Journal of Statistical Physics, Vol. 65, Nos. 5/6, 1991

Quantum and Stochastic Aspects of Low-Temperature Trapping and Reaction Dynamics

P. E. Parris[1]

We discuss theoretical work on motion-limited trapping kinetics at very low temperatures where theories based upon strictly diffusive models break down due to quantum mechanical effects. In addition, we present numerical results which confirm earlier asymptotic predictions regarding the survival probability for one-dimensional chains, and discuss the important role fluctuations and self-averaging (or the lack thereof) play in the analysis of finite systems.

KEY WORDS: Trapping; reactions; coherence.

1. INTRODUCTION

Many of the most commonly used experimental methods for investigating exciton transport in organic and inorganic molecular solids rely on the interaction of mobile excitations with other localized species, such as impurity atoms or molecules, which have been doped into the solid so as to irreversibly trap or quench the excitation.[1-14] The prevalence of such methods, along with an increasing interest in diffusion-limited processes arising in other areas of chemistry and physics, has led to considerable work on a standard trapping model, in which a single particle moves in a d-dimensional medium containing randomly-placed irreversible traps in fixed concentration.[15-51] This corresponds to a reaction of the type $A + B \to B$, where A represents the mobile excitation and B represents the stationary molecules of lower energy which can trap or quench the excitation. The primary focus of many previous theoretical studies has been the averaged survival probability $P(t)$, which is defined as the probability for

[1] University of Missouri-Rolla, Rolla, Missouri 65401.

a randomly-placed particle to survive in the medium for a time t without being trapped. When particle motion is diffusive it has been rigorously shown[41,42] that at long times the survival probability has a stretched-exponential form,[33–43] $P(t) \sim \exp[-At^{d/(2+d)}]$, which arises from long-lived particles located in rarely-occurring regions of the medium devoid of traps. Such regions lead to asymptotic tails $\rho(\tau) \sim \exp(-\alpha\tau^{d/2})$ in the distribution of trapping times τ for the eigenmodes of the disordered system, and the stretched-exponential decay that results is sometimes referred to as "Lifshitz-tail" behavior in analogy with tails derived by Lifshitz[52] that appear in the density-of-states (DOS) of energetically-disordered quantum mechanical systems.[52–55]

Despite the rigor with which predictions regarding the long-time behavior have been derived, numerical simulations[19,20] and exact enumeration calculations[22] suggest that, except in one dimension, Lifshitz tail effects may be difficult to observe experimentally due to the long times at which they occur. There are some indications that the one-dimensional result $P(t) \sim \exp(-At^{1/3})$ has been inferred from fluorescence[9] and conductivity[51] measurements, but it generally remains true that most high-temperature exciton trapping data have been interpreted using theoretical results obtained from approximate treatments of the Green's function, or upon truncated cumulant expansions—approaches which are valid only over finite times[15–32] and which therefore fail to predict the correct asymptotic behavior. The development of a unified analytical approach to study the *crossover* between these short-time theories and the long-time Lifshitz-tail behavior remains a pressing theoretical problem.

Another interesting question which has attracted some recent attention, however, is how the asymptotic behavior of trapping dynamics is altered when particle motion is not diffusive. Examples of situations in which such questions become important include strongly disordered systems in which anomalous or subdiffusive transport occurs, in classical systems where noninteracting particles move ballistically (or generally with an enhanced or superdiffusive mechanism), or at low temperatures in condensed phases when wave motion obtains due to quantum mechanical effects.

Regarding this latter possibility, there are reasons to believe that quantum mechanical effects play an increasingly important role in exciton trapping experiments conducted at very low temperatures.[44–50] In this limit, the inelastic mean free path of the particle can exceed the average distance between traps, and the standard trapping model (which assumes motion to be diffusive over smaller length scales) no longer applies. Electron spin-echo measurements[56] suggest that such a situation may, in fact, describe low-temperature (~ 2 K) exciton trapping measurements on a number of quasi-one-dimensional molecular solids.[7,8,31,32] A difficulty preventing the universal acceptance of this interpretation is that the low-temperature (coherent) limit of the trapping problem is less well-understood than the diffusive limit. In a set of early analyses, Pearlstein and co-workers[44] obtained a number of exact results characterizing the trapping rate at zero temperature in one-dimensional tight-binding chains containing absorptive traps. Subsequent work by Kenkre,[45] using the Boltzmann equation, and Huber,[46] using multiple-scattering approaches to the Green's function, considered the trapping of coherent Bloch-type excitations in the presence of a finite concentration of randomly distributed traps. The use of generalized master equations by Kenkre and co-workers led to an approach to the quantum trapping problem involving the evolution of probabilities in real space similar to those that had been used for the case of diffusive motion.[28-30]

The validity of these earlier approaches has been difficult to ascertain until recently due to the realtive dearth of numerical approaches for studying the problem. Standard numerical methods[19 20,22] that have been developed to study diffusion-limited trapping are often not directly applicable because they are based upon quantities such as the number of distinct sites visited by a particle—a quantity dependent upon the particle's path and therefore not quantum mechanically well-defined. Experience with the diffusive problem suggests, however, that the kind of analytical approaches adopted earlier will give a reasonable description of the survival probability at short times, but will remain insensitive to the true asymptotic behavior of the survival probability and, specifically, to any Lifshitz-tail effects which might arise at long times. It is on this limit, i.e., that in which the motion of the mobile species is described by a purely wavelike evolution, and in particular on the associated Lifshitz tail effects, that we focus here.

2. MODELS

The starting point of most previous analysis of the low-temperature problem has been with evolution equations that are equivalent to a Liouville–von Neumann equation

$$d\rho/dt = -i[H, \rho] \tag{1}$$

for the reduced single-particle density matrix $\rho(t)$, in which the effective (but non-Hermitian) Hamiltonian, written in the basis of site-localized states $|n\rangle$,

$$H = \sum_{n,m} J_{n,m}(|n\rangle\langle m| + |m\rangle\langle n|) - \sum_{n} |n\rangle\, iV_n \langle n| \tag{2}$$

contains terms which assign to the trap-coupled sites an imaginary component of the energy, thus giving them a finite lifetime arising from the coupling to trap molecules. In this equation, the quantity V_n describes the decay of amplitude from site n due to the coupling to the trap states. The exact form that it takes depends on the nature of the traps which have been doped into the material. Two different models have been studied.[44-49] Substitutional traps occupy normal sites of the lattice, and probability amplitude is assumed to decay into them from the $2d$ nearest-neighbor lattice sites. Interstitial traps are assumed to reside in the interstices, and amplitude decays into them from, e.g., the nearest lattice site. The distinction is of qualitative importance, however, only at zero temperature and in one-dimensional systems with strictly nearest-neighbor transfer matrix elements between lattice sites. In this limit, particles created in a linear chain terminated on each end by substitutional traps can only migrate within the chain or decay to the terminating traps. The dynamical problem then separates into that of: (1) obtaining the solution for each cluster of fixed size N, and then (2) statistically weighting the solutions so obtained by the distribution for clusters of a given size. Thus, as pointed out by Pearlstein and Hemenger,[44] the survival probability can be written

$$P(t) = \sum_N Nq^2(1-q)^{N-1}\, P_N(t) \tag{3}$$

The decay from each cluster will be bounded from below by that for the most slowly decaying eigenstate in each. By analyzing such states, a lower bound for the survival probability can therefore be obtained.[47] Thus, attention is focused on the eigenstates and complex eigenvalues of the non-Hermitian Hamiltonian for a cluster of length N terminated at each end by sites with an imaginary site energy iV. As shown in ref. 47, the eigenstates of such a cluster take the form

$$|k\rangle = \sum_{n=1}^{N} [A_k \cos(kn) + B_k \sin(kn)]\, |n\rangle \tag{4}$$

where the allowed wavevectors satisfy the equation

$$\tan(kN) = \frac{(V^2 + J^2)\sin k}{2iJV - (J^2 - V^2)\cos k} \tag{5}$$

From the solutions k of this equation the real and imaginary components of the energy follow from the formal solution to the eigenvalue equation $E_k = 2J\cos(k) = \varepsilon_k - i\Gamma_k$. Figure 1 contains a plot of the eigenvalues for trap-terminated chains of different length. Notice that it is the long-

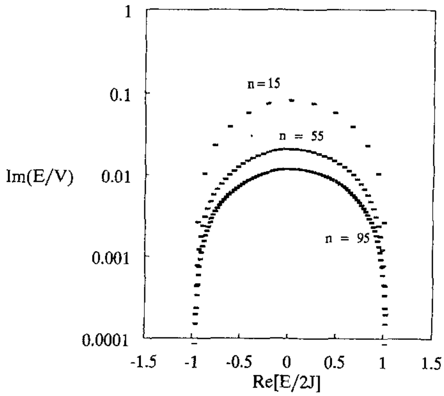

Fig. 1. Eigenvalues for coherent exciton chains of 15, 55, and 95 sites terminated at each end by imaginary site energies of magnitude $V = J$.

wavelength states near the band edges that have the smallest imaginary component of the energy, and thus the longest lifetime. Note also that corresponding states in longer chains live longer than ones in shorter chains. Analysis reveals that the longest-lived states in each chain have a decay amplitude given, asymptotically in N, by the expression[47]

$$\Gamma_N \sim \frac{4VJ^2\pi^2}{V^2+J^2}\frac{1}{N^3} \tag{6}$$

Thus the survival probability for each chain is bounded from below by an expression of the form $P_N(t) > f_N \exp(-2\Gamma_N t)$. By averaging this over the chain length distribution and performing a saddle point analysis, the result $P(t) \sim \exp(-At^{1/4})$ is obtained with $p = (1-q)$ and[47]

$$A = \frac{8 \ln^{3/4}(1/p)}{3}\left(\frac{3VJ^2\pi^2}{2(V^2+J^2)}\right)^{1/4} \tag{7}$$

In the *interstitial* model in any dimension the problem does not divide neatly up into clusters in this fashion.[48,49] The survival probability continues to depend, however, on the eigenstates and complex eigenvalues of the effective Hamiltonian H. Note that the form of H is similar in this limit to that of a tight-binding binary alloy with two types of site energies,[54,55] one of which is strictly imaginary. This has interesting consequences. With *real* site-energy defects, all eigenvalues lie along the real axis. Thus, although the analytic continuation of the averaged Green's function $g^+(z) = \langle (z - H + i0^+)^{-1} \rangle$ can have singularities off the axis (see, e.g., the discussion of Lloyd's model in refs. 54 and 55), the eigenvalue density remains concentrated on the real axis. With random *imaginary* site energies the eigenstates acquire a lifetime, and the eigenvalues themselves move off the axis. This results in an eigenvalue density $\rho(z)$ defined at arbitrary points of the complex plane. We note in passing that it is straightforward to generalize the standard expression for obtaining the density of real energy states from the Green's function to the situation in which the density is distributed over a region of the complex plane. We find, e.g., with $z = \varepsilon - i\Gamma$, that

$$\rho(\varepsilon, \Gamma) = \mathrm{Re}\,\frac{\partial G}{\partial \varepsilon} - \mathrm{Im}\,\frac{\partial G}{\partial \Gamma} \tag{8}$$

where $G = \mathrm{Tr}(g)$. At any case, the asymptotic results that have been obtained thus far for the interstitial model are necessarily less rigorous than for the one-dimensional substitutional model. Nonetheless, it can be argued that, as in the diffusive problem, the long-lived states are those centered in statistically-rare trap-free regions of the medium surrounded by regions of more typical trap density. As in the substitutional model in one dimension, the longest-lived states in these regions are those states of asymptotically long wavelength which decay quickly into the region outside the void where trapping can occur. In ref. 49 an estimate of the asymptotic decay of $P(t)$ was given through a calculation which focused on the average behavior of a transport particle centered in such a trap-free void and which treated the region outside the void using a constant absorptive potential calculated within the virtual crystal approximation (VCA). In this approximation

$$\begin{aligned} V_n \to \langle V_n \rangle &= -iqV \qquad \text{for} \quad |n| > R \\ &\to 0 \qquad\qquad \text{for} \quad |n| < R \end{aligned} \tag{9}$$

By considering small-wavevector (continuum) solutions of the Schrödinger equation, it was shown that the most slowly decaying modes associated with such a potential have eigenvalues, given asymptotically in R, by the expression $E = Jk^2$ with

$$k = k_0 \left(1 - \frac{J \exp(i\pi/4)}{qVR^2} \right) \tag{10}$$

Hence, it is found that

$$E \sim Jk_0^2 - i\,\frac{x_0^2}{R^3} \left(\frac{2J^3}{qV} \right)^{1/2} \tag{11}$$

where $x_0 = Rk_0$ is the first root of the lowest d-dimensional solution to the radial Helmholtz equation. It is reasonable to expect, then, that the decay of particles created in trap-free voids of radius R will be bounded from below by an expression of the form

$$P_R > f(R) \exp(-2\Gamma_R t) \tag{12}$$

where Γ_R can be estimated from the imaginary part of Eq. (11), i.e.,

$$\Gamma_R \sim x_0^2 \left(\frac{2J^3}{qV}\right)^{1/2} R^{-3} \tag{13}$$

By averaging the decay associated with this rate over the size R of the trap-free region in which the particle initially finds itself, we then obtain an estimate for $P(t)$, namely

$$\begin{aligned} P(t) &\sim \int dR\, f(R) \exp(-\alpha R^d - \beta t R^{-3}) \\ &\sim \exp[-At^{d/(d+3)}] \end{aligned} \tag{14}$$

Within this simple phenomenological argument, then, we find the following results as a function of dimensionality:

1D: $\quad P(t) \sim \exp(-At^{1/4})$

$$A = \frac{8 \ln^{3/4}(1/p)}{3} \left(\frac{3\pi^2}{2}\right)^{1/4} \left(\frac{J^3}{qV}\right)^{1/8} \tag{15}$$

2D: $\quad P(t) \sim \exp(-At^{2/5})$

$$A = \frac{5}{2}\left(\frac{2\pi \ln(1/p)}{3}\right)^{3/5} \left(\frac{x_0^4 J}{2qV}\right)^{1/5} \tag{16}$$

3D: $\quad P(t) \sim \exp(-At^{1/2})$

$$A = \left[16\pi^2 \ln\left(\frac{1}{p}\right)\right]^{1/2} \left(\frac{J}{18qV}\right)^{1/4} \tag{17}$$

3. NUMERICAL RESULTS FOR ONE-DIMENSIONAL CHAINS

To test some of these ideas regarding the asymptotic dynamics of the coherent limit of the trapping problem—more specifically, regarding the validity of the approximate arguments which have been applied to the interstitial model—we have performed a series of numerical diagonalizations of the effective (non-Hermitian) Hamiltonian for one-dimensional chains containing a random distribution of interstital traps. In Fig. 2 we present a typical eigenvalue distribution for a chain of 800 total sites. A certain fraction of these sites were associated, with probability $q = 0.1$, with interstitial traps modeled using an imaginary component of the site energy of magnitude $V = J$. Notice the Lifshitz tails of states with small imaginary components (decay amplitudes) associated with real energies near the band edges. These states clearly represent the type which were the focus of the approximate treatment leading to Eqs. (15)–(17). In addition, there are a large number of states with decay amplitudes in the region of $0.1V$, which would correspond to states which are effectively delocalized throughout the chain and thus have $\langle \mathrm{Im}\, H \rangle \sim -qV$. There are also a smaller number of states with decay amplitudes very close to V, corresponding to states which are localized on clusters of trap states (so that $\langle \mathrm{Im}\, H \rangle \sim -V$). Such states could be important in a more complete theory which addresses the short to intermediate-time behavior that we have neglected. In ref. 48, we have shown how the distribution of the tails of the imaginary part of the eigenvalue distribution is correctly predicted by the approximate treatment presented above.

Turning to the time dependence, we show in Figs. 3 and 4 numerical calculations of $P(t)$ based upon the numerical diagonalizations of 800-site random chains of the type whose eigenvalue spectrum is displayed in Fig. 2. In Fig. 3 we present the survival probability for six separate chains, using an initial condition in which the probability of any eigenstate of the chain being initially occupied is equal to $1/N$, N being the number of sites in the chain. Each curve, therefore, represents the quantity $\langle \exp(-\Gamma_i t) \rangle$,

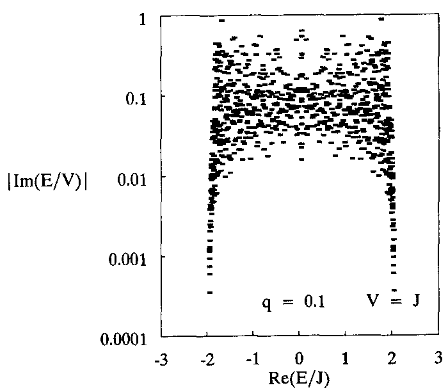

Fig. 2. Eigenvalues for an 800-site chain with a concentration $q = 0.1$ of imaginary site energies of magnitude $V = J$.

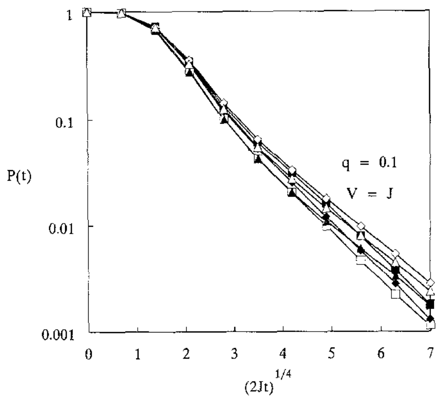

Fig. 3. Survival probability for six 800-site chains containing a concentration $q = 0.1$ of imaginary site energies of magnitude $V = J$, for an initial condition corresponding to equal population of all eigenstates in the chain.

averaged over all eigenstates of the corresponding chain. Note the significant spread in the curves for these six chains showing how, even for chains of this length, the survival probability is not totally self-averaging, but rather is subject to significant fluctuations which depend upon the distribution of trap-free regions in each chain. In Fig. 4 we present the average of these curves with statistical error bars indicating the average deviation of the mean about the corresponding average values. The curve is plotted as a function of $t^{1/4}$ to facilitate the identification of the nearly linear behavior of the logarithmic plot, in agreement with the predictions of Eq. (15). It is

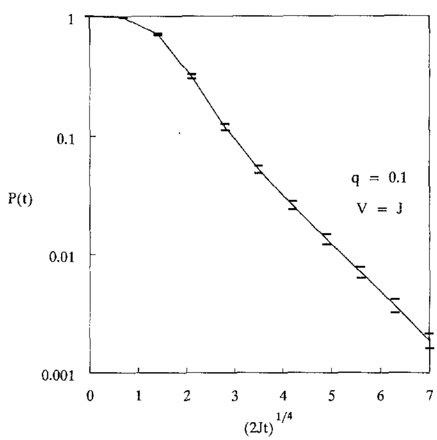

Fig. 4. Average value of the six curves in Fig. 3 with error bars indicating the standard deviation of the mean, showing the stretched-exponential behavior predicted by Eq. (15).

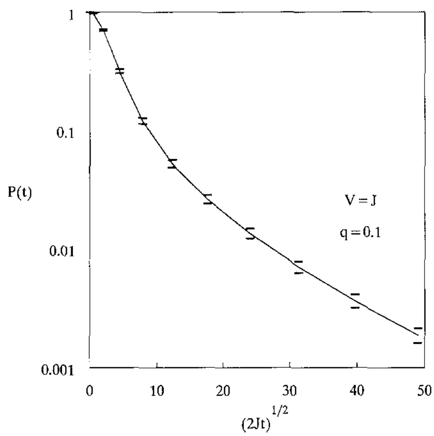

Fig. 5. Average value of the six curves in Fig. 3 with error bars indicating the standard deviation of the mean, plotted as a function of $t^{1/2}$, to be compared with Fig. 4.

interesting to note that this behavior does not occur at inordinately long times. We have also plotted in Fig. 5 the same data as a function of $t^{1/2}$, a form which has appeared in recent numerical work of Huber and Ching on a similar problem. The asymptotic region in the $t^{1/2}$ plot deviates more from linearity than that which appears in Fig. 4. We note, however, that the initial condition of Huber and Ching, corresponding to the initial population of a $k = 0$ mode of the ordered chain, is significantly different than what we have assumed here and thus the two results are not necessarily contradictory, particularly at these relatively short times.

In summary, we have investigated the long-time, Lifshitz-tail behavior of the survival probability for the quantum mechanical counterpart to the standard trapping model. We have found it to be of a stretched-exponential form which is slower in any dimension than in the corresponding diffusive problem. As in earlier treatments of the problem, we have found that spectral analyses of the eigenvalue distribution provide a useful way of obtaining insight into the dynamics of the underlying physical problem. It is hoped that these techniques will prove of similar value in elucidating the short to intermediate-time behavior of the trapping kinetics in very low-temperature condensed phases.

ACKNOWLEDGMENTS

I would like to thank the authors of ref. 59 for making copies of their work available prior to publication, and K. Lindenberg for bringing to my attention the important early work on this problem reported in refs. 44.

REFERENCES

1. H. C. Wolf, in *Advances in Atomic and Molecular Physics*, Vol. 3, D. R. Bates and I. Estermann, eds. (Academic Press, New York, 1976); R. C. Powell and Z. Soos, *J. Lumin.* **11**:1 (1975).
2. H. Auweter, A. Braun, U. Mayer, and D. Schmid, *Z. Naturforsch.* **34A**:761 (1979); H. Auweter, U. Mayer, and D. Schmid, *Z. Naturforsch.* **33A**:651 (1978).
3. A. Braun, U. Mayer, H. Auweter, H. C. Wolf, and D. Schmid, *Z. Naturforsch.* **37A**:1013 (1982).
4. A. H. Francis and R. Kopelman, in *Excitation Dynamics in Molecular Solids*, W. M. Yen and P. M. Selzer, eds. (Springer-Verlag, Berlin, 1981).
5. R. Kopelman, in *Spectroscopy and Excitation Dynamics of Condensed Molecular Systems*, V. M. Agranovich and R. M. Hochstrasser, eds. (North-Holland, Amsterdam, 1983).
6. R. Kopelman, *J. Stat. Phys.* **42**:185 (1986).
7. D. D. Dlott, M. D. Fayer, and R. D. Wieting, *J. Chem. Phys.* **67**:3808 (1977).
8. D. D. Dlott, M. D. Fayer, and R. D. Wieting, *J. Chem. Phys.* **69**:2752 (1978).
9. R. A. Auerbach and G. L. McPherson, *Phys. Rev. B* **33**:6815 (1986).
10. J. R. G. Thorne, R. G. Denning, T. J. Barker, and D. I. Grimley, *J. Phys.* (Paris) **C7**:125 (1985).
11. J. Cibert, M. C. Terrile, and Y. Merle d'Aubigne, *J. Phys.* (Paris) **C7**:135 (1985).
12. U. Even, K. Rademann, J. Jortner, N. Manor, and R. Reisfeld, *Phys. Rev. Lett.* **58**:285 (1987); **42**:2164 (1984).
13. W. D. Dozier, J. M. Drake, and J. Klafter, *Phys. Rev. Lett.* **56**:197 (1986).
14. T. Kirski, J. Grimm, and C. von Borczyskowski, *J. Chem. Phys.* **87**:2062 (1987).
15. E. W. Montroll, *Proc. Symp. Appl. Math. Am. Soc.* **16**:193 (1964); E. W. Montroll and G. H. Weiss, *J. Math. Phys.* **6**:167 (1965).
16. G. H. Weiss, *Proc. Natl. Acad. Sci. USA* **77**:4391 (1980); G. H. Weiss and R. J. Rubin, *Adv. Chem. Phys.* **52**:363 (1983).
17. H. B. Rosenstock, *Phys. Rev.* **187**:1166 (1969); *J. Math. Phys.* **21**:1643 (1980).
18. S. Smoluchowski, *Ann. Phys.* (Leipzig) **48**:1103 (1915).
19. G. Zumofen and A. Blumen, *Chem. Phys. Lett.* **88**:63 (1982); A. Blumen and G. Zumofen, *Chem. Phys. Lett.* **70**:387 (1980).
20. J. Klafter, G. Zumofen, and A. Blumen, *J. Phys. Lett.* (Paris) **45**:L49 (1984).
21. J. Klafter and R. Silbey, *J. Chem. Phys.* **72**:849 (1979); **74**:3510 (1981).
22. S. Havlin, B. Trus, G. H. Weiss, and D. J. Ben-Avraham, *J. Phys. A* **18**:L247 (1985); S. Havlin, M. Dishon, J. E. Kiefer, and G. H. Weiss, *Phys. Rev. Lett.* **53**:407 (1984).
23. D. L. Huber, in *Excitation Dynamics in Molecular Solids*, W. M. Yen and P. M. Selzer, eds. (Springer-Verlag, Berlin, 1981); *Phys. Rev. B* **20**:2307, 5333 (1979).
24. D. L. Huber, *Phys. Rev. B* **20**:2307 (1979).
25. D. L. Huber, *Phys. Rev. B* **20**:5333 (1979).
26. D. L. Huber, *J. Chem. Phys.* **75**:4749 (1981); K. K. Gosh and D. L. Huber, *Phys. Rev. B* **23**:4441 (1981).
27. K. Lakatos-Lindenberg, R. P. Hemenger, and R. M. Pearlstein, *J. Chem. Phys.* **56**:4852 (1972).
28. V. M. Kenkre and Y. M. Wong, *Phys. Rev. B* **22**:5716 (1980); **23**:3748 (1981).
29. V. M. Kenkre and P. E. Parris, *Phys. Rev. B* **27**:3221 (1983).
30. V. M. Kenkre, in *Exciton Dynamics in Molecular Crystals and Aggregates*, K. G. Hohler, ed. (Springer, Berlin, 1982).
31. V. M. