Effect of scattering on the dynamic localization of a particle in a time-dependent electric field

D. H. Dunlap and V. M. Kenkre
Department of Physics and Astronomy, University of New Mexico, Albuquerque, New Mexico 87131
(Received 7 October 1987)

The theory of dynamic localization of a charged particle moving on a lattice under the influence of a time-dependent electric field developed in a previous paper is extended to include the effect of scattering of the particle by imperfections in the lattice. The description used to include scattering is that provided by the stochastic Liouville equation. Exact solutions for the mean-square displacement are obtained, and the average diffusion constant is calculated for a sinusoidal ac field. Scattering is found to play a dual role: It increases diffusion by preventing localization, and decreases it by increasing the amount of incoherence. Viewed another way, an alternating electric field is seen to result in an increase in the scattering rate in general and the appearance of singularities in the rate for specific values of the field magnitude and/or the field frequency. These singularities represent the phenomenon of dynamic localization and indicate the possibility of inducing anisotropy in isotropic materials through the application of strong time-varying electric fields.

I. INTRODUCTION AND THE STOCHASTIC LIOUVILLE EQUATION

In a previous paper[1] (henceforth referred to as I) we addressed the motion of a charged particle moving among the sites of a lattice under the combined action of intersite transfer interactions and a time-dependent electric field. From an explicit solution of the time-dependent Schrödinger equation, we found that, for an ac field, a charged particle will undergo dynamic localization for certain values of the frequency and the magnitude of the field. The present paper is an extension of the theory in I to include the effect of scattering of the charge carrier caused by its interactions with imperfections of the lattice. The effect of scattering is taken into account through a stochastic Liouville equation.[2-7] Exact solutions are found for the mean-square displacement and used to investigate the effects of the applied field on the scattering rate and the diffusion constant of the particle. Explicit calculations of the current are also given.

As in I, we begin our analysis with the simple case of a one-dimensional lattice of sites m $(-\infty < m < \infty)$ with nearest-neighbor intersite interactions V and lattice constant a. If e is the charge on the carrier, and $E(t)$ the electric field, the Hamiltonian of the carrier in the absence of scattering is [see Eq. (1.1) of I]

$$H(t)=V\sum_m \{ |m\rangle\langle m+1| + |m+1\rangle\langle m| \} - eE(t)a\sum_m m\{ |m\rangle\langle m| \} . \quad (1.1)$$

Following the notation of I, we write the quantity $eE(t)a$ as $\mathcal{E}f(t)$, where the dimensionless $f(t)$ contains the time dependence of the field and $\mathcal{E}$ is the magnitude of the field energy, i.e., the product of the field magnitude and ea, express the particle state $|\psi(t)\rangle$ as a linear combination of Wannier states $|m\rangle$, i.e., $|\psi(t)\rangle = \sum_m C_m(t)|m\rangle$, and take $\hbar=1$. The Wannier-state amplitudes, $C_m(t)$, then obey the evolution equation [see Eq. (1.4) of I],

$$i\frac{dC_m}{dt} = -m\mathcal{E}f(t)C_m + V(C_{m+1}+C_{m-1}) . \quad (1.2)$$

In order to include the effect of scattering, we first convert the amplitude equation (1.2) into an equation for the density matrix elements $\rho_{m,n}(t) \equiv C_m^*(t)C_n(t)$,

$$i\frac{\partial}{\partial t}\rho_{m,n} = \mathcal{E}f(t)(m-n)\rho_{m,n} + V(\rho_{m,n+1}+\rho_{m,n-1}-\rho_{m+1,n}-\rho_{m-1,n}) . \quad (1.3)$$

We then add to the right-hand side of (1.3) a term describing the destruction of the off-diagonal elements of the density matrix in the site representation at a constant rate α. The resulting equation,

$$i\frac{\partial}{\partial t}\rho_{m,n} = \mathcal{E}f(t)(m-n)\rho_{m,n} + V(\rho_{m,n+1}+\rho_{m,n-1}-\rho_{m+1,n}-\rho_{m-1,n}) - i\alpha(1-\delta_{m,n})\rho_{m,n} , \quad (1.4)$$

is the stochastic Liouville equation (SLE) in its simplest form. The SLE has been used widely as a simplified tool for the unified description of coherent and incoherent transport.[2-7] The ratio V/α measures the mean free path of the particle in units of the lattice constant. Equation (1.4) is our starting point in this paper.

While the SLE can be solved exactly[2-7] in the absence of the electric field, solutions for the probabilities are difficult to find in the presence of the field. However, explicit solutions can indeed be obtained for derived quantities. We present such an exact solution for the mean-square displacement in Sec. II. In Sec. III we use that solution to calculate the average diffusion constant for the case that the electric field is sinusoidal in time. An investigation of the diffusion constant exhibits the phenomenon of dynamic localization in the limit that the field frequency is large in comparison to the scattering rate. In Sec. IV we calculate the ac current through a slight generalization of the SLE to arbitrary temperatures. A discussion of our results as well as an explicit extension of the analysis to dimensions higher than one comprise Sec. V.

II. EXACT SOLUTION FOR THE MEAN-SQUARE DISPLACEMENT

In attempting to solve (1.4) for $\rho_{m,n}(t)$, one may proceed in a fashion analogous to that used in I. A discrete Fourier transform over the site labels m and n produces the momentum-space form of the SLE:

$$i\frac{\partial}{\partial t}\rho^{k,q}=i\mathcal{E}f(t)\left[\frac{\partial}{\partial k}+\frac{\partial}{\partial q}\right]\rho^{k,q}$$
$$+2V[\cos(k)-\cos(q)]\rho^{k,q}-i\alpha\rho^{k,q}$$
$$+i\alpha\sum_m \rho_{m,m}e^{i(k-q)m}\ , \tag{2.1}$$

where $\rho^{k,q}=\sum_{m,n}\rho_{m,n}e^{i(km-qn)}$. The definition

$$\eta(t)=\int_0^t dt' f(t') \tag{2.2}$$

and the method of characteristics detailed in I can be shown to lead from (2.1) to

$$\rho^{k,q}(t)=\rho_0^{k-\mathcal{E}\eta(t),q-\mathcal{E}\eta(t)}\psi^{k,q}(t,0)e^{-\alpha t}$$
$$+\alpha\int_0^t dt'\left[\sum_m e^{i(k-q)m}\rho_{m,m}(t')\right]$$
$$\times\psi^{k,q}(t,t')e^{-\alpha(t-t')}\ , \tag{2.3}$$

which expresses $\rho^{k,q}(t)$ in terms of its initial value $\rho_0^{k,q}$, with k and q replaced by $k-\mathcal{E}\eta(t)$ and $q-\mathcal{E}\eta(t)$, respectively. The "density-matrix propagator" $\psi^{k,q}(t,t')$ appearing in (2.3) is defined as

$$\psi^{k,q}(t,t')=\exp\left[2Vi\int_{t'}^t dt''\{\cos[k-\mathcal{E}\eta(t)+\mathcal{E}\eta(t'')]\right.$$
$$\left.-\cos[q-\mathcal{E}\eta(t)+\mathcal{E}\eta(t'')]\}\right]. \tag{2.4}$$

