Excess carrier diffusion in the field of deep trapping centers

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Diffusional spreading of excess carriers in the presence of deep trapping centers is studied under conditions of time-dependent thermal equilibrium. Exact formulas are used to show that for intermediate times corresponding to rapid trapping of carriers into spatially distributed localized states, the problem reduces to calculating the spectrum of a quantum particle in a disordered system. This approach is used to establish that for intermediate times, the diffusion obeys a universal law which is determined by the dimensionality of the system and is essentially nonlinear. The relaxation for large times is exponential even for weak fields, and the average relaxation time τ is inversely proportional to the square of the electric field E and is independent of the order parameters. The existence is predicted of a critical field above which the relaxation for large times is determined by the drift of carriers in randomly distributed trapping centers directed along the applied field; in this case, τ is $\sim 1/E$. The coherent potential approximation (CPA) correctly describes the final stage of excess carrier thermalization.

1. INTRODUCTION

Studies of the current associated with injected carriers provide an effective method for analyzing disordered materials.[1,2] In systems where the Fermi level ε_F lies below the edge ε_c of the conduction band, the relaxation of injected carriers with energy $\varepsilon \geqslant \varepsilon_c$ i.e., their distribution over a region of localized states for the entire system, is governed by the requirement that the system be in thermal equilibrium. As far as the kinetics of the excess carriers is concerned, the initial disorder can be modeled as a random spatial distribution of localized states which act as traps with randomly distributed energy levels. We will focus our attention primarily on the spatial disorder of the trapping centers, because this aspect of the problem has not been adequately studied. The previous work along these lines (reviewed in Refs. 1 and 2) invoked the mean potential approximation; on the other hand, the deep centers that trap the carriers most effectively are clearly randomly and sparsely distributed. We will see below that we thus arrive at a problem which is similar to the original problem of calculating the properties of a quantum particle in a random potential.

Carriers can make transitions between different localized states by diffusing along a region of delocalized states (along bands). We will assume for the sake of generality that the carriers move by a sequence of classical "hops," because the states involved in carrier transport along a band may be assumed to be continuous. On the other hand, it is also clear that the carrier motion near the edge of the conduction band will also have some of the features characteristic of "hopping" transport. The above model could be applied directly to the class of amorphous semiconductors with deep impurity levels in the forbidden band. The reader may consult Refs. 1 and 2 for further applications and physical discussions. In this model, the equations

$$\frac{\partial}{\partial t} D_n = u_n^- P_n - u_n^+ D_n + \sum_g (w_{n,n+g} D_{n+g} - w_{n+g,n} D_n), \qquad (1)$$

$$\frac{\partial}{\partial t} P_n = u_n^+ D_n - u_n^- P_n \qquad (2)$$

describe the dynamics of the excess carriers.

Here D_n (P_n) is the conditional probability for finding a carrier at site n in a conducting (localized) state if the carrier was present at site m in a delocalized state at time $t = 0$; $W_{n+g,n}$ is the probability that the carrier will hop between two adjacent sites and is given by

$$w_{n+g,n} = \bar{\omega} \exp\left(-2\frac{|\mathbf{r}_n - \mathbf{r}_{n+g}|}{r_B} + \frac{e(\mathbf{r}_{n+g} - \mathbf{r}_n)\mathbf{E}}{2kT} \right) \qquad (3)$$

when a field E is present. Here $\mathbf{r}_n$ is the position vector of the n-th site, $\bar{\omega}$ (usually comparable to the phonon frequency) is the number of "attempted" hops, r_B is the Bohr radius, and E is the electric field. The quantity $u_n^- = \nu \exp(-|\varepsilon - \varepsilon_c|/kT)$ in (1) and (2) is the probability for a transition from a localized state of energy ε into the region of delocalized states. The probability u_n^+ for trapping into a localized state is comparable to the phonon frequency, $u_n^+ \sim \nu$. The traps are assumed to be randomly distributed among the lattice sites with concentration c; the sites themselves form a regular lattice with unit cell parameter a equal to the effective hopping length.

The case of band transport follows from (1) if we let $a \to 0$, i.e., take the continuous limit. In this case, $1/w$ is the average time to trapping and a is the corresponding mean free path. The above model is valid only if $a \ll l$, because the carriers move between the traps by diffusion only in this case. Here $l = \rho^{-1/d}$ is the average distance between the traps, ρ is the trap concentration, and d is the dimensionality of the lattice. On the other hand, $a \ll l$ is clearly false for deep localized states or for states near the edge of the conduction band. In principle, the mean potential model, e.g., can be used and the frequent trapping by shallow traps can be treated by renormalizing the parameters in Eqs. (1), (2) [cf. below, however]. We note that w is typically $\ll \nu$ for the hops but that $w \gg \nu$ for band transport; however, both w and ν are apparently model parameters only—our basic results are independent of the ratio w/ν.

