Classical Diffusion in One-Dimensional Disordered Lattice

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Classical diffusion of localized excitations is investigated on a one-dimensional chain with (energy-independent) nearest-neighbor transfer rates $W_{n,n+1}=W_{n+1,n}$ that are independently distributed according to a probability density $\rho(W)$. An exact formal solution is derived for $\langle P_0(t)\rangle$, the time development of the initial excitation. The long-time decay of $\langle P_0(t)\rangle$, determined by the behavior of $\rho(W)$ near $W=0$, is analyzed in detail for arbitrary probability densities $\rho(W)$.

Based on classical rate equations, we investigate the diffusion of localized excitations (or particles) in one-dimensional disordered systems. Specifically, we consider an infinite one-dimensional lattice with only nearest-neighbor transfer rates $W_{n,n+1}=W_{n+1,n}$ that are distributed (independently) according to a probability density $\rho(W)$. At time $t=0$ a single site is excited, $P_n(t=0)=\delta_{n,0}$, and the excitation amplitude P_n at the nth site develops according to

$$\frac{dP_n}{dt}=W_{n,n+1}(P_{n+1}-P_n)+W_{n-1,n}(P_{n-1}-P_n). \quad (1)$$

The purpose of this Letter is to calculate the long-time development of the initial excitation, $\langle P_0(t)\rangle$, and to establish its dependence on the functional form of the probability density $\rho(W)$. The average $\langle\cdots\rangle$ is defined with respect to the distribution of the (independent) random variables $W_{n,n+1}$.

Our problem is relevant to the study of transport properties in a number of different physical systems. It is particularly appropriate to fluorescent-line-narrowing experiments.[1] An inhomogeneously broadened optical line is subjected to a narrow laser pulse, and the subsequent time development of the emission profile is governed by phonon-assisted energy transfer between sites with different excitation energies (spectral diffusion). The above model corresponds to a situation where the site-site transfer rates are energy-mismatch independent,[2] and the emission amplitude of those sites initially excited is then represented by $\langle P_0(t)\rangle$.[3]

The quantity $\langle P_0(t)\rangle$ is also related to the diffusibility of a particle in a disordered one-dimensional lattice. It represents the probability of finding the particle after time t at its initial position. Further, $\langle P_0(t)\rangle$ is the Laplace transform of the energy-eigenvalue density of states $N(\epsilon)$. Our long-time results for $\langle P_0(t)\rangle$ therefore determine the low-energy behavior of $N(\epsilon)$ for systems which are described by eigenvalue equations analogous to those generated by Eq. (1)[4]: lattice vibrations of a linear chain with random force constants, electronic impurity bands in one dimension, and low-temperature excitations of random one-dimensional ferromagnets.

An exact formal solution for $\langle P_0(t)\rangle$ can be derived for the type of one-dimensional systems defined above. If $\tilde{P}_0(\omega)$ denotes the Laplace transform of $P_0(t)$, we obtain

$$\langle\tilde{P}_0(\omega)\rangle=\int dg_1\,dg_2\,f_\omega(g_1)f_\omega(g_2)\frac{1}{\omega+g_1+g_2}, \quad (2)$$

where $f_\omega(g)$ denotes the probability density of the following infinite continued fractions:

$$g=\cfrac{1}{\cfrac{1}{W_{0,1}}+\cfrac{1}{\omega+\cfrac{1}{\cfrac{1}{W_{1,2}}+\cdots}}}. \quad (3)$$

As the $W_{n,n+1}$ are independent random variables, we can construct an exact integral equation for $f_\omega(g)$, with ω real:

$$f_\omega(g)=\int dg'f_\omega(g')\int dW\rho(W)\delta\left(g-\left[\frac{1}{W}+\frac{1}{\omega+g'}\right]^{-1}\right)=\int_{g-\omega}^{\infty}dg'f_\omega(g')\left[\frac{g'+\omega}{g'+\omega-g}\right]^2\rho\left(\frac{g(g'+\omega)}{g'+\omega-g}\right). \quad (4)$$

Notice that the support of f_ω is $\{g|g\geqslant 0\}$. The solution of Eq. (4) determines $\langle P_0(\omega)\rangle$ for real ω, and $\langle P_0(t)\rangle$ is given by the inverse Laplace transform of its analytic continuation into the right half-plane.