An alternate derivation[8] of (2.3) from (1.4) may be of interest. The definition

$$g_{m,n}(t)=\rho_{m,n}(t)e^{i\mathcal{E}(m-n)\eta(t)} \tag{2.5}$$

removes the first term on the right-hand side of (1.4):

$$i\frac{d}{dt}g_{m,n}(t)=V[(g_{m,n+1}-g_{m-1,n})e^{-i\mathcal{E}\eta(t)}$$
$$+(g_{m,n-1}-g_{m+1,n})e^{i\mathcal{E}\eta(t)}]$$
$$-i\alpha(1-\delta_{m,n})g_{m,n}\ . \tag{2.6}$$

When transformed into momentum space, (2.6) takes the form

$$i\frac{d}{dt}g^{k,q}(t)=2V\{\cos[q-\mathcal{E}\eta(t)]-\cos[k-\mathcal{E}\eta(t)]\}g^{k,q}$$
$$-i\alpha g^{k,q}+i\alpha\sum_m g_{m,m}e^{i(k-q)m}\ , \tag{2.7}$$

which is then solved formally to give (2.3).

Equation (2.3) is by no means a real solution for $\rho^{k,q}(t)$ since its right-hand side contains the unknown $\rho_{m,n}(t)$. Following a procedure used by Kenkre and Brown[4] to solve the field-free SLE in the absence of the field, we set $k=k'$ and $q=k'-k$ in (2.3) and obtain the evolution of the quantity $P^k(t)$ defined by

$$P^k(t)=\sum_{k'}\rho^{k',k'-k}(t)=(1/N)\sum_m \rho_{m,m}e^{ikm}\ , \tag{2.8}$$

where N is equal to $\sum_m 1$. This Fourier transform of the probability of site occupation is seen to obey

$$P^k(t)=e^{-\alpha t}\theta^k(t)+\alpha\int_0^t e^{-\alpha(t-t')}P^k(t')\Psi^k(t,t')dt'\ . \tag{2.9}$$

Here $\theta^k(t)$ is $P^k(t)$ in the absence of scattering. It involves the particular initial condition in the problem, and is given by

$$\theta^k(t)=\sum_q \rho^{q-\mathcal{E}\eta(t),q-k-\mathcal{E}\eta(t)}(0)\psi^{q,q-k}(t)\ . \tag{2.10}$$

The quantity $\Psi^k(t,t')$ in (2.9) is simply the Fourier transform of the probability propagator[1] evaluated in I:

$$\Psi^k(t,t')=\sum_m e^{ikm}J_m^2([u^2(t,t')+v^2(t,t')]^{1/2})$$
$$=J_0(2[u^2(t,t')+v^2(t,t')]^{1/2}\sin(k/2))\ , \tag{2.11}$$

where J_m is the ordinary Bessel function of the first kind of order m, and

$$u(t,t')=\int_{t'}^t dt''\cos[\mathcal{E}\eta(t'')]\ ,$$
$$v(t,t')=\int_{t'}^t dt''\sin[\mathcal{E}\eta(t'')]\ . \tag{2.12}$$

Like (2.3), Eq. (2.9) is also no more than a "formal" solution since $P^k(t)$ appears on both sides of Eq. (2.9). The Kenkre-Brown analogue of (2.9) contains a $\Psi^k(t,t')$ which is of the difference form $\Psi^k(t-t')$ and, consequently, can be solved explicitly[4] via Laplace transforms. Here, however, the time dependence of the applied field makes the Hamiltonian nonstationary and destroys the difference nature of the propagator. One can always iterate (2.9) to obtain $P^k(t)$ to arbitrary accuracy,

$$P^k(t)=e^{-\alpha t}\left[\theta^k(t)+\alpha\int_0^t dt'\Psi^k(t,t')\theta^k(t')+\alpha^2\int_0^t dt'\int_0^{t'} dt''\,\Psi^k(t,t')\Psi^k(t',t'')\theta^k(t'')+\cdots\right] \quad (2.13)$$

and then substitute it in (2.3) to obtain the solution for all elements of the density matrix. Much can be learned, however, from the mean-square displacement $\langle m^2\rangle$, which often has more practical usefulness than the probabilities. The mean-square displacement can be extracted *exactly* from (2.9) as follows.

The definition of the mean-square displacement,

$$\langle m^2\rangle(t)=-\frac{\partial^2}{\partial k^2}P^k(t)\bigg|_{k=0}, \quad (2.14)$$

used in conjunction with (2.9) gives

$$\langle m^2\rangle(t)=e^{-\alpha t}\theta''(t)+\alpha\int_0^t dt' e^{-\alpha(t-t')}\langle m^2\rangle(t') +\alpha\int_0^t dt' e^{-\alpha(t-t')}\Psi''(t,t')\ , \quad (2.15)$$

where θ'' and Ψ'' are the second derivatives of θ and Ψ, respectively, with respect to k, evaluated at $k=0$:

$$\theta''(t)=-\frac{\partial^2}{\partial k^2}\theta^k(t)\bigg|_{k=0}, \qquad \Psi''(t,t')=-\frac{\partial^2}{\partial k^2}\Psi^k(t,t')\bigg|_{k=0}. \quad (2.16)$$

A differentiation with respect to t converts (2.15) into

$$\frac{d}{dt}\langle m^2\rangle=e^{-\alpha t}\frac{d}{dt}\theta''(t)+\alpha\int_0^t dt' e^{-\alpha(t-t')}\frac{d}{dt}\Psi''(t,t')\ . \quad (2.17)$$

The substitution

$$\Psi''(t,t')=2V^2[u^2(t,t')+v^2(t,t')] \quad (2.18)$$

and the trigonometric properties of $u(t,t')$ and $v(t,t')$ allow us to obtain, from a reduction of (2.17), the general result

$$\frac{d}{dt}\langle m^2\rangle(t)=e^{-\alpha t}\frac{d}{dt}[\theta''(t)-\Psi''(t,0)] +4V^2\int_0^t dt' e^{-\alpha(t-t')}\cos\{\mathcal{E}[\eta(t)-\eta(t')]\}\ . \quad (2.19)$$

Finally, if the initial density matrix is site diagonal, $\theta''(t)=\Psi''(t,0)$, and (2.19) becomes

$$\langle m^2\rangle(t)=\langle m^2\rangle(0) +4V^2\int_0^t dt'\int_0^{t'} dt'' e^{-\alpha(t'-t'')} \times\cos\{\mathcal{E}[\eta(t')-\eta(t'')]\}\ , \quad (2.20)$$

which is one of the central results of this paper.