We can combine Eqs. (1) and (2) and derive the closed equation

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$$D_{nm}(s)=D_{nm}^{0}(s)-\sum_{l}D_{nl}^{0}(s)\gamma_{l}(s)D_{lm}(s) \tag{4}$$

for the Laplace transform of the Green's function $D_{nm}(t)$, where

$$D_{nm}^{0}(s)=\int\frac{d^3k}{(2\pi)^3}\exp[i\mathbf{k}(\mathbf{r}_n-\mathbf{r}_m)]\frac{1}{s+w(0)-w(\mathbf{k})},$$
$$\gamma_l(s)=\frac{su_l^{+}}{s+u_l^{-}},\quad w(\mathbf{k})=\sum_{g}\exp[i\mathbf{k}(\mathbf{r}_{n+g}-\mathbf{r}_n)]w_{n+g,n}. \tag{5}$$

The random parameter γ_e in (4) is nonzero only for sites occupied by traps. The Green's function D_{nm} completely describes the dynamics of the carriers in the delocalized state region. In particular, we will show in the Appendix that the diffusion coefficient for the injected carriers in expressible in terms of D_{nm} (s) by

$$D(t)=D_0\chi(t),\quad \chi(s)=\sum_{n}\langle D_{nm}(s)\rangle=\langle D(s,\mathbf{k}=0)\rangle, \tag{6}$$

where $D_0=a^2w$ is the microscopic diffusion coefficient and $\chi(t)$ is the fraction of the electrons contained in the delocalized state region at time t. Similarly, the injected current is given by

$$\mathbf{I}_p(t)=Q\mathbf{v}_0\chi(t),\quad \mathbf{v}_0=\sum_{g}(\mathbf{r}_{n+g}-\mathbf{r}_n)w_{n+g,n}. \tag{7}$$

Here Q is the total injected charge and v_0 is the microscopic drift velocity; we have $v_0=\mu_0E$ to first order in the field, where the mobility μ_0 is related to the diffusion coefficient by the Einstein relation $\mu_0=(e/kT)D_0$.

2. RELAXATION TO A STEADY-STATE

We note that the random parameter λ_1 in (4) vanishes as $s\to0$. The coherent potential approximation (CPA) therefore correctly describes the exact solution of the problem in the low-frequency limit.[3] We will take the trap concentration to be small, $c\ll1$, since otherwise the system relaxes rapidly over times $\nu^{-1}=10^{-12}-10^{-13}$ s. If we then sum only the diagrams which do not have any intersections (CPA), we find that

$$\langle D_{nm}(s)\rangle=D_{nm}^{0}(s(1+\lambda(s))),$$
$$\lambda=cv[uv+s+svD_{mm}^{0}(s(1+\lambda))]^{-1}, \tag{8}$$

where D_{mm} is the autocorrelator and $u=\exp(-\varepsilon/kT)$; the trap depth ε is measured relative to the edge of the conduction band. Equations (8) and (7) imply that the steady-state drift velocity of the injected carries is

$$v_c=v_0u/(c+u). \tag{9}$$

This result has an obvious interpretation if we note that thermal equilibrium implies that

$$\chi_c=\frac{u}{c+u}=\frac{N\exp(-\varepsilon/kT)}{N\exp(-\varepsilon/kT)+N_t},$$

where N_t is the total number of traps and N is the number of conducting "sites." According to (9), the trapping will be appreciable if $u\ll c\ll1$, as we will henceforth assume.

We now derive the equation that governs the final stage of the thermalization process in the system. For $d=1$ and $s\ll w$, the autocorrelator $\langle D_{mm}\rangle$ is given by the expression

$$\langle D_{mm}(s)\rangle=[4w(s(1+\lambda)+w\Delta^2)]^{-1/2},$$
$$\Delta^2=\frac{v_0^2}{4wD_0}=\left(\frac{eaE}{2kT}\right)^2. \tag{10}$$

For $s\ll c^2w$ and $w\ll v/2c$, Eq. (8) has the solution

$$\lambda=8c^2w\{8wcu+s+[s(s+16cuw)]^{1/2}\}^{-1} \tag{11}$$

for $E=0$. In the limit$\to\infty$, the right-most singularity in $\chi(s)$ (i.e., the pole in λ (s) and the cut in the complex s plane from the origin to the point $s=-16cuw$ needed to make the square root single-valued) contributes to the diffusion coefficient, and we have

$$\frac{D(t)-D_c}{D_c}=\frac{2}{\pi}\int_0^{\infty}dy\left(\frac{1-y}{y}\right)^{1/2}\exp\left[-16y\frac{t}{\tau_c}\right], \tag{12}$$

where $\tau_c=l^2/D_c=1/cuw$ is the mean diffusion time between traps for a system in thermal equilibrium, and $l=a/c^{1/d}$ is the mean distance between the traps. It follows that the final approach to the steady-state regime is described by the power law

$$(D(t)-D_c)/D_c=(\tau_c/4\pi t)^{1/2},\quad t\gg\tau_c.$$

If $E\neq0$, (10) implies that the branch point for the square-root singularity moves from the origin to the point $-w\Delta^2/(1+\lambda)$; the power law for relaxation at large times thus is replaced by the exponential law

$$\frac{v(t)-v_c}{v_c}=\left(\frac{\tau_c}{4\pi t}\right)^{1/2}\exp\left\{-\left(\frac{\Delta}{c}\right)^2\frac{t}{\tau_c}\right\}. \tag{13}$$

