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The spatial evolution of particles diffusing in the presence of randomly placed traps

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The evolution of a particle undergoing a continuous-time random walk in the presence of randomly placed imperfectly absorbing traps is studied. At long times, the spatial probability distribution becomes strongly localized in a sequence of trap-free regions. The subsequent intermittent transfer of the survival probability from small trap-free regions to larger trap-free regions is described as a time-directed variable range hopping among localized eigenstates in the Lifshitz tail. An asymptotic expression for the configurational average of the spatial distribution of surviving particles is obtained based on this description. The distribution is an exponential function of distance which expands superdiffusively, with the mean-square displacement increasing with time as $t^2/\ln^{(2D+4)/D}(t)$ in D dimensions.

I. INTRODUCTION

The effect of transport on the reactions of diffusing species has occupied a prominent position in the theory of chemical transport for most of this century.[1–18] Here we are concerned with the effect of randomly placed stationary traps upon a system of particles which undergo diffusion on a regular lattice. For nearly 20 years it has been known that the transport is anomalous in this circumstance,[2] and that the usual "reaction-diffusion" equation,

$$\frac{\partial}{\partial t}\langle p(r,t)\rangle=\mathscr{D}\nabla^2\langle p(r,t)\rangle-k\langle p(r,t)\rangle, \tag{1.1}$$

attributed to Smoluchowski[3] is inadequate to describe the configuration average of the distribution $p(r,t)$ at long times.[4] For example, it is now known that the configuration average of the survival probability, $P(t)=\langle\int dV\, p(r,t)\rangle$, decays anomalously, not exponentially as in the reaction-diffusion equation, but via the much slower "stretched exponential" decay given by $\exp(-t^{D/(D+2)})$, where D is the dimensionality of the system.[5–8] While this result has suggested to some that the spatial evolution of the distribution of the survival probability might also be anomalous, there has generally been less attention paid to such transport-related quantities. The situation has been exacerbated by the fact that the order in which the configuration averages are taken makes a considerable difference insofar as the spatial distribution is concerned. In one oft-cited paper,[6] for example, the configuration average of the mean-square displacement of surviving particles is given as $\overline{r^2}(t)\sim t^{2/(D+2)}$, which suggests that transport is subdiffusive. However, the quantity actually calculated in Ref. 6,

$$\left\langle\int dV\, p(r,t)r^2\right\rangle \Big/ \left\langle\int dV\, p(r,t)\right\rangle,$$

is the ratio of two configuration averages, a statistic, but not an observable. In contrast, we show in this paper that the observable,

$$\overline{r^2(t)}=\left\langle\int dV\, p(r,t)r^2 \Big/ \int dV\, p(r,t)\right\rangle\sim t^2/\ln^{(2D+4)/D}(t), \tag{1.2}$$

actually describes superdiffusive transport.

That the transport of the survival probability in the presence of randomly placed traps should be nearly ballistic is not obvious at first glance. After all, for the case of periodically placed traps it is generally understood that transport is simply diffusive: Asymptotically, the mean-square displacement of the surviving particles grows linearly in time. It is somewhat surprising, then, that simply introducing spatial disorder in the trap locations causes the mean-square displacement to instead grow superlinearly (in fact, almost quadratically) with time. Indeed, one might wonder how the ordered and the disordered systems are so markedly different as to have entirely different asymptotic time dependencies. One major difference between the two systems shows up clearly in the nature of the eigenfunctions: Those of the disordered system can be spatially localized rather than extended. In 1984, Ebeling, Engel, Esser, and Feistel[19] revealed how this difference plays a key role in understanding the anomalous transport which is observed in the general problem of diffusion in the presence of spatially random multiplicative noise. It was pointed out in Ref. 19 that the eigenstates of a diffusion equation describing transport in the presence of spatially random noise are the same as the localized states studied by Anderson[20] in the context of the Schrödinger equation describing a particle moving on a lattice with site-diagonal disorder. Because the diffusion equation is an imaginary-time Schrödinger equation, however, the time-dependent coefficients in any eigenfunction expansion are real exponentials, rather than oscillatory phase factors. As a result, the localized eigenfunctions at the low-frequency edge of the spectrum dominate any description of the evolution of the distribution $p(r,t)$ at long times, since the participation of high-frequency eigenstates decays away exponen-

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tially. It was shown in Ref. 19 that the normalized spatial distribution of surviving particles,

$$p_n(r,t)=p(r,t)\bigg/\int dV\,p(r,t), \tag{1.3}$$

tends to become localized for long periods of time in between occasional but relatively abrupt transitions during which the population appears to hop to a new location associated with a localized state having a smaller eigenvalue. This interesting intermittent hopping behavior was further described by Zel'dovich *et al.*[21] Subsequent analyses of the problem with Gaussian distributed growth and decay rates have shown[22,23] that the population $P(t)$ increases asymptotically as $\exp(t^3)$. What is remarkable in terms of transport, however, is the anomalously fast rate at which the width of the normalized spatial distribution grows as a result of this hopping from localized state to localized state. Extending the variable range hopping arguments developed by Mott,[24] it can be shown for the case of Gaussian noise that the characteristic width of the spatial distribution increases superdiffusively, scaling as $t/\ln^{1/2}(t)$ in one dimension.[25,26]