In the following we concentrate on the long-time behavior of $\langle P_0(t)\rangle$. It is well known that in the ordered system, i.e., for $\rho(W)=\delta(W-W_0)$, $\langle P_0(t)\rangle$ shows the usual diffusive behavior,

$$\langle P_0(t)\rangle \approx (4\pi W_0)^{-1/2} t^{-1/2}, \quad t\to\infty. \tag{5}$$

In a previous paper,[5] we have calculated $\langle P_0(t)\rangle$ for the case when $\rho(W)$ contains a δ function at $W=0$. This corresponds to a chain with random interruptions, a problem examined in different contexts by a number of authors.[6] We found[5] that in the limit of very long times $\langle P_0(t)\rangle$ decays exponentially to a constant value $P_\infty>0$, so that

$$\langle P_0(t)\rangle - P_\infty \sim \exp(-t^{1/3}). \tag{6}$$

This is a consequence of the localization of the excitations on segments of finite length.

From now on we exclude probability densities $\rho(W)$ which contain a δ function at $W=0$, so that $\langle P_0(t)\rangle$ will eventually decay to zero. We shall see, however, that the decay can be much slower than given by the diffusion result of Eq. (5). To calculate the long-time behavior of $\langle P_0(t)\rangle$ we have to determine the behavior of $\langle \tilde{P}_0(\omega)\rangle$ for small real ω. A careful investigation of the integral Eq. (4) shows that for small ω, $f_\omega(g)$ can be approximated by a δ function, so that

$$f_\omega(g) \approx \delta(g-g_0(\omega)), \quad \omega\to 0, \tag{7}$$

for all $\rho(W)$ that do not contain a δ function at $W=0$. The relative error introduced in averages, e.g., of the form of Eq. (2), vanishes as $\omega\to 0$. Although at the moment our estimations of the corresponding corrections are not yet entirely rigorous,[7] we believe that our results are exact in the limit as $\omega\to 0$ (or $t\to\infty$). They are furthermore very accurately confirmed by numerical calculations, as discussed below. The full details of our investigations will be published elsewhere.

We were able to construct a simple iteration scheme to solve Eq. (4) numerically. The procedure works very well if $f_\omega(g)$ is essentially concentrated on a bounded support and could be applied to equations of a similar type (e.g., those of Dyson[8]). Figure 1 shows results for a uniform distribution of W's,

$$\rho(W)=\begin{cases}1, & 0\le W\le 1,\\ 0, & \text{otherwise},\end{cases} \tag{8}$$

and demonstrates qualitatively the narrowing of $f_\omega(g)$ as ω decreases.

If $f_\omega(g)$ is replaced by a δ function, Eq. (7), it follows that $g_0(\omega)$ is given by

$$g_0=\int_0^\infty dW\,\rho(W)\left(\frac{1}{W}+\frac{1}{\omega+g_0}\right)^{-1}. \tag{9}$$

For very narrow distributions $f_\omega(g)$, the average value of g, $\langle g\rangle = \int dg\, g f_\omega(g)$, is accurately approximated by g_0. We have calculated $\langle g\rangle$ numerically for various $\rho(W)$ by using Monte Carlo methods to generate high-order continued fractions of the type of Eq. (3). Figure 2 shows that the agreement between the numerical results and the predictions of Eq. (9) is excellent. From the small-ω behavior of $\langle \tilde{P}_0(\omega)\rangle$, as determined by Eqs. (2), (7), and (9), we can distinguish the following categories of probability densities $\rho(W)$ which lead to qualitatively different results for the long-time decay of $\langle P_0(t)\rangle$:

(a) If $\rho(W)$ is such that $1/W_{\rm eff} \equiv \int dW\,\rho(W)/W$ exists, we obtain $g_0 \approx (W_{\rm eff}\,\omega)^{1/2}$ as $\omega\to 0$, and $\langle P_0(t)\rangle$ therefore follows the one-dimensional diffusion result,

$$\langle P_0(t)\rangle \approx (4\pi W_{\rm eff})^{-1/2} t^{-1/2}, \quad t\to\infty. \tag{10}$$