III. EVALUATION OF THE DIFFUSION CONSTANT

The general result (2.20) is valid for arbitrary time dependence of the electric field. For a sinusoidal field, $f(t)=\cos(\omega t)$, $\eta(t)=\sin(\omega t)/\omega$, and (2.20) reduces to

$$\langle m^2\rangle(t)=\langle m^2\rangle(0)+4V^2\int_0^t dt'\int_0^{t'} dt'' e^{-\alpha(t'-t'')}\cos\{(\mathcal{E}/\omega)[\sin(\omega t')-\sin(\omega t'')]\}\ . \quad (3.1)$$

Equation (3.1) reduces, in the limit of no field ($\mathcal{E}\to 0$), to the well-known result

$$\langle m^2\rangle(t)-\langle m^2\rangle(0)=(4V^2/\alpha)t=2(D_0/a^2)t\ , \quad (3.2)$$

where D_0 is the field-free diffusion constant $2V^2a^2/\alpha$, a being the lattice constant, as stated in the Introduction. In order to find a general expression for the diffusion constant D in the presence of the field, we expand the cosine function in the integrand of (3.1) as a double sum of Bessel functions. The result is

$$\frac{d}{dt}\langle m^2\rangle=4V^2\int_0^t d\tau\, e^{-\alpha\tau}\sum_{m,n}J_m(\mathcal{E}/\omega)J_n(\mathcal{E}/\omega)\{\cos[(n-m)\omega t]\cos(m\omega\tau)-\sin[(n-m)\omega t]\sin(m\omega\tau)\}\ , \quad (3.3)$$

where we have changed the integration variable t' to $\tau=t-t'$. On performing the indicated integrations, and on considering the limit that $\alpha t\gg 1$, we get

$$\left[\frac{d}{dt}\langle m^2\rangle\right]_{\alpha t\gg 1}=4V^2\sum_{m,n}J_m(\mathcal{E}/\omega)J_n(\mathcal{E}/\omega)\{\alpha\cos[(n-m)\omega t]-m\omega\sin[(n-m)\omega t]\}/[\alpha^2+(m\omega)^2]\ . \quad (3.4)$$

We define the diffusion constant D in the presence of the field as

$$\lim_{t\to\infty}[\langle m^2\rangle(t)-\langle m^2\rangle(0)](\alpha^2/2t) .$$

Equation (3.4) then shows that the relation between D and D_0, i.e., between the values of the diffusion constant in the presence and absence of the field, respectively, is

$$D=D_0\sum_m J_m^2(\mathcal{E}/\omega)\{\alpha^2/[\alpha^2+(m\omega)^2]\} . \tag{3.5}$$

Equation (3.5) is valid for arbitrary relative magnitudes of V, α, $\mathcal{E}$, and ω. Two extreme limits are of special interest: the static limit ($\omega \ll \alpha$) and the dynamic limit ($\omega \gg \alpha$). In the static limit, Eq. (3.5) reduces to a particularly simple form. The factor $\alpha^2/(\alpha^2+m^2\omega^2)$ on the right-hand side of (3.5) may be written as the integral over time (from 0 to ∞) of $\alpha e^{-\alpha t}\cos(m\omega t)$. The sum over m of $\cos(m\omega t)J_m^2(\mathcal{E}/\omega)$ may be expressed exactly as a single Bessel function $J_0[2(\mathcal{E}/\omega)\sin(\omega t/2)]$. An alternate way of writing (3.5) is therefore

$$D=D_0\int_0^\infty dt\,\alpha e^{-\alpha t}J_0[\mathcal{E}ty(t)] , \tag{3.6}$$

where $y(t)=\sin(\omega t/2)/(\omega t/2)$. The static limit allows us to write $y(t)=1$ in (3.6) and evaluate the right-hand side of (3.6) as a Laplace transform. The diffusion constant in the static limit is therefore given by

$$D=D_0[1+(\mathcal{E}/\alpha)^2]^{-1/2} \tag{3.7}$$

The diffusion constant is thus reduced by the electric field.

In the dynamic limit ($\omega \gg \alpha$), only the $m=0$ term in the summation need be retained on the right-hand side of (3.5), and the diffusion constant is given by

$$D=D_0J_0^2(\mathcal{E}/\omega) . \tag{3.8}$$

A reduction in D caused by the field occurs also in this dynamic limit. One also observes the dramatic feature peculiar to this limit that the diffusion constant vanishes whenever $\mathcal{E}/\omega$ is a root of J_0. The particle is then confined to a limited region of the lattice. This is the phenomenon of dynamic localization whose investigation we began in I. In the general case wherein neither limit is applicable, there will be contributions from the other terms in the sum in (3.5). Since it is not possible to select a value of $\mathcal{E}/\omega$ which will simultaneously be a root of *all* the Bessel functions in the sum, it is clear that dynamic localization in its strictest sense cannot occur in the general case, i.e., the diffusion constant will not vanish. Nevertheless, when the first term dominates the series, the motion will be substantially reduced whenever $\mathcal{E}/\omega$ is a root of J_0. We plot the average diffusion constant as a function of $\mathcal{E}/\omega$ for different values of α/ω in Fig. 1. It is clear from Fig. 1 that increasing the size of α with respect to ω makes dynamic localization less apparent.

It is also of interest to examine the effect of the applied field as manifested in the modification of the carrier velocity and the collision time. In the absence of the field, the velocity of the particle is proportional to V, the scattering rate (the reciprocal of the collision time) is proportional to α, and the diffusion constant to V^2/α. In the presence of the field of frequency ω, one can say that, if the field changes sign many times during collision-free motion of the particle, i.e., if $\omega \gg \alpha$, the velocity is reduced by the factor $J_0(\mathcal{E}/\omega)$. In other words, $V_{\text{eff}}=VJ_0(\mathcal{E}/\omega)$. The velocity vanishes for certain values of the frequency, causing the onset of dynamic localization. If, on the other hand, the particle suffers many collisions within the period of the field, i.e., if $\omega \ll \alpha$, the collision time is decreased by the factor $[1+(\mathcal{E}/\alpha)^2]^{-1/2}$.

An alternate way of stating the effect of the field for *all* frequencies is to say that what it affects is not the velocity of the particle, but *always* the collision time (equivalently, the scattering rate). The general expression for the effective scattering rate for arbitrary relative magnitude of ω and α, is

$$\alpha_{\text{eff}}=\alpha\Big/\left[\sum_m[1+(m\omega/\alpha)^2]^{-1}J_m^2(\mathcal{E}/\omega)\right] . \tag{3.9}$$

The limiting cases discussed above are recovered immediately from (3.9). In the static limit ($\omega \ll \alpha$), Eq. (3.9) reduces to

$$\alpha_{\text{eff}}=\alpha[1+(\mathcal{E}/\alpha)^2]^{1/2} , \tag{3.10}$$

while, in the dynamic limit ($\omega \gg \alpha$) it yields

$$\alpha_{\text{eff}}=\alpha/J_0^2(\mathcal{E}/\omega) . \tag{3.11}$$

Figures 2(a) and 2(b) show this effect pictorially. The peaks in α_{eff} represent dynamic localization.