Equations (12) and (13) can be interpreted physically as reflecting the effects of multiple scattering of the carriers by the traps over long periods of time. These effects are appreciably attenuated in a nonzero electric field, so that the relaxation become exponential.[4]

We now consider the three-dimensional case. The autocorrelator D_{mm} for small s can then be expressed in the form

$$\langle D_{mm}(s)\rangle=b_0-b_1[s(1+\lambda)+w\Delta^2]^{1/2}, \tag{14}$$

where

$$b_0=\frac{0{,}506}{2w},\quad b_1=\frac{1}{4\pi}\left(\frac{1}{w}\right)^{3/2}.$$

Solving (8) by iteration for $s\to0$, we find that

$$\lambda=\frac{c}{u}-s\frac{c}{u^2}\left(\frac{1}{v}+b_0\right)+b_1\frac{c^{3/2}}{u^{5/2}}s\left[s+\frac{u}{c}w\Delta^2\right]^{1/2}+O(s^2). \tag{15}$$

The mobility consequently relaxes to the steady-state value $\mu_c=(u/c)\mu_0$ in accordance with the equation

$$\frac{\mu(t)-\mu_c}{\mu_c}=\left(\frac{\tau_c}{4\pi t}\right)^{3/2}\exp\left\{-\left(\frac{\Delta}{c^{1/3}}\right)^2\frac{t}{\tau_c}\right\}, \tag{16}$$

which is analogous to (13); here $\tau_c=c^{1/3}/uw$. The behavior (16) can be traced to the right-hand singularity in λ (s), i.e., to the cut in the s-plane. Since it is clear on physical grounds that this singularity describes repeated trapping processes,

we will rewrite Eq. (8) for $\Delta = 0$ and $s \ll \Omega$ (where $1/\Omega = 1/\nu + b_0 = 1/\nu + 1/4w$) in the form

$$s = -u\Omega[1-\Omega(b_1\eta^{1/2}+c/\eta)]^{-1}. \tag{17}$$

Here $\eta = s(1+\lambda(s))$. This shows that s is not analytic as a function of η for η between 0 and η_m, where η_m is defined by

$$-\frac{c}{\eta_m^2}+\frac{b_1}{2}\frac{1}{\eta^{1/2}}=0.$$

We thus find that the endpoint of the cut lies at $\eta_m = (2c/b_1)^{2/3}$, which corresponds to $s_m = -u\Omega$.

In exactly the same way, we find that

$$\frac{\chi(t)-\chi_c}{\chi_c} = \frac{\tau_c}{4\pi t}\left(\ln\frac{t}{\tau_c}\right)\exp\left\{-\frac{\Delta^2}{c}\frac{t}{\tau_c}\right\} \tag{18}$$

for $d = 2$ and $t \gg \tau_c = 1/uw$. In this case the logarithmic singularity of $\lambda(s)$ dominates the behavior of (18), and the endpoint of the cut is

$$s_m = -4\pi w u\left(\ln\frac{1}{c}\right)^{-1} \quad \text{for } w \ll \frac{\nu}{4\pi}\ln\frac{1}{c}.$$

Equation (18) describes the final stage of carrier relaxation in a system in thermal equilibrium, i.e., how the already trapped carriers become redistributed among the localized states. Because this redistribution involves repeated carrier trapping and escape, the function $\chi(t)$ is small with respect to the parameter u for times t during the transient stage. Thus, in the low-temperature limit $u \to 0$ most of the drop in the observable quantities occurs during the initial stage of relaxation, when the free carriers are trapped into localized states. We will study the early relaxation process in the next section.

