the absence of the Fermi energy in our model. It results from the normalization factor (38).

D. Transient current

The differences between the ac conductivity for hopping transport in weakly energy dependent densities of states in the vicinity of the Fermi energy and the ac-conductivity in an exponential density of states for charge carriers far from the Fermi energy, also affect the relaxation current. If we use Eq. (44) in the calculation of the integral (34) we find that for large times,

$$\sigma(\tau)-\sigma(\infty)=kT\sigma(0,0)\frac{e^{3/2}}{\sqrt{2\pi}}\frac{\rho_t}{\rho_0}\frac{e^{-\tau/e}}{\tau^{5/2}}. \tag{55}$$

Here $\tau=\omega_0(0)t$. Equation (55) is applicable if $\tau\gg 1$. This has to be in contrast to the time dependence of the relaxation current in the conventional theory, which leads to[1,26] $\sigma(\tau)-\sigma(\infty)\propto\exp(-\tau/e)/\tau^{3/2}$, where again $\tau=\tilde{\omega}_0 t$. Thus, on the one hand, as a function of τ the decay of the deviation of the transient current from its stationary value appears to be faster. On the other hand, since $\omega_0(0)\ll\omega_0(\epsilon_t)$, the exponential decay is only observed at a much larger time scale than in the conventional theory for hopping transport in a weakly energy-dependent density of states in the vicinity of the Fermi energy.

Since the exponential decay is observed at a much larger time scale than in the ordinary situation, where the carriers jump in a weakly energy dependent density of states in the vicinity of the Fermi energy, the transport regime of anomalous diffusion becomes even more important than in the conventional situation. Anomalous diffusion is realized for $\tau \ll 1$. If $\tau \ll 1$ but $\tau\rho_0 \gg 1$ we can again use Eq. (44) for the calculation of the integral (34). Doing so, we obtain

$$\sigma(\tau)-\sigma(\infty)=kT\sigma(0,0)\frac{\rho_t}{\rho_0}\frac{1}{\tau}. \tag{56}$$

Again this expression has to be contrasted with the known results for hopping transport in a weakly energy-dependent density of states in the vicinity of the Fermi energy, which in this case leads to a time dependence of the form $\sigma(\tau)-\sigma(\infty)\propto\tau^{-1+\alpha}$, where $\alpha>0$, is in general, a function of τ (see, e.g., Ref. 1, and references therein). Thus the present theory leads to $\alpha=0$ for $\tau\gg 1/\rho_0$. Note that, when talking about the exponent α, we are talking about properties of the transient current close to equilibrium. This exponent is different from that which is obtained in the nonequilibrium situation in relaxation current experiments far from equilibrium, as investigated, e.g., in Ref. 13, and references therein. For the investigation of the transient current for $\nu t\gg 1$ and $\tau\rho_0 \ll 1$, we can use Eq. (46) in the calculation of the integral (34). Since, however, the frequency dependence of the diffusion coefficient is not affected by the density of states in the Pollak-Geballe limit, the results for the time dependence of the transient current agree in this limit with those from the conventional theory. Within that range we have $\alpha=\alpha(t)>0$. Note that, since ρ_0 is very large, this regime is applicable also for relatively large times.

VI. DISCUSSION OF THE QUASIELASTIC APPROXIMATION

An important ingredient in our calculation is the quasielastic approximation. This approximation relies on the notion that only hops with small energy transfer are characteristic for a particle ensemble. Hard hops, that is, hops with large energy transfer, are considered as rare, and thus are ignored. In our calculation we have used this assumption in the calculation of the effective transition probability (21). This raises the question, whether there are physical reasons to believe that this assumption is justified, and whether there are also physical systems that can be compared experimentally with the results of our calculation.