In deriving (3.5), we integrated (3.4) to a time t and took the limit of the quotient of the resulting expression and t as $t\to\infty$. Because the average value of the oscillating terms in (3.4) is zero, only the constant terms contributed in expression (3.5). It is important to note that this derivation is incorrect in the case that the frequency ω is identically zero. For, in this case, there will be no oscillating terms in (3.4). In order to find the mean-square

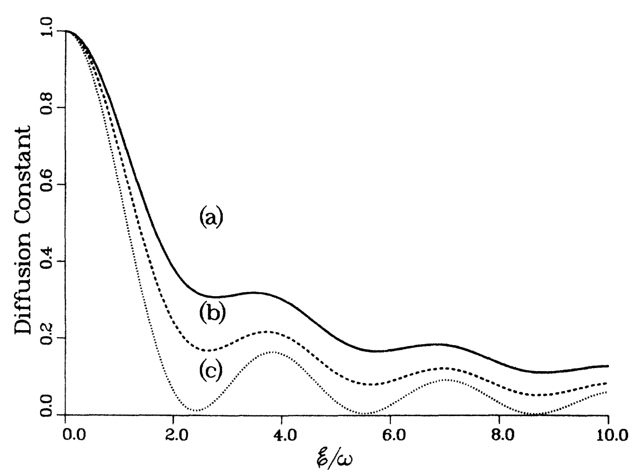

FIG. 1. Dynamic localization as manifested in the diffusion constant. The diffusion constant D normalized to its value D_0 in the absence of the field, from (3.5), shown as a function of the field-frequency ratio $\mathcal{E}/\omega$ for different values of the ratio of the scattering ratio to the field frequency α/ω: (a) $\alpha/\omega=2$, (b) $\alpha/\omega=1$, and (c) $\alpha/\omega=0.2$. The oscillations of the diffusion constant which arise from the phenomenon of dynamic localization are visible for small scattering, but are washed out for large scattering.

displacement in this case, it is easiest to set $\omega=0$ in expression (3.1). We have, from (3.1),

$$\langle m^2\rangle(t)=\langle m^2\rangle(0)+4V^2\int_0^t dt'\int_0^{t'} dt'' e^{-\alpha(t'-t'')} \times\cos\mathcal{E}(t'-t'') \ . \tag{3.12}$$

At long times, such that $\alpha t \gg 1$, the integration of the expression in (3.12) is given by

$$\langle m^2\rangle-\langle m^2\rangle(0)=4V^2[\alpha/(\alpha^2+\mathcal{E}^2)]t \ . \tag{3.13}$$

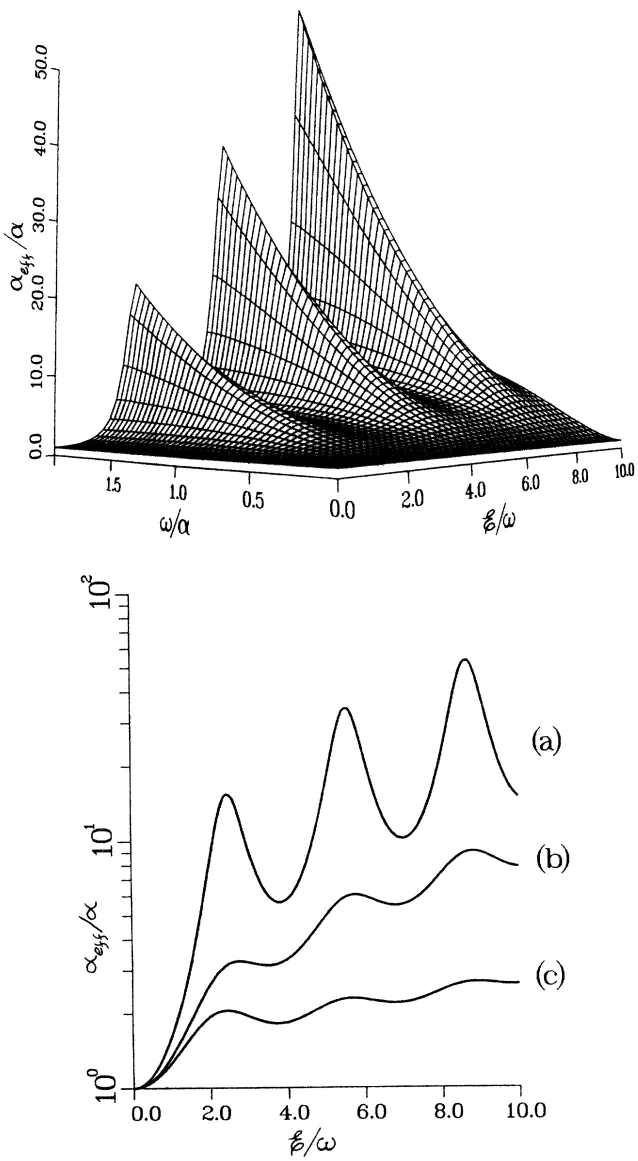

FIG. 2. Dynamic localization as manifested in the effective scattering rate. The effective scattering rate, α_{eff}, is plotted as a function of the dimensionless ratios α/ω and $\mathcal{E}/\omega$. When α/ω is small (small scattering), and $\mathcal{E}/\omega$ is near a root of J_0, the effective scattering rate is strongly peaked, showing dynamic localization. The full plot is shown in (a). Three cross sections of the α surface are shown in (b) for three values of α/ω: (a) 0.5, (b) 2, and (c) 10.

On dividing the right-hand side of (3.13) by $2t$ and taking the limit as $t\rightarrow\infty$, we find that the diffusion constant in the dc case is given, not by (3.7), but by

$$D=D_0[\alpha^2/(\alpha^2+\mathcal{E}^2)] \ . \tag{3.14}$$

This also means that the effective scattering rate in the dc case is given by

$$\alpha_{\text{eff}}=\alpha(1+\mathcal{E}^2/\alpha^2) \tag{3.15}$$

rather than by (3.10).

Equations (3.15) and (3.10) are both obtained in the static limit. For large scattering ($\alpha\gg\mathcal{E}$), they give essentially identical results (except for a proportionality constant). However, for large fields ($\mathcal{E}\gg\alpha$), they are quite different. Which of the two results is applicable in a given measurement is really a question of measurement time. This is clearly demonstrated in Fig. 3, where we plot the mean-square displacement in the static regime for an ac field. We note that, if the scattering rate is small compared to the magnitude of the electric field, the graph of the mean-square displacement as a function of time looks like a staircase, with alternating risers and treads. The mean-square displacement increases quickly when the ac field is zero, and all the lattice sites are energetically equivalent. This is indicated by the risers in the staircase. On the other hand, the mean-square displacement increases slowly when the ac field is nonzero. This forms the treads. The time average of the diffusion constant is given by the slope of the staircase. If the measurement time is long compared to the period of the ac

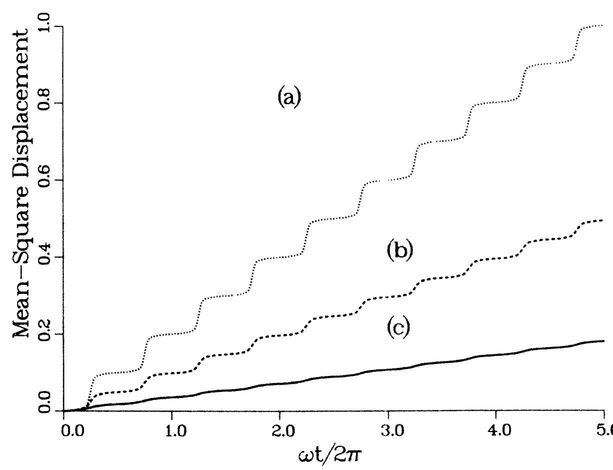

FIG. 3. The mean-square displacement in the static regime for an ac field. The mean-square displacement plotted as a function of reduced time $\omega t/2\pi$ for different values of $\alpha/\mathcal{E}$: (a) $\alpha/\mathcal{E}=0.1$, (b) $\alpha/\mathcal{E}=0.2$, and (c) $\alpha/\mathcal{E}=0.5$. We take $\alpha/\omega=10$ in all cases. The onset of a "staircase" when $\alpha/\mathcal{E}$ is small indicates that motion occurs primarily when the oscillating field passes through its zeros. The measured diffusion constant will be given by (3.14), i.e., the slope of a single "tread" of the staircase if the measurement time is small compared to the period of the ac field. On the other hand, if the measurement time is large compared to the period of the ac field, the measured diffusion constant will be the average over both "risers" and "treads," and will be given by (3.7).