3. EQUIVALENCE WITH THE ANDERSON MODEL

We now consider the times for which $s \gg u^-$ (the time scales will be classified in more detail later). The distribution of the random parameter γ_1 in (4) then takes the form

$$\Pi(\gamma) = (1-c)\delta(\gamma)+c\delta(\gamma-\nu). \tag{19}$$

The problem for a continuous distribution of localized states can also be reduced to the form (19). Indeed, let $\rho(\varepsilon)$ be the distribution density for the localized states, with

$$\int_0^\infty d\varepsilon\,\rho(\varepsilon) = \frac{N_t}{N}.$$

By the definition of γ [cf. (5)], the states with $\varepsilon > \bar{\varepsilon} = kT\ln(\nu/s)$ may be regarded as deep trapping centers. Here $\gamma = \nu$ and the concentration of these centers is given by

$$c(s) = \int_{\bar{\varepsilon}}^\infty \rho(\varepsilon)\,d\varepsilon. \tag{20}$$

States with $\varepsilon < \bar{\varepsilon}$ correspond to shallow traps. In this case $\gamma \sim s$ and the CPA model[3] can be used to treat the shallow trapping ($\gamma \to s\lambda$). According to (4), this amounts to dividing the frequencies w and ν by the factor $(1+\lambda)$. This approach can be used to analyze an arbitrary distribution $\rho(\varepsilon)$; it also implies that the approximation (19) holds quite generally for appropriately bounded time intervals and low temperatures.

We see immediately from (4) that this equation coincides with the Dyson equation for the single-particle Green's function in the Anderson model, which is described by the Hamiltonian

$$H = \sum_{n,g} w_{n+g,n}(a_n^+ a_n - a_{n+g}^+ a_n) + \sum_n \gamma_n a_n^+ a_n. \tag{21}$$

Here a_n^+ and a_n are the creation and annihilation operators for an electron at site n, and the $w_{n+g,n}$ can now be regarded as overlap integrals. The equivalence between the two models can be expressed formally by the equations

$$D_{nm}(s) = -G_{nm}(-s), \tag{22}$$

$$G_{nm}(\omega) = \langle 0|a_n\frac{1}{\omega-H}a_m^+|0\rangle. \tag{23}$$

The Anderson model with diagonal disorder has been much studied recently (cf. the reviews in Refs. 5-7). However, before we can take over the results for the Anderson model, we must find a way to handle problems with overlap integrals which are symmetric with respect to the lattice sites. We can do this by noting the following exact equation for the Green's functions:

$$D_{nm}(s,E) = \exp\left(\frac{e(\mathbf{r}_n-\mathbf{r}_m)\mathbf{E}}{2kT}\right)D_{nm}(s+w\Delta^2,0), \tag{24}$$

$$w\Delta^2 = \sum_g [w_{n+g,n}(E) - w_{n+g,n}(0)]. \tag{25}$$

The proof is quite obvious for ordered systems and consists in choosing a new Boltzmann distribution corresponding to the final state to which the system relaxes in a nonzero electric field E. It can be shown that (24) remains valid for arbitrary types of disorders, including the disorder assumed in the bonding problem. We can verify (24) readily for the case of interest by directly substituting it into (4). The usefulness of (24) is evident, since it can be recast immediately in terms of the averaged Green's functions; this makes it possible to express the Green's function for $E \neq 0$ in terms of the Green's function for $E = 0$.

4. WEAK FIELDS

We will first analyze the autocorrelator D_{mm} in order to illustrate the use of Eqs. (22)–(25). According to (22) we can write

$$\langle D_{mm}(t)\rangle = e^{-\Delta^2 wt}p(t), \quad p(t) = \frac{-1}{2\pi i}\int_{-i\infty+0}^{i\infty+0} ds\,e^{st}\langle G_{mm}(-s)\rangle. \tag{26}$$

As a function of the complex variable s, function $G_{mm}(s)$ is well known to have a cut along the positive axis $\mathrm{Re}(s) > 0$, and the discontinuity across the cut is equal to the one-electron state density $n(s)$ for the Hamiltonian (21). We can thus recast the expression for $p(t)$ as

$$p(t) = \int_0^\infty ds\,n(s)e^{-st}. \tag{27}$$

We note that the obvious initial condition $p(0) = 1$ is satisfied. The state density $n(s)$ for the model (19), (21) behaves as[5]

$$n(s) \propto \exp\left\{-b_d c\left(\frac{2w}{s}\right)^{d/2}\right\} \tag{28}$$

near the true edge of the band, provided the Lifshits conjecture is valid; the numbers b_d were calculated in Ref. 8. If we substitute (28) into (27) and evaluate the resulting integral by the method of steepest descent, we get

$$p(t) = \exp\{-B_d (t/\tau_d)^{d/(d+2)}\}, \tag{29}$$

where

$$B_1 = 3(\pi/2)^{2/3}, \quad B_2 = 8{,}85,$$

$$B_3 = {}^5/_3 (2\pi^4)^{2/5}, \quad \tau = l^2/D_0 = 1/wc^{2/d}.$$

Here τ is the average diffusion time between the traps, and the saddle point used in evaluating (29) is given by

$$s_c = \frac{2}{\tau}\left(\frac{db_d}{4}\frac{\tau}{t}\right)^{2/(d+2)}$$

We recall that our analysis is valid only for $s \gg u^-$ because of our initial assumptions. The asymptotic formula (29) is therefore guaranteed to be valid only for times