In the present paper we develop a description of hopping transport in the presence of randomly distributed partially absorbing traps in the spirit of that introduced in Ref. 19 for the problem of multiplicative noise. In such a case, the localized states of interest are those with long wavelengths centered about asymptotically large but rare trap-free domains with eigenvalues making up the so-called Lifshitz tail of the spectrum.[27] We describe the transport as time-directed variable range hopping among localized states in the Lifshitz tail. Furthermore, adapting a formulation of variable range hopping which was developed by Apsley and Hughes[28] we derive an analytic expression for the configuration average of the normalized spatial distribution function,

$$P_r(r,t)=\langle p_n(r,t)\rangle. \tag{1.4}$$

For periodically placed traps $P_r(r,t)$ is a Gaussian, and the underlying process is purely diffusive. We show here, in contrast, that for the disordered case the asymptotic distribution is an exponential function

$$P_r(r,t)\sim\frac{1}{\tau^D D V_D}\exp(-r/\tau), \tag{1.5}$$

of the radial distance from the origin, where in Eq. (1.5),

$$V_D=\frac{2\pi^{D/2}}{D\Gamma(D/2)} \tag{1.6}$$

is the volume of the unit sphere in D dimensions, Γ is the Gamma function, and

$$\tau\sim t/\ln^{(D+2)/D}(t) \tag{1.7}$$

is a logarithmically scaled time variable. It follows from Eqs. (1.7) and (1.5) that the transport of the survival probability is superdiffusive, with the nth moment

$$\overline{r^n}\sim\tau^n \tag{1.8}$$

proportional to the nth power of the rescaled time.

The rest of the paper is as follows. The next section lays down the theoretical foundations of our investigation by casting the problem in terms of an appropriate master equation. This approach, when coupled with an eigenfunction expansion, exposes the importance of localization for transport in the presence of random traps. Section III presents a calculation of the survival probability which exploits the variable-range hopping theory of Apsely and Hughes.[28] From this calculation emerges the spatial distribution function as expressed by Eqs. (1.5) and (1.7), and its moments as expressed by Eq. (1.8). Section IV presents a discussion of these results, and the relevant results of others. Section V concludes this work with a summary, and some speculations about possible applications.

II. THE MODEL

A chemical species diffusing among randomly placed traps may be described by a master equation[14]

$$\frac{d}{dt}P_m(t)=\sum_s F[P_{m+s}(t)-P_m(t)]-\gamma_m P_m. \tag{2.1}$$

In Eq. (2.1), the population at site m at time t is $P_m(t)$, and F is the rate for hopping between nearest-neighbor sites m and $m+s$ which are separated by a lattice constant a. The rates γ_m are taken from a bivalued distribution: if m labels a trap site, then the trapping rate $\gamma_m=\gamma$; otherwise $\gamma_m=0$.

It is instructive to express $P_m(t)$ in Eq. (2.1) as an eigenfunction expansion. We write $P_m(t)$ as the projection of the site state $|m\rangle$ onto the state of the system, $|\psi(t)\rangle$

$$P_m(t)=\langle m|\psi(t)\rangle. \tag{2.2}$$

The state vector can be expressed as a sum

$$|\psi(t)\rangle=\sum_j \langle\phi_j|0\rangle e^{-E_j t}|\phi_j\rangle \tag{2.3}$$

over the eigenstates $|\phi_j\rangle$ of the operator governing the disordered master equation,

$$\hat{H}=\sum_{m,s}\gamma_m|m\rangle\langle m|-F(|m\rangle\langle m+s|-|m\rangle\langle m|). \tag{2.4}$$

In Eq. (2.3) we have taken the localized initial condition $P_m(0)=\delta_{m,0}$. The eigenstates $|\phi_j\rangle$ and eigenvalues E_j obey the equation

$$\hat{H}|\phi_j\rangle=E_j|\phi_j\rangle, \tag{2.5}$$

Equations (2.2) and (2.3) may be combined to obtain

$$P_m(t)=\sum_j \langle m|\phi_j\rangle\langle\phi_j|0\rangle e^{-E_j t}. \tag{2.6}$$

All of the eigenvalues (or "frequencies") of the operator $\hat{H}$ are positive and lie in the range $4DF+\gamma\geqslant E_j>0$. Since the contribution to Eq. (2.6) from states with larger eigenvalues decays exponentially in time, it follows that eventually only the states in the neighborhood of $E=0$ are important.

It is understood that the states in the neighborhood of $E=0$ are exponentially localized in space. Indeed, the operator $\hat{H}$ is the same operator studied in the context of Anderson localization. With a binary distribution describing the diagonal elements of $\hat{H}$, that part of the spectrum near $E=0$ is often referred to as the Lifshitz tail. In what follows, we will describe these eigenfunctions approximately by ascribing to each the same localization length l. We refer to the average position of a localized state $|\phi_j\rangle$ as $\mathbf{r}_j$, and we refer to the average position of a state in the site basis as $\mathbf{r}_m$. The projection of a site state $|m\rangle$ onto a localized state $|\phi_j\rangle$ is therefore given by

$$\langle m|\phi_j\rangle \cong f_{m,j} e^{-\alpha|\mathbf{r}_m-\mathbf{r}_j|}, \tag{2.7}$$

where $\alpha=1/l$, and $f_{m,j}$ is a phase factor which is algebraic in $\mathbf{r}_j$ and $\mathbf{r}_m$. Substitution of Eq. (2.7) into Eq. (2.6) leads to

$$P_m(t) \cong \sum_j f_{j,0} f_{m,j} e^{-\alpha|\mathbf{r}_m-\mathbf{r}_j|} e^{-\alpha|\mathbf{r}_j|} e^{-E_j t}. \tag{2.8}$$

As we have already remarked, at long times only a diminishing fraction of low-frequency states in the Lifshitz tail are important for the description of $P_m(t)$. Such states are spatially few and far between. Because the participation of each localized state in the sum in Eq. (2.8) depends exponentially on both position and time, the terms in Eq. (2.8) are rather disparate. At long times it becomes increasingly likely that the contribution of a single state, $|\phi_i\rangle$, say, will exponentially dominate the others. In such a case,

$$P_m(t) \sim (f_{i,0} f_{m,i} e^{-\alpha|\mathbf{r}_m-\mathbf{r}_i|}) \times e^{-\mathfrak{R}_i}, \tag{2.9}$$

where the exponent $\mathfrak{R}_i$ is

$$\mathfrak{R}_i = \alpha|\mathbf{r}_i| + E_i t = \min_j[\alpha|\mathbf{r}_j| + E_j t]. \tag{2.10}$$