field, the measurement probes the average of the slope of the risers and the treads, and (3.7) and (3.10) apply. However, if the process of measurement is characterized by a very short time, and addresses only the treads in the staircase, (3.14) and (3.15) apply. The crucial quantity is thus the ratio of the measurement time to the time over which a single tread occurs (which equals half the period of the field).

IV. FINITE-TEMPERATURE CALCULATIONS

A well-known[2,4,9] drawback of the stochastic Liouville equation in its simple form (1.4) is that its treatment of the scattering is appropriate only to an infinite-temperature system. This drawback arises from the fact that a single scattering rate α appears in the equation. As we shall see below, a consequence of this feature of the SLE is that the current vanishes at long times no matter what the magnitude of the electric field. This is clearly undesirable. In order to address the current, it is therefore necessary to modify the SLE and remove this infinite-temperature characteristic. This can be done in a straightforward fashion. If we set $k=q$ in (2.1), we get the Boltzmann equation for $F_k(t)$, the diagonal part of $\rho^{k,q}(t)$:

$$\frac{\partial F_k(t)}{\partial t}+\mathcal{E}f(t)\frac{\partial F_k(t)}{\partial k}=-\alpha[F_k(t)-1/N] , \quad (4.1)$$

where N is the number of sites in the lattice. The term on the right-hand side of (4.1) results in an approach to equilibrium (in the absence of the field) in the relaxation-time approximation, but the equilibrium distribution is $F_k(\infty)=1/N$, which is the case when the temperature is infinite. In order to include the effect of finite temperature, we replace $1/N$ by the appropriate Boltzmann distribution, i.e, by $e^{-\beta\varepsilon(k)}/\sum_q e^{-\beta\varepsilon(q)}$, which is the correct value of $F_k(\infty)$. Here $\varepsilon(k)$ is the energy of the carrier in the momentum state $|k\rangle$. The diagonal part (in k space) of the SLE, thus modified, is, therefore

$$\frac{\partial F_k(t)}{\partial t}+\mathcal{E}f(t)\frac{\partial F_k(t)}{\partial k}=-\alpha[F_k(t)-F_k(\infty)] . \quad (4.2)$$

The solution of (4.2) is

$$F_k(t)=F_{k-\mathcal{E}\eta(t)}(0)e^{-\alpha t} +\alpha\int_0^t dt' e^{-\alpha(t-t')}F_{k-\mathcal{E}\eta(t)+\mathcal{E}\eta(t')}(\infty) , \quad (4.3)$$

where by $F_{k-\mathcal{E}\eta(t)}$ we mean the quantity obtained by replacing k by $k+\mathcal{E}\eta(t)$ in F_k. Since the particle velocity, v_k, is an odd function of k, it is apparent from (4.3) that the *unmodified* SLE results in a vanishing current at long times. The integration over k of the product of v_k and $F_{k-\mathcal{E}\eta(t)+\mathcal{E}\eta(t')}(\infty)$, which is $1/N$ for the unmodified SLE, is zero, and that part of the current due to the initial distribution decays to zero exponentially in time. If $F_k(0)=F_k(\infty)$, (4.3) can be written, upon integrating by parts, in the form

$$F_k(t)=F_k(\infty)+\beta\int_0^t dt'\mathcal{E}(t')e^{-\alpha(t-t')} \times F_{k-\mathcal{E}\eta(t)+\mathcal{E}\eta(t')}(\infty) \times\frac{d}{dk}\{\varepsilon[k-\mathcal{E}\eta(t)+\mathcal{E}\eta(t')]\} . \quad (4.4)$$

The current, $j(t)$, is equal to $e\sum_k v_k F_k(t)$. The particle velocity v_k is proportional to the k derivative of $\varepsilon(k)$, and $\sum_k v_k F_k(\infty)=0$. Making these substitutions in (4.4), we obtain the general expression for the current $j(t)$:

$$j(t)=e\beta\int dt'\mathcal{E}(t')e^{-\alpha(t-t')} \times\sum_k F_k(\infty)v_k v_{k-\mathcal{E}\eta(t)+\mathcal{E}\eta(t')} . \quad (4.5)$$

Expression (4.5) is an exact consequence of (4.2), and is valid for arbitrary $\varepsilon(k)$. For the system under consideration in the present paper, it takes on a simpler form. Here, the band energy $\varepsilon(k)$ equals $2V\cos(k)$.[10] Thus,

$$F_k(\infty)=\exp[\beta 2V\cos(k)]/I_0(2V\beta) , \quad (4.6)$$

where I_0 is the modified Bessel function of order zero. The matrix elements v_k of the particle velocity are given by $2Va\sin(k)$. Substituting $\sin(\omega t)$ for $f(t)$, and $\tau=t-t'$, we find the expression for the current,

$$j(t)=2Vea[I_1(2V\beta)/I_0(2V\beta)]\alpha \times\int_0^t d\tau\, e^{-\alpha\tau}\sin(\mathcal{E}/\omega\{\sin[\omega(t)]-\sin[\omega(t-\tau)]\}) . \quad (4.7)$$

The long-time limit of the current may be obtained by expanding (4.7) as an infinite sum of Bessel functions, and taking the limit $\alpha t\to\infty$. We then have

$$\lim_{\alpha t\to\infty} j(t)=2Vea[I_1(2V\beta)/I_0(2V\beta)]\alpha\sum_{m,n}J_n(\mathcal{E}/\omega)J_m(\mathcal{E}/\omega)/[(n\omega)^2+\alpha^2]\{n\omega\cos[(n-m)\omega t]+\alpha\sin[(n-m)\omega t]\} . \quad (4.8)$$

Equation (4.8) shows that the response to the driving ac field, $\mathcal{E}\cos(\omega t)$, has many higher Fourier components in general. It is interesting to examine this result in the dynamic limit, $\alpha\ll\omega$. In this limit, only the term with $n=0$ is large in the sum in (4.8). One thus has

$$\lim_{\alpha/\omega\to 0} j(t)=2Vea[I_1(2V\beta)/I_0(2V\beta)]J_0(\mathcal{E}/\omega)\sin[(\mathcal{E}/\omega)\sin(\omega t)] =2Vea[I_1(2V\beta)/I_0(2V\beta)]J_0(\mathcal{E}/\omega)\sum_m J_m(\mathcal{E}/\omega)\sin(m\omega t) . \quad (4.9)$$

We observe two important aspects of the current in the dynamic limit from (4.9). The first is that the $m=0$ component of the current is 90° out of phase with the field, i.e., the conductivity is purely imaginary. The second is that, when $\mathcal{E}/\omega$ is a root of J_0, i.e., dynamic localization occurs, the ac current is identically zero.