$$t \ll \tau(\tau_t/\tau)^{(d+2)/d}, \tag{30}$$

where $\tau_t = 1/u^-$ is the average time during which a carrier remains confined in a trap. However, there are indications that (29) may also be correct even for t not satisfying (30). We saw previously (cf. Fig. 1) that for finite τ_t the function $\chi(s)$ has a cut in the complex plane from the origin to the point $s = s_m$. We can thus rewrite inequality (30) in the form $s_c \gg |s_m|$, which reduces to

$$t \ll \tau\left(\frac{c}{u}\right)^{3/2}, \quad t \ll \tau\left(\frac{c}{u}\ln\frac{1}{c}\right)^2, \quad t \ll \tau\left[\left(1+\frac{4w}{\nu}\right)\frac{c^{2/3}}{u}\right]^{5/2} \tag{31}$$

for $d = 1, 2, 3$, respectively.

Similarly, we can try to use Eqs. (6), (22), and (24) to find the time dependence of the function $\chi(t)$. Unlike the case of the autocorrelator, however, in this case we must know the analytic properties of the functions $G_{nm}(s)$ for $n \neq m$ as well as for $n = m$. It can be shown that the leftmost singularity of $G_{nm}(s)$ will again have the form of a cut, as we found above. The instanton method[7] can be used to find the discontinuity in $G_{nm}(s)$ across the cut, and a suitable technique was developed in Ref. 8 for solving the quantum problem of interest to us. A straightforward generalization of the results in Ref. 8 gives the leading term

$$\operatorname{Im} G_{nm}(s+i0) \propto n(s)\,\theta\left(\left|\frac{2w}{s}\right|^{1/2} - \frac{1}{a}|\mathbf{r}_n - \mathbf{r}_m|\right) \tag{32}$$

for the jump in $G_{nm}(s)$ across the slit as $s \to 0$ (here $\theta(x)$ is the Heaviside function). We have so far neglected the more detailed coordinate dependence because it is not needed to find the leading term in the asymptotic expansion. If we now combine Eqs. (6), (22), (24), and (32) and take inverse Laplace transforms, we find that

$$\chi(t) = \exp\left[-\Delta^2 wt - B_1\left(1-\frac{\Delta}{c}\right)^{2/3}\left(\frac{t}{\tau}\right)^{1/3}\right], \quad d=1, \tag{33}$$

$$\chi(t) = \exp\left[-\Delta^2 wt - B_d\left(\frac{t}{\tau}\right)^{d/(d+2)}\right], \quad d=2,3. \tag{34}$$

The result for $d = 1$ of course agrees with the exact solution found in Refs. 9 and 10. This asymptotic formula is useful because the first term in (33), (34) dominates for large times even for weak fields E. Indeed, the average relaxation time is $\sim 1/E^2$. The physical explanation is that the number of carriers is further decreased by the electric field because the system contains clusters of finite size.

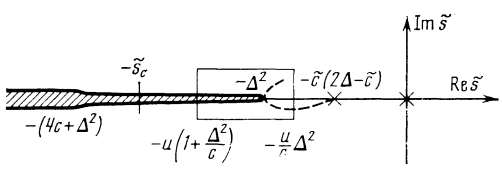

FIG. 1. Location of the singularities of $\chi(s)$ ($d = 3$) in the $\tilde{s}$-plane, where $\tilde{s} = s/w$, $w \ll \nu$. The value $\tilde{s} = -(4c + \Delta^2)$ corresponds to the square-root singularity in $\chi(s)$ and lies at the edge of the effective band found in the coherent potential approximation. The actual cut should in fact extend as far as $\tilde{s} = -\Delta^2$; however, the jump in $\chi(s)$ across the cut is exponentially small for $\tilde{s} > -4c + \Delta^2$. The value $\tilde{s} = -s_c$ corresponds to the saddle point in (29). For $u \neq 0$, this cut overlaps a second cut $[-u(1 + \Delta^2/c), -u\Delta^2/c]$ which determines the final stage of the relaxation process. In this case the residue at the pole $s = 0$ becomes nonzero. For strong fields $\Delta > \tilde{c}$, the pole at $\tilde{s} = -\tilde{c}(2\Delta - \tilde{c})$ leaves the second sheet by passing through the origin of the cut and enters the first (physical) sheet.

5. THE DRIFT REGIME

In the previous section we used Eqs. (22) and (24) to calculate $\chi(t)$; however, we considered only the contribution of the branch-point singularity of the function $G_{nm}(s)$. There are grounds for believing that G_{nm} has no other singularities as a function of s. The question then arises of determining whether all the singularities of $\chi(s)$ are caused by the branch point singularities. This question is of interest, because $\chi(s)$ is given as a sum of $G_{nm}(s)$ over the index n with factors that grow exponentially along the applied field. We thus anticipate that $\chi(s)$ may contain new singularities when the field is nonzero. In order to solve this problem we must know the complete Green's function $G_{nm}(s)$, at least near $s = 0$. In principle, this information could be obtained from the dispersion equations[7] for a specified imaginary part Im (s). However, this would require knowing the discontinuity in G_{nm} across the entire cut. On the other hand, we know that Im $(G_{nm}(s + i0))$ is exponentially small in the fluctuation region below the edge of the effective band (cf. below). This suggests that the CPA method should correctly describe the qualitative form of the Green's function far from the edge of the effective band. We will thus use the CPA method to look for other singularities (we will also give additional arguments in support of the final results).

