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We consider the transport of electrons or excitons through a random environment in the presence of constant site off-diagonal dephasing processes. It is shown that dephasing of this form will always defeat Anderson localization at long times. Some recent theories of quantum localization which depend upon such dephasing processes are, therefore, seen to be inconsistent.

A quantum particle moving through a random environment is a common scenario for electron and exciton transport in numerous disordered materials.[1–7] The most frequently studied model that has been advanced in this context is due to Anderson.[1] In the Anderson model, an electron hops among a set of energetically disordered lattice sites.[1] Delocalized (current carrying) states fail to occur when the energetic degeneracy of the lattice sites is raised above some critical value. The quantum percolation model, in which the sites are all energetically degenerate, exhibits a similar localization transition when the fraction c of removed sites exceeds a critical value c^*. In both models, localization arises purely from elastic scattering, which tends to destroy the spatial phase coherence of the quasiparticle wave functions.[3] In real disordered materials, however, the question of observing quantum localization is complicated by the fact that inelastic processes, due to electron–electron and electron–phonon interactions, can mask the purely quantum mechanical nature of Anderson localization. In fact, the effects that phonon interactions and quasiparticle correlations have on quantum localization are topics of considerable current interest.[3–5]

In a series of recent papers,[6,7] Loring and Mukamel (LM) have presented a self-consistent dephasing theory of quantum localization which is based upon an effective-medium type treatment of the Liouville equation for the single particle density matrix. Their approach is surprisingly simple to implement and, at first sight, appears to correctly recover the main features of the currently accepted (zero-temperature) scaling theory of localization developed by Abrahams *et al.*[2] While we feel that the idea of a density matrix approach similar to that suggested by LM has considerable merit, we feel the need to point out that the approximate manner in which they have closed their self-consistent equations is fundamentally inconsistent and therefore inappropriate as a tool for studying quantum localization. In particular, we show in this paper that the starting equations of LM are inherently incapable of yielding any information about quantum localization at the (infinite) time scales relevant to their analysis. Hence, it is our conclusion that the apparent agreement of their work, with the results of quantum transport theory, is largely coincidental and stems from the artificial manner in which they close their self-consistent equations. In fact, their results are attributable, we believe, to classical divergences in the diffusive propagator describing density fluctuations, and are not directly related to the real source of quantum localization.[8]

The starting point of the analysis of Loring and Mukamel is a stochastic Liouville equation (SLE) for the time evolution

$$\frac{d\rho}{dt} = -i[H,\rho] - \Lambda\rho \qquad (1)$$

for the single particle density matrix ρ.[6,7] The Hamiltonian H describes quasiparticle transport in the disordered system. In Refs. 6 and 7, H is taken to be of the tight binding, nearest neighbor form, although this restriction is not crucial to the observations which follow. The last term in Eq. (1), $\Lambda\rho$, is a dephasing term which can be simply written as

$$\begin{aligned}[\Lambda\rho]_{mn} &= \Gamma(1-\delta_{mn})\rho_{mn}\\ &= [\Gamma(1-P)\rho]_{mn}\,,\end{aligned} \qquad (2)$$

where P projects out the diagonal elements of a matrix in the site representation. The use of a dephasing term such as in Eq. (2) has a long history in the study of translationally invariant systems, where it has provided a convenient but phenomenological way to introduce scattering into the coherent motion described by the single particle Hamiltonian H.[9,10] In particular, it provides a simple means for describing the extremes of coherent and incoherent motion through a single parameter Γ/J, where J is the width of the band. Much is known about the solutions to the SLE for translationally invariant or ordered systems. For example, it is known that the mean square displacement of an initially localized particle grows as t^2 for times $t \ll \Gamma^{-1}$, while eventually, for $t \gg \Gamma^{-1}$, it grows linearly in time, hence describing a diffusive process. It is worth noting, however, that dephasing of this and more complex forms have primarily been used to model inelastic processes arising from electron–phonon or electron–electron interactions.

Nonetheless, while stochastic Liouville equations for ordered systems have been studied extensively, the effects of disorder on the SLE, or conversely, the general effects of various types of dephasing on localization have not. Some interesting work in this direction has been done recently, but very few exact results are known.[4,5] For example, while it is easily shown that a translationally invariant system de-

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scribed by such an SLE equilibrates to a uniform, delocalized particle distribution, there is apparently some question as to whether this is intimately connected to dephasing, or is in fact just a reflection of the translational invariance of the eigenstates of the Hamiltonian. Hence, it may not necessarily follow that in a strongly disordered system equilibration to a delocalized particle distribution will also occur. Indeed, if a disordered Hamiltonian has only localized eigenstates, one might imagine that the corresponding SLE, if it equilibrates at all, would do so to a localized distribution which reflects the localization of the eigenstates of the Hamiltonian. Although not explicitly stated, this assumption implicitly underlies the analysis of Loring and Mukamel, who base their results regarding quantum localization on an analysis of the infinite time (actually zero-frequency) solution to Eq. (1) for a particle initially located at a single site.[6,7] Their reliance upon this assumption is most strongly revealed by the fact that at *no time* do they take the limit $\Gamma \to 0$, which is the usual starting point for an analysis of the quantum localization problem. Indeed, their results for the diffusion coefficient above the "threshold" actually diverge in this limit because of the dependence on the pure crystal diffusion constant which is proportional to Γ^{-1}. The resultant implication of the LM analysis that the mean square displacement necessarily grows a t^2 above the threshold for $\Gamma = 0$ is in disagreement with the currently held view.[1-5,8-10]

More specifically, however, the assumption of localized equilibration underlying the analyses of Loring and Mukamel is *not correct.* In fact, for any initial condition the solutions to Eq. (1) approach a stationary distribution which is spatially delocalized. This can be shown in a number of different ways; the proof which follows, however, is succinct and clearly demonstrates the essential physics involved. We first consider the evolution of the norm of the density matrix $\|\rho\| = \mathrm{Tr}\,\rho^2$, under the action of the stochastic Liouville operator implicitly defined in Eq. (1), and for some physically relevant initial condition. The exact form of the initial density-matrix is not important except that it must be Hermitian. The time derivative of $\|\rho\|$ may be written

$$\frac{d}{dt}\|\rho\| = 2\,\mathrm{Tr}\,\dot{\rho}\rho$$

$$= 2\,\mathrm{Tr}\{H\rho\rho - \rho H\rho\} - 2\,\mathrm{Tr}\{(\Lambda\rho)\rho\}. \quad (3)$$

The term involving H vanishes since we may cyclically permute operators under the trace. This leaves the dephasing term, from which we obtain the evolution

$$\frac{d}{dt}[\mathrm{Tr}\,\rho^2] = -2\Gamma\,\mathrm{Tr}[\rho(1-P)\rho], \quad (4)$$

for the norm of ρ. Recall that $1-P$ projects onto the off-diagonal part of a matrix in the site representation. Consequently, the trace on the right hand side of Eq. (4) is simply the sum of the absolute squares of the off-diagonal elements of the density matrix in the site representation. The right-hand side of Eq. (4) is therefore negative semidefinite. Thus, at any time during which the density matrix has nonzero site off-diagonal elements, $\|\rho\|$ will monotonically decrease. The norm of ρ, however, is positive definite and so it is bounded from below by zero. Thus, there is irreversible relaxation until $\|\rho\|$ reaches its minimum value. In this limit the time derivative is zero and thus the off-diagonal elements ρ_{nm} vanish identically.