(b) If $\rho(W)$ is finite at $W=0$, we have $g_0 \sim (-\omega/\ln\omega)^{1/2}$ as $\omega\to 0$, and

$$\langle P_0(t)\rangle \sim (\ln t/t)^{1/2}, \quad t\to\infty. \tag{11}$$

(c) If $\rho(W)$ diverges as $W^{-\alpha}$ for $W\to 0$, g_0 be-

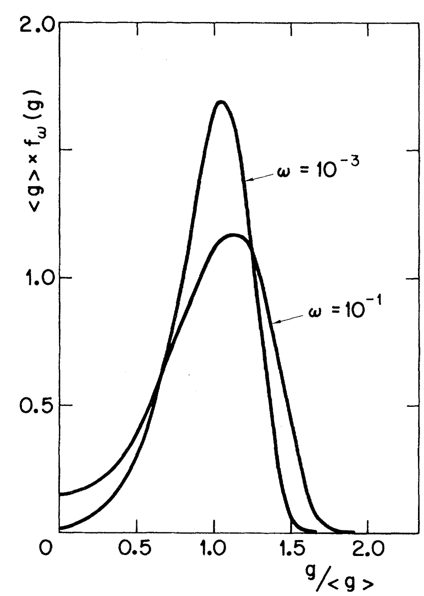

FIG. 1. Results of a numerical solution of Eq. (4) for the uniform probability density of Eq. (8).

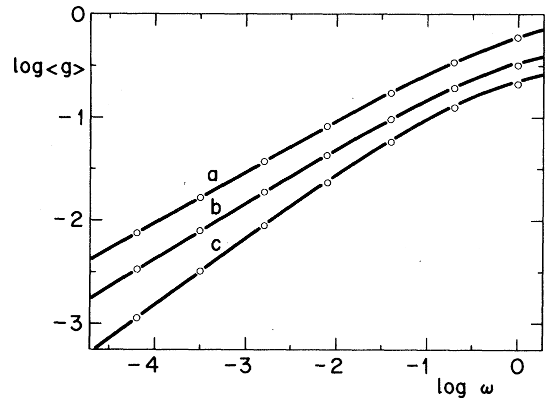

FIG. 2. $\langle g \rangle$, the average value of g [Eq. (3)] for three different distributions of W's: (a) W uniformly distributed in $\frac{1}{2} \leq W \leq \frac{3}{2}$; (b) W uniformly distributed in $0 \leq W \leq 1$; (c) $\rho(W) = \frac{1}{2} W^{-1/2}$ for $0 \leq W \leq 1$. Monte Carlo results (circles) are compared with the predictions $g_0 \approx \langle g \rangle$ of Eq. (9) (full curves).

haves asymptotically as $g_0 \sim \omega^{1/(2-\alpha)}$, and

$$\langle P_0(t)\rangle \sim t^{-(1-\alpha)/(2-\alpha)}, \quad t \to \infty. \tag{12}$$

In conclusion, we have investigated in detail a simple one-dimensional model for spectral diffusion in disordered systems. We have shown how the long-time decay of the initial excitation is determined by the distribution of transfer rates, $\rho(W)$. Depending on the form of $\rho(W)$ near $W = 0$ we can distinguish four categories with qualitatively different long-time behavior. Compared with the ordinary diffusion result, Eq. (10), the long-time decay becomes slower and slower as the fraction of very small transfer rates increases. Corresponding to Eqs. (11) and (12) we may speak of weak and strong *quasilocalization* of the excitations. Systems with distributions $\rho(W)$ that contain a δ function at $W = 0$ have to be studied by different methods.[5] Here we have *true localization* of the excitations, and the initial excitation decays exponentially to an enhanced constant value as $t \to \infty$ (see Ref. 5). These conclusions supplement the results of Theodorou and Cohen[9] concerning the localization of eigenstates in one-dimensional systems with off-diagonal disorder.

Our very general results for $\langle P_0(t)\rangle$ may be of direct use in fluorescent-line-narrowing experiments, and (via the energy density of states) for the analysis of low-temperature specific-heat measurements in one-dimensional systems with random site-site couplings.[10] It is not clear whether we may expect similar results in higher dimensions. The recent investigations of Antoniou and Economou[11] seem to exclude the occurrence of some sort of quasilocalization as we pass from ordinary diffusive behavior to true localization (in percolation systems).

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