Here the index i labels that state whose contribution to the series in Eq. (2.8) dominates all others at time t. Summing Eq. (2.9) over all sites, we assume that the total probability remaining in the system at time t is well expressed by the contribution from this single dominating term, i.e.,

$$P(t) \sim q_i e^{-\mathfrak{R}_i}, \tag{2.11}$$

where the algebraic factor

$$q_i = \sum_m f_{i,0} f_{m,i} e^{-\alpha|\mathbf{r}_m-\mathbf{r}_i|} \tag{2.12}$$

is essentially the product of the area under the ith eigenfunction and the value of the ith eigenfunction at the origin. Equation (2.11) indicates how much has yet to decay from the system. But it also tells us where that which has not decayed may be found: Because the eigenstates are localized, we can associate the remaining probability in Eq. (2.11) with the location $\mathbf{r}_i$ of the eigenstate $|\phi_i\rangle$ which is dominating at time t, i.e., the state which has the smallest exponent $\mathfrak{R}$ at this time. Alternatively, the dominating state can be viewed as that state which is "closest" to the initial site in a space which consists of the normal spatial variables "augmented" by the "temporal distance" Et. Thus, what remains in the system is overwhelmingly concentrated in the state which is, in the sense described above, a "nearest-neighbor" to the origin at time t. As time increases, the temporal distance of each state increases at a rate proportional to its eigenvalue so that an exchange occurs in which the nearest-neighbor status is somewhat abruptly handed off to a state with a smaller

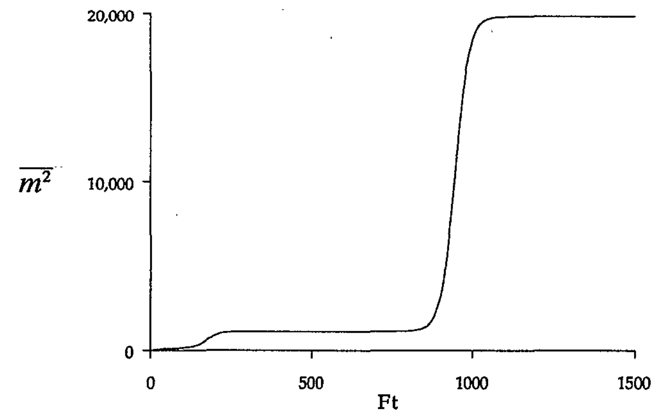

FIG. 1. For one dimension, the mean-square displacement, $\overline{m^2} = (\Sigma_m m^2 P_m(t))/(\Sigma_m P_m(t))$ of the distribution of surviving particles is shown as a function of reduced time Ft for a single configuration. The treads on the stair-step pattern arise because the distribution of that which remains tends to stay localized in large trap-free segments. The risers are indicative of hops, where the distribution of that which remains shifts abruptly to a larger trap-free segment. Hops occur twice for this configuration of traps: Once when $Ft \sim 160$, and again when $Ft \sim 880$. The concentration of traps was taken to be $C=0.5$, and the trapping rate $\gamma=0.5F$.

eigenvalue. Such an exchange occurs when two terms in Eq. (2.8) are of the same order of magnitude, i.e., when

$$e^{-\mathfrak{R}_i} \sim e^{-\mathfrak{R}_j}. \tag{2.13}$$

Equation (2.13) describes the state of affairs when the ith state, which has been dominating the sum, is about to be overshadowed by the jth state, which has a smaller eigenvalue. When the exchange occurs, the distribution of remaining probability appears to "hop" to a new, and more distant, spatial location. Between exchanges, however, the probability distribution pauses at the location of the dominant eigenstate for some time. Substituting Eq. (2.9) in Eq. (2.12), we can solve for the transition time

$$t_{i,j} = \frac{\alpha(|\mathbf{r}_j| - |\mathbf{r}_i|)}{E_i - E_j}, \tag{2.14}$$

where the location and eigenvalue of the state which will dominate next are $|\mathbf{r}_j|$ and E_j respectively. In order to determine which state will be the subsequent nearest-neighbor, we substitute for $|\mathbf{r}_j|$ and E_j in Eq. (2.14) the location $|\mathbf{r}|$ and eigenvalue E, respectively, of every state in the system with $E<E_j$, and then compute the corresponding transition time: the subsequent nearest-neighbor is the state for which $t_{i,j}$ is smallest.[19] Occasionally it happens that the eigenvalue of a nearby state which is not the closest to the origin in real space is, nevertheless, small enough that it becomes the subsequent-nearest-neighbor in the augmented space. In such a case, the remaining probability appears to "leap-frog" over other states in real space. The aficionado will recognize this as characteristic of what is often referred to as "variable range hopping." In Fig. 1 we show the results of a numerical integration of Eq. (2.1) for a single configuration. The mean-square displacement, shown as a function of time, resembles a stair case; the treads indicate times during which the probability is stationary, and the risers occur when the distribu-

tion abruptly "hops" to a state with a smaller eigenvalue. In Fig. 2 we show several snapshots of the normalized distribution of remaining probability and its correlation with the underlying distribution of traps. It is apparent that this hopping is associated with abrupt movement between large trap-free segments. That this is essentially equivalent to the description discussed above of hopping from state to state is clear, for the localized eigenfunctions near the band edge are in fact centered about such large trap-free segments.

III. SPATIAL DISTRIBUTION OF SURVIVING PARTICLES

In order to find the configurational average of the spatial distribution of surviving particles, it is necessary to calculate the distribution of nearest-neighbor distances $\mathfrak{R}$ in the augmented space referred to above, in which the states $|\varphi_j\rangle$ are associated with points (r_j,E_jt). Following the procedure of Apsely and Hughes[28] for variable-range hopping, we require the probability $P_{\mathfrak{R}}$ that the region in the augmented space which is bounded by the the r axis, the E axis, and the line $\mathfrak{R}=\alpha r+Et$ is devoid of localized eigenstates. Ignoring excluded-volume effects due to the sparsity of the states of interest, this can be written as an exponential function[28]

$$P_{\mathfrak{R}}=\exp\left[-\frac{V_D\eta_D}{(\alpha a)^D}\int_0^{\mathfrak{R}/t} dE\ \rho_D(E)(\mathfrak{R}-Et)^D\right], \tag{3.1}$$

in which η_D is that fraction of the total number of states described by the D-dimensional Lifshitz density $\rho_D(E)$, and a is the lattice spacing. The probability that the closest state is located at the distance $\mathfrak{R}$ in the hypershell of area $DV_Dr^{D-1}\ dr\ dE$ at time t is therefore given by

$$P(r,E;t)DV_Dr^{D-1}\ dr\ dE$$
$$=\frac{DV_Dr^{D-1}\ dr}{a^D}\ \eta_D\rho_D(E)dE\exp\left[-\frac{V_D\eta_Dt}{(\alpha a)^D}\right.$$
$$\left.\times\int_0^{\alpha r/t+E} dE'\ \rho_D(E')(\alpha r/t+E-E')^D\right]. \tag{3.2}$$

Thus the probability $P_E(E,t)dE$ that a randomly chosen member of the ensemble is dominated at time t by an eigenstate of frequency E is the integral

$$P_E(E,t)dE=\eta_D\rho_D(E)dE\int_0^\infty \frac{DV_Dr^{D-1}\ dr}{a^D}$$
$$\times\exp\left[-\frac{V_D\eta_Dt}{(\alpha a)^D}\int_0^{\alpha r/t+E} dE'\ \rho_D(E')\right.$$
$$\left.\times(\alpha r/t+E-E')^D\right], \tag{3.3}$$

of Eq. (3.2) over spatial variables. Similarly, the probability $P_r(r,t)DV_Dr^{D-1}\ dr$ that r is the spatial separation between the origin and the dominating state at time t is the integral

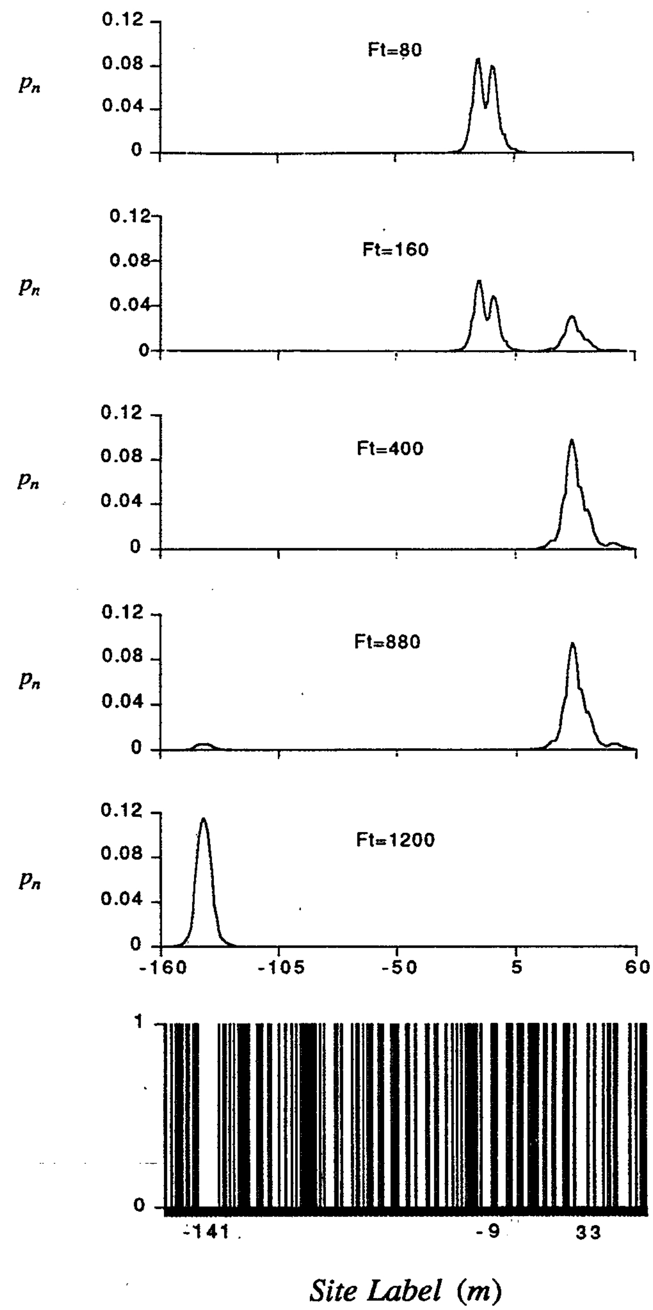

FIG. 2. For the same configuration of traps which was used in producing Fig. 1, we show here a time sequence of the normalized distribution p_n of surviving particles as a function of location. For $Ft=80$, the distribution is localized about two 4-site trap-free segments with an average position $\bar{m}=-9$: One segment is 5 sites to the left of the origin and the other segment is 12 sites to the left of the origin. For $Ft=160$, the distribution is in the process of hopping to a trap-free region located nearly 33 sites to the right of the origin. This region is dominated by a 15 site region containing only 3 traps. For $Ft=400$ the distribution is appears exponentially localized within this relatively trap-free domain. For $Ft=880$ the distribution has just begun the process of hopping to a 9-site trap-free segment located 141 sites to the left of the origin, and for $Ft=1200$ the distribution is again exponentially localized in a single region. This behavior is consistent with Fig. 1. Beneath these plots is a superimposed histogram showing the underlying trap configuration, with vertical lines indicating the presence of traps.