From the physical point of view there are several reasons to believe that the energy scale ω exists, and is small. All of them have their origin in peculiarities of the electron-phonon coupling in strongly localized systems, which lead to a restriction of the amount of energy transferable in one jump. It is important to realize that not all phonons can interact with localized electrons equally well. The high energetic phonons are localized in such systems, and thus do not contribute much to transport. Practically, such phonons can only contribute to transport if the electron is sitting purely by chance, on such a site, that carries a localized mode, which is unlikely. Thus, already on the first glance we have to conclude, that only phonons with wave vectors $q \ll q_{\mathrm{Debye}}$, where q_{Debye} is the Debye wave vector, are important. If the material is, e.g., a material with weak electron-phonon coupling, we can restrict the consideration to one-phonon processes. If we then use the argument given above, we immediately conclude that the energy scale ω exists, and that this energy scale is much smaller as the Debye energy. However, if we further investigate the electron-phonon interaction in such materials, we find that even phonons with moderate wave vectors are not very important. The reason for this is simply that the electron-phonon coupling constant is, strictly speaking, not a constant, but a function of the phonon wave vector q, that is finally of the energy transferable in one jump. It is proportional to both to the overlap integral between the electron wave functions and a phonon wave and to the Fourier transformed electromagnetic potential, which describes the electromagnetic coupling between the electron and the phonon. The overlap integral tends rapidly to zero for phonons with wave vectors $q>2\alpha$ and thus also renders those modes with $q>q_{\mathrm{Debye}}\alpha a$ ineffective, where a is the lattice constant of the host material. The Fourier transformed electromagnetic potential is considered as a constant in most calculations of the transition probabilities (see, e.g., Ref. 1). However, this assumption is not reasonable, since from the physical point of view it means that the electric field of the phonon wave is immediately screened out. Such an approximation is reasonable for a metal. In the relevant class of materials, however, there are no electrons which could strongly screen out this field. As a consequence, the Fourier transformed potential is the Fourier transform of a potential with long range, and therefore already tends to zero rapidly for tiny q. Consequently, we have to conclude that ω is small.

It might be argued that due to the imperfections, the relevant class of materials, such as the band tails of amorphous semiconductors or organics, always tend to be materials with strong electron-phonon coupling. But we would like to stress again that such an assertion is an empty statement, since the electron-phonon coupling constant is not a constant but a function of q, and the question is not how strong the electron-phonon interaction is in general, but how strong the electron-phonon coupling constant is for those modes that most likely contribute to transport. In general, we believe that the assumption that the electron-phonon interaction for the relevant modes is not strong is used in all of the papers referred to in our references, since transition probabilities have not been considered, in any of the papers which are adequate for strong electron-phonon coupling.

If, however, the assumption that the transitions can be considered as quasielastic is physical then the predictions of the quasielastic approximations should not be in conflict with the experiments, and thus also not in conflict with those properties of the inelastic theories, which have been verified experimentally. For the exponential density of states there are at present three observable quantities, which have been investigated both by our quasielastic theory and by theories, which have not used this assumption. These quantities are the energy relaxation rate at lowest temperatures,[27] the relaxation current at lowest temperatures,[28] and the dc conductivity [Eq. (42)]. If we look on our expression (42) for the dc conductivity we note that the energy scale ω has dropped out. The same expression for the dc conductivity is also obtained in the inelastic theory (see, e.g., Ref. 19), that is, with-out the assumption of quasielasticity. Accordingly, the energy scale ω cannot be detected in any measurement of the dc conductivity. In a recent paper[28] we have also shown that the time dependence of the relaxation current in quasielastic approximation agrees with the time dependence of the relaxation current in the inelastic theory[13] up to a number, which scales the attempt-to-escape frequency ν by the ratio ω/Δ at appropriate places. Unfortunately, it turns out to be hard to extract this number from a relaxation current experiment. However, fortunatly the same number can be obtained from an energy relaxation experiment. For the energy relaxation experiment both methods predict a time dependence of the type[13,27]

$$\epsilon(t) \approx -\Delta \ln \ln(\nu_0 t) \tag{57}$$

for the mean energy of the particle packet in energy space. However, while in the inelastic situation[13] $\nu_0 = \nu$, quasielasticity yields $\nu_0 = \omega\nu/\Delta$. Measurements on amorphous hydrogenated silicon yield $\nu_0 = 10^{12}$ Hz. Thus, if the inelastic theory holds, $\nu = 10^{12}$ Hz. On the other hand, the theoretical calculations yield values of the order $\nu \approx 10^{18}$ Hz and larger (see, e.g., Refs. 29 and 30) for the band tail of amorphous hydrogenated silicon. These estimates have been verified experimentally by frequency resolved photoluminescense measurements (see, e.g., Ref. 31) and linewidth electron spin resonance measurements.[32] Accordingly, the predictions of the inelastic theory are not in line with the experiments, but differ from the experimental values by six orders of magnitude. Therefore, it cannot be claimed that the inelastic theory can account for the experimental situation. By contrast, no problem arises for our quasielastic theory. For the quasielastic theory this fact merely confirms that ω can indeed be considered as super small. We conclude, that the existing experiments on transport in band tails of amorphous hydrogenated silicon are much better described by our quasielastic theory than by their inelastic counterparts.