It is important to notice that the general current expression (4.5) we have obtained above contains a natural and practical generalization of the Kubo linear-response formula.[11] In order to clarify this statement, we rewrite (4.5) in the Kubo form

$$j(t)=\int_0^t dt'\phi(t,t')\mathcal{E}(t') \tag{4.10}$$

and note that the response function $\phi(t,t')$ is given by our analysis as

$$\phi(t,t')=e\beta e^{-\alpha(t-t')}\sum_k F_k(\infty)v_k v_{k-\mathcal{E}[\eta(t)-\eta(t')]} . \tag{4.11}$$

In the limit of small field ($\mathcal{E}\rightarrow 0$), (4.11) reduces to

$$\phi(t,t')=\phi(t-t')=e\beta e^{-\alpha(t-t')}\sum_k F_k(\infty)v_k v_k , \tag{4.12}$$

which is the standard Kubo result for our system. The stationary nature of the Kubo response function, equivalently, the fact that ϕ depends on t and t' only through their difference, arises from the fact that the Kubo formalism involves a *linear* approximation in the applied field. Our response function (4.11), on the other hand, contains the applied field to all orders. This nonlinearity is reflected in the nondifference nature of our response function.

V. DISCUSSION

The starting point for the analysis in this paper is the stochastic Liouville equation (SLE) (1.4), and its diagonal form modified in order to include finite-temperature effects, (4.2). The main results of this analysis are (2.20) for the mean-square displacements, (3.5) for the diffusion constant, (3.9) for the effective scattering rate, and (4.6) for the current. They are exact consequences of the SLE. The first three arise from (1.4) and the fourth from (4.2). The results complete our investigation of the phenomenon of dynamic localization begun in I by extending the theory in I to include the effects of scattering of the charge carrier by such agents as imperfections and vibrations of the lattice.

From a mathematical point of view, one of the contributions of the present paper is the exact solution (2.20) for the mean-square displacement in the presence of scattering, which we have been able to obtain despite the fact that the probabilities of site occupation cannot be written down explicitly [see Eq. (2.13)]. This is different from the case for no scattering analyzed in I where we obtained explicit results for probabilities as well as for the mean-square displacement. From the physical point of view, a number of new results have emerged, some of which were expected. One of such is the quantitative justification of the statement we made in I that the observation of dynamic localization would require that the scattering rate α be small with respect to the field frequency ω. This is clear from Eq. (3.5).

The introduction of the scattering rate α separates the parameter space into two limiting regimes: the static limit and the dynamic limit. These show quite different behaviors. In the static limit, i.e., for $\alpha\gg\omega$, the effect of the ac electric field is to cause the diffusion constant to decrease monotonically with increasing field intensity. However, in the dynamic limit, i.e., for $\alpha\ll\omega$, an increase in the field causes the diffusion constant to decrease in a nonmonotonic, indeed in an oscillatory, fashion. Drastic reduction in the diffusion constant occurs whenever the quantity $eEa/\hbar\omega$ approaches a root of the Bessel function of order zero. These oscillations, which constitute the phenomenon of dynamic localization, are shown in Figs. 1 and 2.

In Table I below we compare the results obtained in Ref. 1 for the mean-square displacement $\langle m^2\rangle$ in the absence of scattering to the corresponding ones obtained here in the presence of scattering. We have retained in the table only long-time terms, and have neglected oscillatory ones except in the one case in which an oscillatory term is the only one present. Entry (I1) in the table shows that scattering impedes motion,[2] converting, as it does, the time dependence of $\langle m^2\rangle$ from t^2 to t. The characteristic transport quantity changes from a velocity $2^{1/2}Va$ in the absence of scattering to a diffusion constant $2V^2a^2/\alpha$ in its presence. Entry (I2) shows, however, the intriguing result that, in the presence of a dc electric field, scattering *assists motion,* as it converts a bounded, oscillatory $\langle m^2\rangle$ into an unbounded quantity which grows linearly with t. Physically, the scattering removes the energy mismatch created by the dc field. Entries (I3) and (I4) show that, as in the field-free case, the motion is generally impeded by scattering: the dependence of $\langle m^2\rangle$ is converted from t^2 to t by scattering. However, the phenomenon of dynamic localization is exhibited in (I3) whenever the scattering rate is small with respect to the field frequency. The characteristic transport quantity

TABLE I. Mean-square displacement of an initially localized particle.

Electric field $\mathcal{E}\cos(\omega t)$	Long-time $\langle m^2\rangle$ in the presence of scattering ($\alpha\neq 0$)	Long-time $\langle m^2\rangle$ in the absence of scattering ($\alpha=0$)	
$\mathcal{E}=0$, no field	$(2V^2/\alpha)t$	$2V^2t^2$	(I1)
$\omega=0$, dc field	$2V^2[\alpha/(\alpha^2+\mathcal{E}^2)]t$	$(8V^2/\mathcal{E}^2)\sin^2(\mathcal{E}t/2)$	(I2)
$\omega\neq 0$, $\alpha\ll\omega$	$(2V^2/\alpha)J_0^2(\mathcal{E}/\omega)t$	$2V^2J_0^2(\mathcal{E}/\omega)t^2$	(I3)
$\omega\neq 0$, $\alpha\gg\omega$	$2V^2/(\alpha^2+\mathcal{E}^2)^{1/2}t$		(I4)

(velocity in the absence of scattering, diffusion constant in its presence) oscillates as the field intensity is varied and is greatly reduced for certain values. The case when the scattering rate is large with respect to the field frequency is described by entry (I4). The difference between the diffusion constants as given by the entries (I2) and (I4) arises from differences in measurement times, has been discussed in Sec. III, and is depicted in Fig. 3.

The localization in the *dc case* seen in entry (I2) of the table is well understood and indeed is textbook material.[12,13] While it is by no means the focus of the present paper, we point out in passing that it may be described physically in at least three different ways. One of them is in terms of the eigenstates of the Hamiltonian, which are well known[14–22] to be localized Stark ladder states. The phase relations among the coefficients of the Wannier state expressed in terms of the Stark ladder states are known to be such that there is no tendency to evolve to a delocalized state. A second way of understanding the localization in a dc field is from a momentum-space perspective. The dc field causes Bloch oscillations[23–26] in the Brillouin zone and returns the system repeatedly to its original state, increasing and decreasing the velocities in the band periodically without change. A third way to approach the problem is in terms of the evolution of a two-state nondegenerate system with energy difference $\mathcal{E}$ and interaction matrix element V, initially occupying one of the states. The energy mismatch $\mathcal{E}$ causes localization in the initial state in the sense that the average probability in the initial state never fully reduces to zero. This effect can be thought of as occurring repeatedly in our system as the particle attempts to move to more and more distant sites. The energy mismatch occurs at every step and returns the particle to the initial site. For each of the three interpretations for the localization by a dc field, there is an interpretation of the role of the scattering rate in causing delocalization. Scattering introduces mixing among the Stark ladder states with coefficients representative of a state spread out in the crystal, makes the Bloch oscillations incomplete and leaves the k-distribution asymmetrical in the band and thus descriptive of spatial motion, and provides a nonsingular density of states for the transition between site states required in the sense of the Fermi golden rule.

The physics behind the *ac-field* effect analyzed in I and the present paper can be described[27] as arising from a synchronization of two processes: the natural time evolution of the particle in the Brillouin zone when driven by a field, and the time evolution of the field itself. When the field is sinusoidal in time and the band energy (consequently, the velocity in the k state) is also sinusoidal in k, it is possible for the period and the magnitude of the field to be such that the average of the velocity over a period of the field vanishes *no matter what the initial value of* k. This condition is precisely the dynamic localization condition. Arbitrary dispersion relations and arbitrary time dependencies of the field will not, in general, result in exact dynamic localization, but the overall effect of reduction in transport quantities as the ratio of the magnitude and frequency of the field approaches certain values will occur.

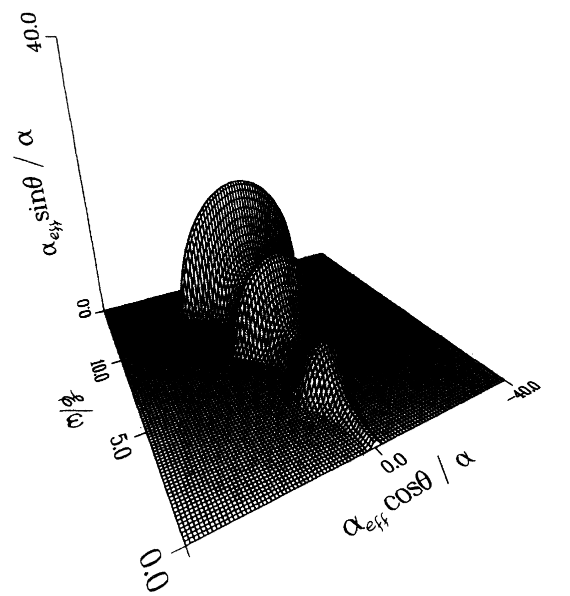

FIG. 4. Projection of the effective scattering in two dimensions. The projection of the scattering in a 2D solid as a function of the ratio $\mathcal{E}/\omega$. Each intersection of the plotted surface with a constant $\mathcal{E}/\omega$ plane constitutes a polar plot of the effective scattering $\alpha_{\text{eff}}(\theta)$, normalized to its value α in the absence of the field. The ratio $\alpha/\omega = 1$, and the electric field is directed at 45° to the x and y axes. Dynamic localization is manifested as bulges in the surface, representing an increase in the effective scattering rate.

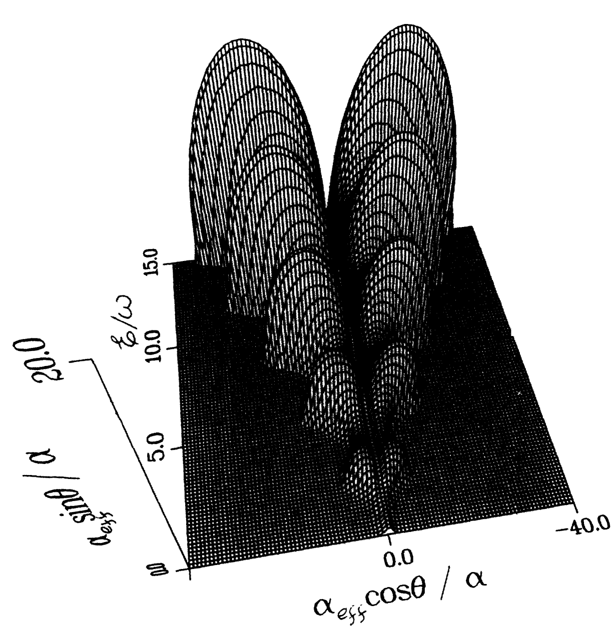

FIG. 5. Anisotropic effects arising from dynamic localization. A projection of the effective scattering rate in a two-dimensional solid, as in Fig. 4, except that the angle that the electric field makes with the x axis is $\theta = 0°$. Scattering will be field enhanced in the x direction, and will be independent of the field in the y direction. The result is a highly anisotropic α surface.

Many of the assumptions underlying our analysis have been commented on in I. We will not repeat that discussion here. We add only that the SLE we have used here to describe the effects of scattering possesses both the advantage of tractability, which has encouraged many workers to employ it,[2–7] and the disadvantage that it is actually an infinite-temperature transport instrument[2,4,9] which must be modified, for instance, in the manner we have used in Sec. IV, before it can address finite temperatures.

In order to investigate in I the effect of dynamic localization in higher dimensions, we extended the model to include motion on a three-dimensional (simple-cubic) lattice. We considered the case that the nearest-neighbor interactions were orthogonal, so that the motion along each axis was independent of the motion along the other two. We conclude the present paper with the same extension to higher dimensions, but in the presence of scattering. In I, we found that the mean-square displacement along a particular axis r ($r=x$, y, or z) in the simple-cubic model obeys the same equation as in one dimension, but with the electric field $\mathcal{E}$ replaced by the projection of the electric field along the r axis, $\mathcal{E}_r$, and with the appropriate transfer matrix element along the r axis, V_r. In the present case, we need only add to this the scattering rate for motion along the r axis, α_r. Thus, in analogy with I, the average diffusion constant along the r axis, D_r, in the presence of a sinusoidal ac field, is given by

$$D_r = 2V_r^2 a_r^2/\alpha_{r,\mathrm{eff}} , \tag{5.1}$$

where a_r is the lattice constant in the r direction, and where the effective scattering rate along the r axis, $\alpha_{r,\mathrm{eff}}$, is given by

$$\alpha_{r,\mathrm{eff}} = \alpha_r \Big/ \left[\sum [1+(m\omega/\alpha_r)^2]^{-1} J_m^2(\mathcal{E}_r/\omega) \right] . \tag{5.2}$$

In order to show the anisotropies that an ac field can impose on an initially isotropic material, we consider the two-dimensional case of the above, i.e., $V_z=0$. If we assume that $\alpha_x=\alpha_y=\alpha$, the projection of the effective scattering rate along a direction which makes an angle θ with the x axis is given by

$$\begin{aligned}\alpha_{\mathrm{eff}}(\theta) &= (\alpha_{x,\mathrm{eff}}^2\cos^2\theta+\alpha_{y,\mathrm{eff}}^2\sin^2\theta)^{1/2}\\ &= \alpha\left[\left[\sum J_m^2(\mathcal{E}_x/\omega)/[1+(m\omega/\alpha)^2] \right]^{-2}\cos^2\theta + \left[\sum J_m^2(\mathcal{E}_y/\omega)/[1+(m\omega/\alpha)^2] \right]^{-2}\sin^2\theta \right]^{1/2} .\end{aligned} \tag{5.3}$$

Figures 4 and 5 are polar plots of the ratio of $\alpha_{\mathrm{eff}}(\theta)/\alpha$ as a function of $\mathcal{E}/\omega$. The ratio α/ω has been set to unity, so neither the dynamic nor the static limit results are completely dominant. The magnitude of the effective scattering in a particular direction is indicated by the height of the surface above the origin in that direction. In Fig. 4 the electric field is directed at an angle of 45° to the x and y axes, and the surface is isotropic. We notice that, as the electric field is increased, the effective scattering becomes greater in general, which is in agreement with expression (3.10) calculated in the static limit. The oscillations peak whenever $\mathcal{E}/2^{1/2}\omega$ is a root of J_0, which is in agreement with properties of the dynamic limit, expressed in (3.11). In Fig. 5 the electric field is directed along the x axis. Hence, in the y direction the effective scattering is independent of the field, whereas it oscillates with the roots of J_0 in the x direction.

We hope that the analysis given in I and the present paper will prompt experimentalists to carry out observations of the dynamic localization effect. The importance of the effect would lie in the possibility of inducing transport anisotropy in otherwise isotropic substances by the application of time-varying fields and of generally tailoring dynamically the mobility properties of materials.

ACKNOWLEDGMENT

This work was supported in part by the National Science Foundation (NSF) under Grant No. DMR-85-06438.

[1] D. H. Dunlap and V. M. Kenkre, Phys. Rev. B **34**, 3625 (1986).
[2] See, e.g., V. M. Kenkre, in *Energy Transfer Processes in Condensed Matter,* edited by B. Di Bartolo (Plenum, New York, 1984), p. 205.
[3] H. Haken and G. Strobl, Z. Phys. **262**, 135 (1973).
[4] V. M. Kenkre and D. W. Brown, Phys. Rev. B **31**, 2479 (1985).
[5] T. S. Rahman, R. S. Knox, and V. M. Kenkre, Chem. Phys. **44**, 197 (1979).
[6] R. Silbey, Annu. Rev. Phys. Chem. **27**, 203 (1976).
[7] R. P. Hemenger, K. Lakatos-Lindenberg, and R. M. Pearlstein, J. Chem. Phys. **60**, 3271 (1974).
[8] This is a trivial generalization of a method pointed out to us by Charles Hart in connection with the amplitude equation (1.2).
[9] K. Lindenberg and B. West, Phys. Rev. Lett. **51**, 1370 (1983).
[10] In this paper we have taken k to be the reciprocal to m (rather than to ma), and therefore to be dimensionless.
[11] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
[12] P. W. Anderson, *Concepts in Solids* (Benjamin, New York, 1963).
[13] N. W. Ashcroft and N. D. Merman, *Solid State Physics* (Holt, Rinehart and Winston, New York, 1976), p. 244.
[14] M. Ferrari, V. Grecchi, and F. Zironi, J. Phys. C **18**, 5825 (1985).
[15] J. E. Avron, Ann. Phys. (N.Y.) **143**, 33 (1982).

[16]G. H. Wannier and J. P. Van Dyke, J. Math. Phys. **9**, 899 (1968).
[17]G. H. Wannier, Phys. Rev. **117**, 432 (1960).
[18]G. H. Wannier and D. R. Fredkin, Phys. Rev. **125**, 1910 (1962).
[19]E. N. Adams, Phys. Rev. **107**, 698 (1957).
[20]P. Roblin and M. W. Muller, J. Phys. C **16**, 4547 (1983).
[21]H. Fukuyama, Phys. Rev. B **8**, 5579 (1973).
[22]M. Luban and H. H. Luscombe, Phys. Rev. B **34**, 3674 (1986).
[23]F. Bloch, Z. Phys. **52**, 555 (1928).
[24]J. B. Krieger and G. J. Iafrate, Phys. Rev. B **33**, 5494 (1986).
[25]J. B. Krieger and G. J. Iafrate, Physica B + C **134B**, 228 (1985).
[26]M. Luban, J. Math. Phys. **26**, 2386 (1985).
[27]D. H. Dunlap and V. M. Kenkre, Phys. Lett. (to be published).

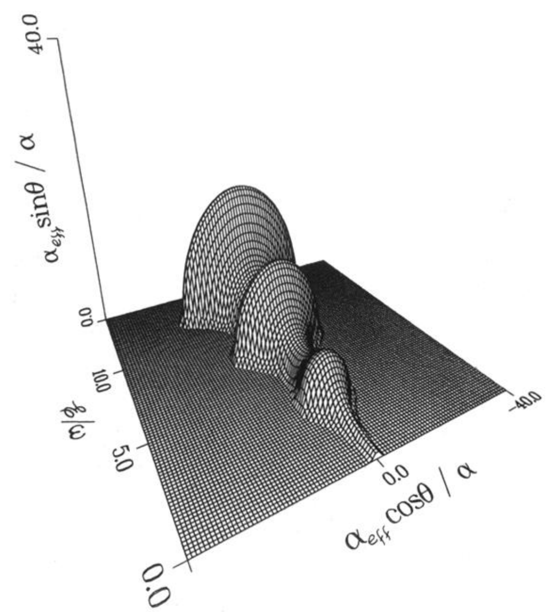

FIG. 4. Projection of the effective scattering in two dimensions. The projection of the scattering in a 2D solid as a function of the ratio $\mathcal{E}/\omega$. Each intersection of the plotted surface with a constant $\mathcal{E}/\omega$ plane constitutes a polar plot of the effective scattering $\alpha_{\text{eff}}(\theta)$, normalized to its value α in the absence of the field. The ratio $\alpha/\omega=1$, and the electric field is directed at 45° to the x and y axes. Dynamic localization is manifested as bulges in the surface, representing an increase in the effective scattering rate.

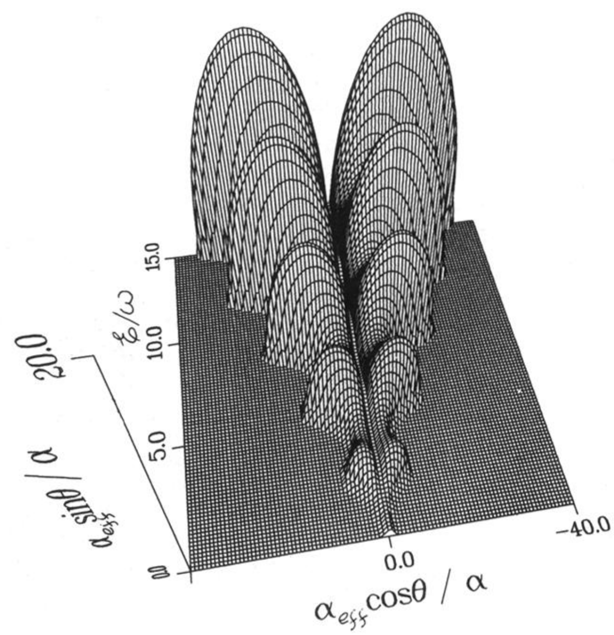

FIG. 5. Anisotropic effects arising from dynamic localization. A projection of the effective scattering rate in a two-dimensional solid, as in Fig. 4, except that the angle that the electric field makes with the x axis is $\theta=0°$. Scattering will be field enhanced in the x direction, and will be independent of the field in the y direction. The result is a highly anisotropic α surface.