It is the vanishing of the off-diagonal elements ρ_{nm} at long times which leads to a delocalized spatial population. This can be seen most clearly from the equation of motion for an off-diagonal element,

$$\frac{d\rho_{nm}}{dt} = -i\sum_r [H_{rm}\rho_{nr} - H_{nr}\rho_{rm}] - \Gamma\rho_{nm}, \quad (5)$$

which will be seen to depend upon the diagonal elements only through the term $H_{nm}(\rho_{nn} - \rho_{mm})$. At long times both ρ_{nm} $(n \neq m)$ and its derivative vanish by the arguments already given and so Eq. (5) reduces to $\rho_{nn} = \rho_{mm}$ if $H_{nm} \neq 0$. Thus, any two sites directly connected by a nonzero matrix element have equal populations as $t \to \infty$. By transitivity this equality can be extended to any two sites which have a path of nonzero matrix elements of the Hamiltonian connecting them. As advertised then, we see that purely off-diagonal dephasing of this type thwarts quantum localization, and leads at long times to an equal population at all sites. That is, at long times a particle obeying Eq. (1) will explore the entire region of the lattice to which it is initially connected. In the context of the percolation problem, therefore, the solution to the SLE at long times for any finite amount of dephasing necessarily has the localization properties characteristic of *classical* not quantum percolation. It is worth noting that in a later paper from the same group[11] an analysis is given of the classical site percolation problem in a manner closely resembling the treatment of the "quantum" site percolation problem presented in Ref. 6. Although the underlying physics treated in these two papers is drastically different, the transport properties predicted are identical in every aspect. As the analysis above shows, this agreement is at least internally self-consistent since the SLE in Eq. (1) does in fact describe classical percolation at long times. This reinforces the fact that any attempt to obtain information about the localization of eigenstates of H from the asymptotic properties of the SLE in Eq. (1) is impossible.

Our criticism of the analyses of LM then is as follows. The manner in which they close their self-consistent equations necessitates that they include a constant off-diagonal dephasing term in their starting equations if they are to obtain a quantum transition. In fact, unlike the ordinary situation, their results depend crucially upon Γ being finite. We have shown, however, that whenever the off-diagonal dephasing term has a finite real part as $t \to \infty$, the off-diagonal elements of the density matrix will decay to zero. Because of the structure of the SLE this state of affairs leads at long times to an equal population at all connected sites, that is, to a delocalized probability distribution. Hence, for finite Γ a quantum transition *cannot* occur, in contradiction to the results obtained by Loring and Mukamel. While it is certainly true that at times short relative to Γ^{-1}, the quasiparticle will appear to be localized, in the long time limit (or equivalently, at a time scale long relative to Γ^{-1}, the relevant time scale for the LM analysis) localization cannot occur. That the time scale relevant to the LM work is long compared to Γ^{-1} can be inferred from an approximation [Eq. (16) Ref. 6, for example] in which a factor $\epsilon + \Gamma$ is replaced by Γ. This substitution necessarily implies that ϵ, the Laplace transform variable conjugate to time, is much less than Γ or equivalently the relevant time scale is $t \gg \Gamma^{-1}$. It would seem then that the form of the relaxation matrix used by Loring and Mukamel, while appropriate for inelastic processes, does not have the correct structure to describe the elastic scattering responsible for Anderson localization.

The present discussion makes clear, however, that the real use of the SLE in the context of quantum localization is to be found in an investigation of the effects of inelastic scattering on localization. Indeed an analysis of the frequency dependent diffusion constant discussed by Loring and Mukamel should reveal the manner in which the inelastic scattering length (which will be proportional to Γ) competes with the zero temperature localization length as well as with the percolation correlation length. In particular, it should be possible to use the SLE of Loring and Mukamel to examine the crossover between classical and quantum percolation either as a function of frequency (time), or as a function of the dephasing rate Γ.

Why then does the LM theory appear to predict a quantum percolation transition? Close inspection of the LM theory shows, we believe, that the divergences which give rise to the purported quantum transition are essentially classical in nature and arise from singularities in the diffusive propagator. As pointed out elsewhere,[8] however, the set of diagrams associated with processes of this type cancel when considered to all orders and do not contribute to nonclassical localization effects. Hence any modification of the approach of Refs. 6 and 7 which would allow a finite result as $\Gamma \rightarrow 0$, would suffer from the same drawback. It would appear then that a correct analysis of Eq. (1), describing in detail the crossover from classical to quantum transport in the limit of zero dephasing, remains an open problem.

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