VII. CONCLUSIONS

If we investigate our calculation we conclude that the frequency dependence of the hopping conductivity at low frequencies can significantly be affected by the form of the density of states, if there is no restriction of the transport to a vicinity of the Fermi energy. In the strongly localized regime such a situation occurs, e.g., if the density of states increases strongly with energy, as is the case in the exponential density of states investigated in our paper, where the dc conductivity results from the vicinity of the transport energy.[18,19] The transport energy reflects the competition between the Boltzmann factor, which favors contributions from low energies, and the mobility, which favors contributions with high energies.[19] The latter tendency results from the exponential dependence of the mobility on the characteristic hopping length, which, for a strongly energy dependent density of states increasing with energy, decreases strongly with energy. An alternating electric field leads to a reduction of the characteristic hopping length, and thus affects the above-mentioned competition. For lowest frequencies and in the multiple hopping regime this reduction is described by Eqs. (24) and (26). According to these equations, electrons at the top of the tail are much less affected by the alternations of the electric field than electrons at the bottom of the tail. Thus, although the main contribution to the dc conductivity results from the vicinity of the transport energy, the frequency dependence of the conductivity is determined by the electrons on the bottom of the tail.

Since the frequency dependence is governed by the electrons at the bottom of the tail the characteristic frequency, which governs the impact of the frequency of the alternating electric field, is not of the order of the dc conductivity, but of the order of the spectral conductivity at the bottom of the tail. Since the spectral conductivity in the vicinity of the bottom of the tail is much smaller than the spectral conductivity in the vicinity of the transport energy the dynamical conductivity already depends on frequency for very low frequencies. Furthermore, owing to the same reasons, the multiple hopping regime is very narrow. The Pollak-Geballe limit is practically already reached at relatively low frequencies.

If we compare the frequency dependence of the dynamical conductivity in the multiple hopping regime in the exponential tail further with that in the ordinary situation we also find that the character of the frequency dependence is different. In contrast to the ordinary situation, in which an increase of the frequency in the multiple hopping regime results in a strong increase of the real part of the conductivity, we find that the loss in the tail, although it is increasing with frequency, is practically extremely small. Furthermore, both s' and s'' are equal to one, within the multiple hopping regime.

The peculiarities in the frequency dependence also manifest themselves in the relaxation current. Since the characteristic frequency is tiny small anomalous diffusion is also observed at relatively large times. Within the range of applicability of the Pollak-Geballe limit the excess current decays like $\delta j(t) \propto t^{-[1-\alpha(t)]}$, with $1 > \alpha > 0$, but in the range of applicability of the multiple hopping regime just like $\delta j(t) \propto t^{-1}$. Owing to the same reasons discussed above, the Pollak-Geballe limit is also observed in the excess current for relatively large times.

Unfortunately, it turns out to be very difficult to compare our results directly with experiments, e.g., with results of measurements of the frequency dependence of the conductivity in amorphous hydrogenated silicon. The reason for this is that usually the idealized situation is not metin the experiment. In the experiments, both the contributions from holes in the valence band tail, from the electrons in the conduction band tail and from electrons in the conduction band, mix. Despite this fact, our results are qualitatively in line with the experimental results. According to the experiments[33–35] the Pollak-Geballe limit is also observed at low temperatures and at relatively low frequencies. The lowest frequency used in these experiments was 210 Hz. This fact is in line with our result that the characteristic frequencies are determined by the electrons at the bottom of the tail, since otherwise the Pollak-Geballe could clearly not be observed at such low frequencies. At higher temperatures the authors of the Refs. 33–35 find that the frequency dependence of the conductivity is well described by Eq. (1). They argue that this result is not due to hopping of electrons in the conduction band tail, but due to macroscopic inhomogenities. Our calculation gives further support to these hypotheses.

APPENDIX: SOME INTEGRALS

Here we discuss the main steps in the calculation of the integral (43). To this end we write Eq. (43) in the form

$$\sigma(s)-\sigma(0)=\frac{1}{5}\,\frac{e^2n(T)}{(kT)^2}\,\frac{\nu\Delta}{(2\alpha)^2}\,\frac{1}{\rho_0^{\rho_t-1}}I(\tilde{s}), \tag{A1}$$

where

$$I(\tilde{s})=\int_1^{\rho_0}d\rho\,\exp(\rho_t\ln\rho-\rho)(\exp\{W[\tilde{s}\rho\exp(\rho)]\}-1), \tag{A2}$$