For the range of parameter values of interest we can rewrite Eq. (8), which follows from the CPA approximation, directly for the function $\chi(s) = 1/s(1 + \lambda) = 1/\eta$ as follows:

$$s = \eta - c\nu[1 + \nu D_{mm}^0(\eta)]^{-1}, \tag{35}$$

where the autocorrelator $D_{mm}^0(\eta)$ is given by Eqs. (10) and (14). We will take $s \ll w$ and $\Delta \ll 1$ in what follows; moreover, if $c \ll 1$ we can invert Eq. (35). For $d = 1$ and $w \ll \nu/2c$, we thus find that

$$\tilde{\eta} = \tilde{s} + 2c^2 + 2c(\tilde{s} + c^2 + \Delta^2)^{1/2}, \tag{36}$$

where the frequency $\tilde{s}$ is now given in units of w. The physical branch in (36) corresponds to $\eta > 0$ for $s > 0$. Equation (36) then yields the expression

$$\chi(s) = [(\tilde{s}+c^2+\Delta^2)^{1/2}-c-\Delta][(\tilde{s}+c^2+\Delta^2)^{1/2}-c+\Delta] / [w(\tilde{s}-2c\Delta)(\tilde{s}+2c\Delta)] \quad (37)$$

for $\chi(s)$. The square-root singularity in (37) coincides with the cut in the function $G_{nm}(s)$ which we discussed above. The value $s = -wc^2$ corresponds to the edge of the effective band. According to (24), the branch point for $E \neq 0$ is shifted by $w\Delta^2$ from the edge of the band. The contribution from the cut in the square root function can be expressed in the form

$$\chi(t) = \frac{c}{\pi}\int_0^\infty \frac{y^2\,dy\,\exp[-wt(\Delta^2+c^2+y^2)]}{[y^2+(\Delta+c)^2][y^2+(\Delta-c)^2]} \quad (38)$$

[cf. (33)]. The pole for fields $\Delta > c$ in (37) is new; its contribution to $\chi(t)$ is given by

$$\chi_p(t) = (1-c/\Delta)\exp(-2c\Delta wt). \quad (39)$$

On the other hand, the exact result for $d = 1$ is[9,10]

$$\chi_p(t) = \left(1-\frac{c}{\Delta}\right)\exp[-c(2\Delta-c)wt]. \quad (40)$$

Comparison of (39) and (40) shows that the CPA correctly describes the dependence on the field. We can account for the difference in the exponents (by a factor of 2 when $\Delta = c$) by recalling that in the CPA, the pole must pass through the branch point $s = -w(c^2+\Delta^2)$ of the square root in order to reach the physical sheet; on the other hand, $s = 0$ gives the actual edge of the band in the exact solution, and the initial point on the cut thus lies at $s = -w\Delta^2$. This is responsible for the difference wc^2 between the exponential arguments in (39) and (40).

We now discuss the case when $d = 3$. Proceeding as above, we find for $w \ll \nu$ that

$$\chi(s) = \frac{1}{w}\frac{\tilde{s}+2\tilde{c}^2-2\tilde{c}[\tilde{s}+\tilde{c}^2+\Delta^2]^{1/2}}{\tilde{s}^2-(2\tilde{c}\Delta)^2}, \quad (41)$$

where $\tilde{s} = s/w + 4c$ and $\tilde{c} = 2c/\pi$. Here the position of the square-root singularity corresponds to the edge of the effective band, $s = -4cw[1+0(c)]$ (cf. Ref. 6) shifted by the amount $w\Delta^2$. The contribution of this cut to $\chi(s)$ is given by an expression similar to (38). The existence of the new pole for fields $\Delta > \tilde{c}$ is nontrivial; its contribution to the time dependence $\chi(t)$ is given by

$$\chi_p(t) = (1-\tilde{c}/\Delta)\exp[(-2\tilde{c}\Delta+4c)wt]. \quad (42)$$

If we recall that the pole can only reach the physical sheet by passing through the branch point on the cut that corresponds to the true edge of the band [i.e., $s = -w\Delta^2$ rather than $s = -w(4c+\Delta^2)$], we see that the correct expression for the pole contribution is

$$\chi_p(t) = (1-\tilde{c}/\Delta)\exp[-\tilde{c}(2\Delta-\tilde{c})wt]. \quad (43)$$

This agrees with the exact asymptotic expression (34) for $\Delta = \tilde{c}$. We note that (43) is the dominant contribution for large times.

The asymptotic expressions (40) and (43) can also be derived as follows. It is clear on physical grounds that carrier drifting in large clusters is responsible for the above behavior. We can use perturbation theory to study the relaxation within a given cluster; keeping only the lowest-order term (equal to D^0_{nm}) in perturbation expansion, we can estimate the corresponding contribution to the time dependence $\chi(t)$ by

$$\chi_p(s) \sim \sum_n R_{nm}(c)D_{nm}{}^0(s), \quad (44)$$

where $R_{nm}(c)$ is the statistical weight of the cluster. Since we are interested in the mathematical origin of the new singularities in $\chi(s)$ for nonzero fields, we will consider only clusters with the largest statistical weights in (44); allowance for the other contributions to $\chi(s)$ (including the interaction of carriers with traps in larger clusters) will alter only the coefficient of the exponential in $\chi_p(t)$. In order of magnitude we then clearly have

$$R_{nm}(c) \sim (1-c)^{\varkappa}, \quad \varkappa = \sum_{j=1}^{d} |n_j - m_j|.$$

We can rewrite R_{nm} as

$$R_{nm} \sim \exp[-\rho\sigma|\mathbf{r}_n-\mathbf{r}_m|], \quad (45)$$

in the continuum limit, where $\rho = l^{-d}$ is the trap concentration and $\sigma = a^{d-1}$ is the trapping cross section. The Green's function can then be expressed in the form

$$D_{nm}{}^0(s) \sim \exp\left[-\frac{1}{2D_0}(4sD_0+v_0^2)^{1/2}|\mathbf{r}_n-\mathbf{r}_m| + \frac{1}{2D_0}\mathbf{v}_0(\mathbf{r}_n-\mathbf{r}_m)\right]. \quad (46)$$

Substitution into (44) shows that $\chi(s)$ has a new pole on the physical sheet with $-w\Delta_2 < s < 0$ if $\Delta > c$ (i.e., $v_0 > 2D_0\sigma\rho$). Its contribution to $\chi(t)$ is similar in form to (43):

$$\chi_p(t) \sim \exp\left[-t\left(\frac{1}{\bar{\tau}_{dr}}-\frac{1}{\bar{\tau}_{dif}}\right)\right]. \quad (47)$$

Here $\bar{\tau}_{dr} = \tilde{l}/v_0 \sim c\Delta w/2$ is the average drift time between the traps, and $\tilde{l} = 1/\sigma\rho = l(l/a)^{d-1}$, $\tilde{\tau}_{dif} = \tilde{l}^2/D_0 = (wc^2)^{-1}$ is the time required for a carrier to diffuse a distance $\tilde{l}$. The condition $\Delta > c$ clearly implies that $\tilde{\tau}_{dr} < 2\tilde{\tau}_{dif}$. The calculations can be done for arbitrary fields and concentrations if $d = 1$. For $(1-c)e^{\theta} > 1$ we find that

$$\chi_p(t) \sim \exp\left[-cwt\left(e^{\theta}-\frac{e^{-\theta}}{1-c}\right)\right], \quad \theta = \frac{lEa}{2kT}, \quad (48)$$

which agrees with the exact result.[9,10] The set of diagrams which describe the carrier-trap interaction in large clusters must be considered in order to calculate the coefficient of the exponential in (48); however, its structure is correctly given by Eqs. (39) and (42), which were derived in the coherent potential approximation.

We also note that the above derivation for the asymptotic time behavior of $\chi_p(t)$ in fact exhausts all the contributions and becomes exact for large E, in which case the carriers follow the field. In this case the CPA also gives the exact result (there are no interference diagrams)[11]

$$\chi(t) = \exp[-d\tilde{w}tc], \quad (49)$$

where $\tilde{w}$ is the hopping frequency along the field, which is assumed to lie along the long diagonal of the elementary lattice cell.

We have assumed above that $w \ll \nu$ in the three-dimensional system. In the opposite limit $w \gg \nu$ every carrier that reaches a site occupied by a trap will be captured, and Eq. (43) remains valid. One shows without difficulty (using the CPA, e.g.) that in this case the constant $\tilde{c}$ in (43) is given by

$$\tilde{c} = \frac{2c}{\pi}\left(\frac{\Omega}{4w}\right)^2, \quad \frac{1}{\Omega} = \frac{1}{\nu} + \frac{1}{4w}. \tag{50}$$

For strong fields (directed carrier motion, $\Delta \gtrsim 1$), the expression for $\chi(t)$ for arbitrary ratios ν/w becomes

$$\chi(t) = \exp\left[-c\tilde{\Omega}t\right],$$

where $1/\tilde{\Omega} = 1/\nu + 1/d\tilde{w}$. For $d = 1$ or 2, Eqs. (40) or (47) remain meaningful for $w \ll \nu/2c$ and $w \ll (\nu/4\pi)\ln(1/c)$. Thus the dimensionality of the system affects only the time scales for fields above the critical value.

DISCUSSION

We have derived several exact formulas for the kinetic characteristics of a disordered system in which the relaxation primarily involves transitions of excess carriers to spatially distributed localized states. We can arbitrarily divide this process into two stages corresponding to large and small times. For large times $t \gtrsim \tau_c$, where $\tau_c = l^2/D_c = (uwc^{2/d-1})^{-1}$ is the average diffusion time between traps for a system in thermal equilibrium, the relaxation to a steady-state regime is complete and is described by a $(\tau_c/d)^{d/2}$ law. The relaxation process during this stage is greatly influenced by the electric field; in this case the power law breaks down and the relaxation becomes exponential with time constant $\tau_c(2kT/eEl)^2$.

Most of the decrease in the observable quantities occurs for short times corresponding to the initial trapping of free carriers into localized states. The characteristic time scale during this stage is $\tau = l^2/D_0 = 1/wc^{2/d} \ll \tau_c$, i.e., it is equal to the microscopic carrier diffusion time between the traps; the corresponding behavior is given by Eqs. (33) and (34). We suggest the following interpretation. The relaxation in a cluster of length L is proportional to $e^{-t/T}$, where $T = L^2/D_0$, and the probability of finding such a cluster in the system is equal to $\exp(-\rho L^d)$. The desired expression (34) thus follows by maximizing the product $e^{-t/T} \cdot \exp(-\rho L^d)$ with respect to L.

We found that the initial stage of relaxation is also greatly influenced by the electric field. For weak E, the relaxation is exponential with time constant $\tau = 1/w\Delta^2$ for large times. We note that τ is independent of the order parameters. The physical explanation is as follows. In a nonzero field, the probability that a carrier will remain at a given site decays with time as $\exp(-w\Delta^2 t)$ even if there are no traps [cf. (24)], because the electric field tends to sweep the carriers away. However, for an infinite cluster the carrier concentration at each site must remain constant even for $E \neq 0$ because carriers are supplied from adjacent sites. For finite clusters, this is no longer true—in this case, the carriers are concentrated at the boundary of the cluster and therefore play no role. The electric field thus decreases the number of active carriers by the factor

$$\exp(-w\Delta^2 t) = \exp(-V_0^2 t/4D_0).$$

Another important effect of the field is to direct the motion of the carriers. As a result, the clusters (which are elongated along the field) start to become more important and for field strengths above a critical value, the carrier drifting in the clusters is the slowest process in the system. This process can be analyzed qualitatively by straightforward physical arguments. Indeed, the contribution to the drift current at time t comes from clusters whose length along the field is greater than $v_0 t$ and whose cross sectional area exceeds the trapping cross section σ. In order of magnitude, the relative number of such clusters in the system is equal to $\exp(-\rho v_0 t\sigma)$, which roughly speaking also describes the time behavior $\chi(t)$, since the spreading of the carriers normal to the field retards the falloff in $\chi(t)$ [cf. Eqs. (33), (34)]. If we compare this relaxation behavior with the behavior found for relaxation in isotropic clusters, which is of the form $\exp(-v_0^2 t/4D_0)$, we find that $V_k \approx D_0\sigma\rho$. Thus for strong fields, the carriers drift prior to being captured by the traps, whereas they move by diffusion for weak fields.

We close by noting that the above physical arguments supporting the asymptotic formulas derived in the previous sections are quite general. This suggests that similar time dependences should be observed for other disordered models, e.g., in the bonding problem. This was demonstrated rigorously in Refs. 9 and 10 for the one-dimensional case.

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APPENDIX

Proceeding from first principles, we can write

$$\mathbf{I}_p(t) = Q\frac{d}{dt}\sum_n \langle (\mathbf{r}_n - \mathbf{r}_m)\,(D_{nm}(t) + P_{nm}(t))\rangle$$

for the current produced by carriers injected into the conduction band. We can evaluate the sum by multiplying Eq. (4) by $\mathbf{r}_n - \mathbf{r}_m$ and summing over all n:

$$\sum_n (\mathbf{r}_n - \mathbf{r}_m)\,(D_{nm}(s) + P_{nm}(s))$$

$$= \left[\sum_n (\mathbf{r}_n - \mathbf{r}_l) D_{nl}^0(s)\right]\left[1 - s\sum_l P_{lm}(s)\right].$$

Here we have used $sP_{nm} = \gamma_n D_{nm}$ and $\Sigma_n D^0_{nm} = 1/s$ [cf. Eq. (2)]. We obtain Eq. (1) by averaging the last equation and using conservation of particle number: $\Sigma\langle P_{nm} + D_{nm}\rangle = 1/s$.

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