$$P_r(r,t)DV_Dr^{D-1}\,dr = \frac{DV_Dr^{D-1}\,dr}{a^D}\int_0^\infty \eta\rho_D(E)dE\,\exp\left[-\frac{V_D\eta t}{(\alpha a)^D}\times\int_0^{\alpha r/t+E} dE'\,\rho_D(E')(\alpha r/t+E-E')^D\right]. \tag{3.4}$$

of Eq. (3.2) over all E. By our previous arguments, Eq. (3.4) describes the configurational average of the spatial distribution of remaining particles. For further simplification, it is helpful to introduce the repeated integrals of the spectral density,

$$\rho_D^{(n)}(E)=\int_0^E dE_1\int_0^{E_1} dE_2\cdots\int_0^{E_{n-1}} dE_n\,\rho_D(E_n). \tag{3.5}$$

After integrating Eqs. (3.2) and (3.4) by parts and substituting the function $\rho_D^{(n)}(E)$ defined in Eq. (3.5), we obtain the results

$$P_r(r,t)DV_Dr^{D-1}\,dr = \frac{D\eta_DV_Dr^{D-1}\,dr}{a^D}\int_0^\infty \rho_D(E)dE\times\exp\left[-\frac{D!V_D\eta_Dt^D}{(\alpha a)^D}\,\rho_D^{(D+1)}(\alpha r/t+E)\right]. \tag{3.6}$$

and

$$P_E(E,t)dE=\rho_D(E)dE\int_0^\infty \frac{D\eta_DV_Dr^{D-1}\,dr}{a^D}\times\exp\left[-\frac{D!V_D\eta_Dt^D}{(\alpha a)^D}\,\rho_D^{(D+1)}(\alpha r/t+E)\right]. \tag{3.7}$$

Until now we have made no assumptions about the nature of the spectral density near the band edge. For the particular disordered system under consideration, composed of a random distribution of trapping centers of identical strength, it has been established that the states of interest are those associated with asymptotically large trap-free voids.[29] The lowest eigenstates of asymptotically large spherical voids of radius R and volume $V=V_DR^D$ can be associated with the long-wavelength solutions to the Helmholtz equation for a spherical cavity. Asymptotically,

$$E\sim\epsilon_0(V_D/V)^{2/D}, \tag{3.8}$$

where $\epsilon_0=Fx_0^2$ and x_0 is the first root of the radial Helmholtz equation in D dimensions. In one dimension, $x_0=\pi/2$, in two dimensions, $x_0=2.405$, the first root of the ordinary Bessel function $J_0(x)$, and in three dimensions $x_0=\pi$. When Eq. (3.8) is combined with the Poissonian distribution

$$P_V=\lambda e^{-\lambda V}, \tag{3.9}$$

of trap-free voids of volume V, the spectral density

$$\rho_D(E)\sim\left(\frac{\lambda DV_D}{2\epsilon_0}\right)\left(\frac{\epsilon_0}{E}\right)^{(D+2)/2}\exp[-\lambda V_D(\epsilon_0/E)^{D/2}] \tag{3.10}$$

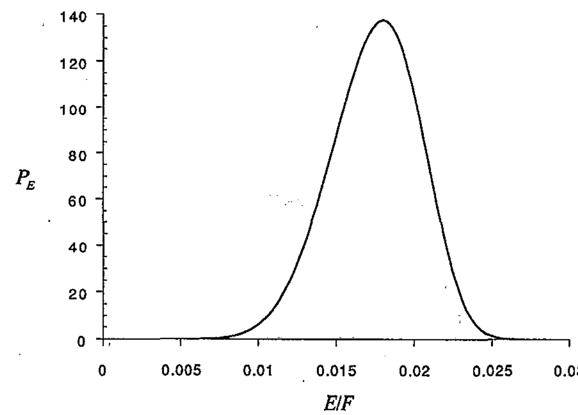

FIG. 3. The distribution of the probability that a member of the ensemble is dominated by an eigenstate of frequency E in one dimension, for $Ft=10^8$, $\gamma=0.5F$, and $C=0.5$. We observe that function cuts off quickly at $E/F\sim0.023$. The curve was generated by performing the indicated integration in expression (3.7). The localization length was taken to be $l\sim 1/\sqrt{\gamma C}=2.0$, and $\eta=8.3$. The localization length of 2.0 was also confirmed numerically, using a transfer matrix algorithm. The parameter η was determined by comparing the mean-square displacement of the spatial distribution as found from numerical simulations with the predictions of expression (3.18), cf. Fig. 5 below.

associated with the trap-free voids is obtained, where $\lambda=|\ln(1-C)|$ and C is the fraction of sites occupied by traps. It is these exponential tails in the spectral density that are often referred to as Lifshitz tails. If we substitute Eq. (3.10) in Eq. (3.5), we obtain, to leading order in E/ϵ_0,

$$\rho_D^{(n)}(E)\sim\left(\frac{\lambda DV_D}{2\epsilon_0}\right)^{1-n}\left(\frac{E}{\epsilon_0}\right)^{(D+2)(n-1)/2}\times\exp[-\lambda V_D(\epsilon_0/E)^{D/2}]. \tag{3.11}$$

A. The distribution of dominating frequencies

In order to examine the distribution of the probability that a member of the ensemble is dominated at the time t by an eigenstate of frequency E, we have numerically integrated Eq. (3.7). The results of this integration for one dimension are shown in Fig. 3. Note that the distribution cuts off sharply on the high frequency side. The probability to find trap-free regions which dominate the series in Eq. (2.8), but which are also smaller than a certain critical volume, becomes exceedingly small at long times. We shall refer to the value of E at which this cutoff occurs as E_c. That the distribution cuts off abruptly above a certain eigenvalue is not surprising. Owing to their scarcity, the spatial separation between successively dominating trap-free regions, being inversely proportional to their density, is a rapidly increasing function of their size. As a result, the pausing time between hops increases exponentially with the size of the trap-free regions being considered for next-nearest-neighbors. It is therefore likely that hopping among the smaller trap-free regions will be completed long before any hop occurs to larger trap-free regions. If the variable z is substituted for $\alpha r/t$ in Eq. (3.7), the distribution of (dominant) eigenvalues can be written

$$P_E(E,t)=\left(\frac{t}{\alpha a}\right)^D \rho_D(E)\int_0^\infty D\,\eta_D V_D z^{D-1}\,dz\,\Omega_D \times(z+E,t), \tag{3.12}$$

where

$$\Omega_D(E,t)=\exp\left[-\frac{D!V_D\eta_D t^D}{(\alpha a)^D}\,\rho_D^{(D+1)}(E)\right]. \tag{3.13}$$

In the limit in which $t\to\infty$, the function $\Omega_D(E,t)$ is a step function, $1-\Theta(E-E_c)$. It is precisely this behavior which accounts for the abrupt cutoff seen in Fig. 3. To find an analytic expression for the cutoff, we choose E_c to be that frequency at which $\Omega_D(E,t)$ is equal to 1/2. If we set the logarithm of Eq. (3.13) equal to ln(1/2), and then substitute Eq. (3.11) for $\rho_D^{(D+1)}(E)$, an equation for E_c may be obtained by iteration

$$\left(\frac{E_{\rm cut}}{\epsilon_0}\right)^{D/2}=\frac{\lambda V_D/(D+2)}{\ln(A)+\ln\left(\dfrac{1}{\ln(A)+\ln(1/\ln(A)+\cdots)}\right)}; \tag{3.14}$$

where

$$A=\frac{\lambda V_D}{D+2}\left(\frac{D!V_D\eta_D}{\ln(2)}\right)^{1/(D+2)}\left(\frac{2\epsilon_0 t}{\lambda\alpha a D V_D}\right)^{D/(D+2)}. \tag{3.15}$$

At long times this has the leading behavior

$$E_c\sim\left[\frac{\lambda V_D}{D\,\ln(2\epsilon_0 t/\lambda\alpha a D V_D)}\right]^{2/D}. \tag{3.16}$$

Thus in the ensemble, the eigenvalue associated with the smallest of the dominating trap-free regions approaches zero as $1/\ln(t)^{2/D}$. Returning to Eq. (3.12), we argue that the integration over z needs to be completed only up to E_c. Asymptotically this yields the result

$$P_E(E,t)\sim\begin{cases}\eta_D V_D\left[\dfrac{(E_c-E)t}{\alpha a}\right]^D\rho_D(E), & 0<E\leqslant E_c,\\ 0, & E_c>E.\end{cases} \tag{3.17}$$

B. The spatial distribution of the dominant eigenstates

That the width of the spatial distribution of the survival probability increases with time is not surprising. As we have discussed above, the hopping process resembles a slow march to larger and larger trap-free regions. This is characterized by the rate at which E_c steadily decreases towards the band edge. Since the average distance between trap-free regions is inversely proportional to the density of states in the Lifshitz tail, these hops occur over ever-increasing distances. This compensates somewhat for the fact that the pausing time between hops is also increasing, and leads to the result that the spatial distribution of the survival probability grows superdiffusively. To demonstrate this, we examine the moments of the spatial distribution function expressed by Eq. (3.6). The nth moment can be written

$$\overline{r^n}=\int_0^\infty r^n P_r(r,t) D V_D r^{D-1}\,dr$$
$$=\int_0^\infty \frac{D\,\eta_D V_D r^{D+n-1}\,dr}{a^D}\int_0^\infty \rho_D(E)dE \times\exp\left[-\frac{D!V_D\eta_D t^D}{(\alpha a)^D}\,\rho_D^{(D+1)}(\alpha r/t+E)\right]. \tag{3.18}$$

One of the two integrations in Eq. (3.18) can be performed if the coordinate system is rotated. Changing integration variables from r and E to $\mathfrak{R}=\alpha r+Et$ and $\aleph=\alpha r-Et$ (3.18) becomes

$$\overline{r^n}=\frac{D\,\eta_D V_D}{2\alpha t}\int_0^\infty d\mathfrak{R}\int_{-\mathfrak{R}}^{\mathfrak{R}} d\aleph\,\frac{(\mathfrak{R}+\aleph)^{D+n-1}}{a^D(2\alpha)^{D+n-1}}\,\rho_D\left(\frac{\mathfrak{R}-\aleph}{2t}\right) \times\exp\left[-\frac{D!V_D\eta_D t^D}{(\alpha a)^D}\,\rho_D^{(D+1)}(\mathfrak{R}/t)\right]. \tag{3.19}$$

Integrating over $\aleph$ and reintroducing the integration variable $E=\mathfrak{R}/t$, we find

$$\overline{r^n}=\frac{(D-1+n)!\,\eta_D D V_D t^{D+n}}{(\alpha a)^D\alpha^n}\int_0^\infty dE\,\rho_D^{(D+n)}(E) \times\exp\left[-\frac{D!V_D\eta_D t^D}{(\alpha a)^D}\,\rho_D^{(D+1)}(E)\right]. \tag{3.20}$$

If we now perform the integration over E by parts, we obtain the general result,

$$\overline{r^n}=t^n\times\frac{(D-1+n)!}{(D-1)!\alpha^n}\int_0^\infty dE\,\Omega_D(E,t)\,\frac{d}{dE}\left[\frac{\rho_D^{(D+n)}(E)}{\rho_D^{(D)}(E)}\right] \tag{3.21}$$

for $n>0$. The integrand in Eq. (3.21) is the product of an algebraic function of E and the function $\Omega_D(E,t)$, which is asymptotically a step function. The asymptotic expression for the nth moment is therefore

$$\overline{r^n}\sim t^n\times\frac{(D-1+n)!}{(D-1)!\alpha^n}\int_0^{E_c} dE\,\frac{d}{dE}\left(\frac{\rho_D^{(D+n)}(E)}{\rho_D^{(D)}(E)}\right)$$
$$=t^n\frac{(D-1+n)!}{(D-1)!\alpha^n}\,\frac{\rho_D^{(D+n)}(E_c)}{\rho_D^{(D)}(E_c)}. \tag{3.22}$$

If we now substitute Eq. (3.11) in Eq. (3.22), we obtain

$$\overline{r^n}\sim t^n\,\frac{(D-1+n)!}{(D-1)!\alpha^n}\left(\frac{2\epsilon_0}{\lambda D V_D}\right)^n\left(\frac{E_c}{\epsilon_0}\right)^{(D+2)n/2}. \tag{3.23}$$

Finally, substituting Eq. (3.16) for E_c, we obtain the asymptotic form

$$\overline{r^n}\sim\frac{(D-1+n)!}{(D-1)!}\,\tau^n \tag{3.24}$$

for the moments of $P_r(r,t)$, where

$$\tau=2\left(\frac{\lambda V_D}{D^{D+1}}\right)^{2/D}\frac{\epsilon_0 t/\alpha}{\ln^{(D+2)/D}(\epsilon_0 t/\alpha a)}. \tag{3.25}$$

That transport is superdiffusive follows from Eqs. (3.24) and (3.25). We note, in fact, that the variable τ is nearly proportional to time: were it not for the logarithmic factor in Eq. (3.25), the transport here might be described as ballistic.

From the moments, it is straight forward to obtain the asymptotic spatial distribution function. To this end, it is convenient to introduce the Fourier transform

$$\tilde{P}_r(k,t)=\int_0^\infty e^{ikr}P_r(r,t)dr\ r^{D-1}DV_D. \qquad (3.26)$$

Expanding the right-hand side of Eq. (3.26) in powers of k, we obtain a power series for $\tilde{P}_r(k,t)$ in terms of the moments expressed by Eq. (3.24),

$$\tilde{P}_r(k,t)=\sum_{n=0}^{\infty}\frac{(ik\tau)^n}{n!}\frac{(D+n-1)!}{(D-1)!}=\frac{d^{D-1}}{d\tau^{D-1}}\left(\frac{\tau^{D-1}}{1-ik\tau}\right). \qquad (3.27)$$

The Fourier inversion of Eq. (3.27),

$$P_r(r,t)=\frac{d^{D-1}}{d\tau^{D-1}}\left(\frac{\tau^{D-1}}{2\pi}\int_{-\infty}^{\infty}\frac{dk\ e^{-ikr}}{DV_Dr^{D-1}}\frac{1}{1-ik\tau}\right) \qquad (3.28)$$

requires a straightforward integration, with the result that in any dimension $P_r(r,t)$ is simply an exponential function

$$P_r(r,t)=\frac{e^{-r/\tau}}{\tau^D DV_D} \qquad (3.29)$$

of the radius r.

IV. DISCUSSION

For periodically placed traps it is useful to consider a description of the system on a length scale much larger than the separation between traps. On this scale the system is homogeneous with an effective uniform trapping rate which is given by the smallest eigenvalue in the system. Therefore, except for an overall exponential decay, the spatial distribution is described by a Gaussian, the mean-square displacement of which increases linearly in time. In view of this, the main results of this paper, expressed in Eqs. (3.24) and (3.29) are remarkable for at least two reasons, apart from any future applications. First, it is amazing that such dramatically different transport behavior should arise from the apparently subtle change in the arrangement of the traps produced by disorder. Second, even if it were anticipated that disorder might radically alter the transport, it is still not obvious that it should cause superdiffusion. Of course, we (and others) have made much of the fact that the eigenstates of the disordered system are localized, whereas the eigenstates of the ordered system are extended, but these considerations do not by themselves constitute a physical picture. Such a picture may be had by discussing the underlying diffusion in terms of particles undergoing a continuous-time random walk. If the traps are placed randomly, then some trap-free voids will be larger than others, and furthermore there will be remote voids which are larger than any of those found near the origin. At long times, the surviving particles will be predominantly those which have arrived in those larger and more distant voids, and their paths must necessarily be those which tend to avoid early capture. Thus the randomly formed voids act as a filter, preferentially preserving those rare particles which travel to the large voids quickly, or in nearly a straight line. Particles which take more tortuous paths take longer to reach the larger voids, and are more likely to be trapped. The process of diffusion is described by considering all paths. This includes typical paths, which are quite tortuous, in addition to those rare paths which are nearly ballistic. On the other hand, superdiffusion, as described by Eq. (3.24), comes about when the nearly ballistic paths are preferentially singled out by considering only that which has not yet been trapped.

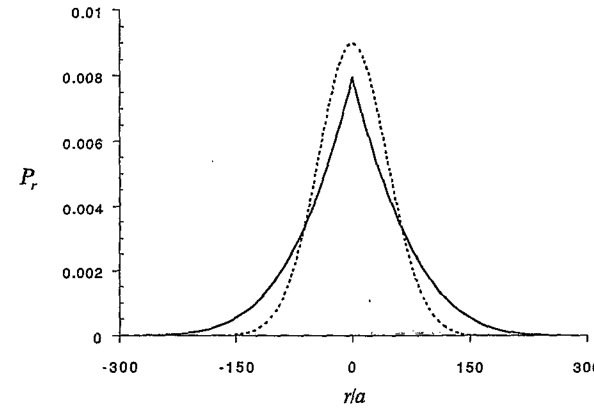

FIG. 4. The average spatial distribution of surviving particles as a function of the distance from the origin in one dimension, for $Ft=1000$, $\gamma=0.5F$, $C=0.5$, $l=2.0$ and $\eta=8.3$. The solid curve was generated by performing the integration in expression (3.6) numerically. The dotted line shows the Gaussian distribution which is obtained if the same traps are placed periodically, rather than at random. The width of the distribution grows more quickly with time in the disordered case, as is shown in Fig. 5.

In order to illustrate the preceding discussion, we have displayed in Fig. 4 the spatial distribution of surviving particles for $Ft=1000$, in one dimension, as expressed by Eq. (3.6). In this calculation the trapping rate γ was chosen to be $0.5F$, and the trap concentration C was taken to be 0.5: This gives rise to an inverse localization length α of 0.5. On the same graph we have shown the distribution of surviving particles which would obtain at $Ft=1000$ if traps of the same strength and concentration were instead placed on the lattice periodically. We see here that what remains untrapped is distributed over a much larger region for the disordered case. For the same concentration and trapping rate used in Fig. 4, we have numerically integrated Eq. (2.1) in one dimension for 1000 random trapping configurations. The configurational average of the resulting mean-square displacement is shown in Fig. 5. The smooth curve which has been superimposed is Eq. (3.21). Again for comparison, in the same figure we have also shown the mean-square displacement of the survival probability for the case in which the traps have been placed on the lattice periodically, rather than at random. It is apparent that the disorder begins to have a noticeable affect on the mean-square displacement at rather short times, when $Ft\sim 100$.

The effects discussed here have wide application. Let us suppose that Eq. (2.1) describes a diffusing chemical species

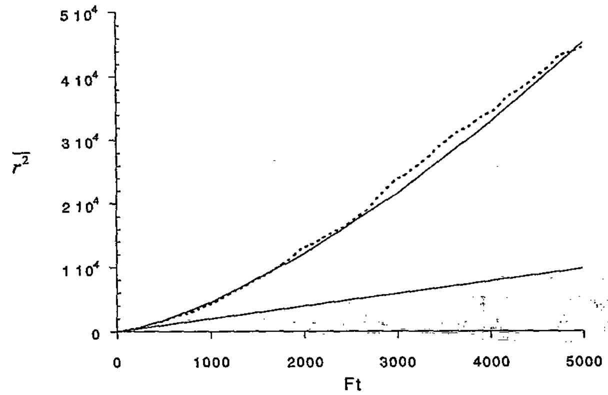

FIG. 5. The configuration average of the mean-square displacement $\overline{r^2}$ (in units of a^2) of the spatial distribution of surviving particles as a function of reduced time Ft, in one dimension. The trapping rate $\gamma=0.5F$ and the concentration of traps $C=0.5$. The dotted line is the average of results obtained by numerically integrating Eq. (2.1) for 1000 randomly chosen trap configurations. The solid line which has been superimposed was calculated by integrating Eq. (3.18) for $l=2.0$. A reasonable agreement with the numerical simulations is had by setting the parameter $\eta=8.3$. The fact that these lines are superlinear indicates that motion is superdiffusive. For the case in which the traps are placed periodically with the same concentration (every other site is a trap), the mean-square displacement is linear with time asymptotically, such that $\overline{r^2} \sim 8a^2Ft/\sqrt{16+(\gamma/F)^2} = 1.985\ a^2\ Ft$. This line has been shown on the graph for comparison. We note that at $Ft \sim 100$ the onset of superdiffusive behavior begins to clearly distinguish the disordered case from the ordered case.

which may react with fixed centers. In such a case, our analysis shows that the unreacted species will necessarily be spread over a much wider volume than would be expected from normal diffusion. The reaction could be, for example, the trapping of charge carriers in disordered molecularly doped polymers. In such a case, the distribution of untrapped carriers will affect observables such as the width of the signal in a time-of-flight measurement of the mobility. Bioremediation of contaminated soils presents another application, in which activated bacteria act as randomly placed traps for the contaminant molecules. The equations describing the infiltration of the plume reduce to diffusionlike equations for low concentrations. From the results above, we see that the part of the plume which remains unremediated is likely to be located further from the contaminant source than what would be predicted if the bacteria were assumed to be homogeneous.

V. CONCLUSION

In this paper we have presented an analysis of diffusion in the presence of randomly placed traps, as described by Eq. (2.1). We have described the evolution of Eq. (2.1) through an eigenfunction expansion, and have focused on interesting new effects which arise because of the localization of the eigenstates at the band edge. We have shown that the distribution of surviving particles evolves as though it were hopping from one trap-free region to the next. Thus we have applied the method of Apsely and Hughes, originally developed to describe variable range hopping, to calculate the spatial distribution of the surviving particles. We have found that the distribution is an exponential function, with a width which increases almost linearly with time. To our knowledge this is the first time an asymptotic expression for the spatial distribution function of surviving particles has been obtained either for the random trapping problem, or for any other variation of Eq. (2.1). Whether the distribution remains a simple exponential function for other functional forms of the trapping rates remains an open question.

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