and $\tilde{s}=s/(6\nu)$. To calculate the integral we focus on its derivatives with respect to $\tilde{s}$. We have $I(0)=0$. Furthermore, since for large ρ,

$$\frac{d}{d\tilde{s}}\Big|_{\tilde{s}=0}\exp\{W[\tilde{s}\rho\exp(\rho)]\}\approx\rho\exp(\rho), \tag{A3}$$

we obtain

$$\begin{aligned}I'(0)&=\int_1^{\rho_0}d\rho\,\exp(\rho_t\ln\rho-\rho)\frac{d}{d\tilde{s}}\bigg|_{\tilde{s}=0}\exp\{W[\tilde{s}\rho\exp(\rho)]\}\\&\approx\frac{{\rho_0}^{\rho_t+2}}{\rho_t+2}.\end{aligned} \tag{A4}$$

When calculating the second derivative of $I(\tilde{s})$ with respect to $\tilde{s}$ we again take into account that the main contribution to the integral results from large ρ. In this case

$$\frac{d^2}{d\tilde{s}^2}\exp\{W[\tilde{s}\rho\exp(\rho)]\}=-\frac{\rho^2\exp(\{2\rho-W[\tilde{s}\rho\exp(\rho)]\}}{\{1+W[\tilde{s}\rho\exp(\rho)]\}^3}. \tag{A5}$$

If we use Eq. (A5) and change the variables of integration from ρ to W we obtain the equation

$$\frac{d^2I(\tilde{s})}{d\tilde{s}^2}=-\frac{1}{\tilde{s}}\int_{W(\tilde{s}e)}^{W_0}dW\frac{\exp((\rho_t+1)\ln[\rho(W)])}{(1+W)^2}. \tag{A6}$$

Here $W_0=W[\tilde{s}\rho_0\exp(\rho_0)]$. Since for large ρ,

$$\rho(W)\approx\ln[\exp(W)W/\tilde{s}], \tag{A7}$$

Eq. (A6) can also be written in the form

$$\frac{d^2I(\tilde{s})}{d\tilde{s}^2}=-\frac{1}{\tilde{s}}\int_{W(\tilde{s}e)}^{W_0}\frac{dW}{(1+W)^2}\{\ln[\exp(W)W/\tilde{s}]\}^{\rho_t+1}. \tag{A8}$$

To simplify the integral further, we introduce the new integration variable u to $W=[(1+W_0)u+W_0]$. Doing so, we obtain

$$\begin{aligned}\frac{d^2I(\tilde{s})}{d\tilde{s}^2}=&-\frac{1}{\tilde{s}(1+W_0)}\int_{(W(\tilde{s}e)-W_0)/(1+W_0)}^{0}\frac{du}{(1+u)^2}\\&\times\{\ln[\exp(\rho_0)\rho_0]+(1+W_0)u\\&+\ln[1+(1+W_0)u/W_0]\}^{\rho_t+1}.\end{aligned} \tag{A9}$$

The main contribution to the integral results from the first term in the logarithm. Therefore, we simply obtain

$$\begin{aligned}\frac{d^2I(\tilde{s})}{d\tilde{s}^2}=&-\frac{1}{\tilde{s}(1+W_0)}\int_{(W(\tilde{s}e)-W)_0/1+W_0}^{0}\\&\times\frac{du}{(1+u)^2}\{\ln[\exp(\rho_0)\rho_0]\}^{\rho_t+1}.\end{aligned} \tag{A10}$$

It can be checked that the corrections from the second and the third term in the logarithm are negligible if the inequality $\rho_tW_0/\rho_0\ll1$ is satisfied. In realistic systems this inequality is satisfied in the whole multiple hopping regime. If we use this simplification we obtain

$$\frac{d^2I(\tilde{s})}{d\tilde{s}^2}=-\rho_0\exp(\rho_0)\{\ln[\exp(\rho_0)\rho_0]\}^{\rho_t+1}\frac{\exp(-W_0)}{1+W_0}. \tag{A11}$$

To obtain $I(\tilde{s})$ we integrate Eq. (A11) twice with respect to $\tilde{s}$ and take into account Eq. (A3). Then we obtain

$$\begin{aligned}I(\tilde{s})=&\tilde{s}\frac{{\rho_0}^{\rho_t+2}}{\rho_t}-\rho_0^{\rho_t}\exp(-\rho_0+W_0)[(1-W_0+W_0^2)\\&-\exp(-W_0)],\end{aligned} \tag{A12}$$

which yields Eq. (44).

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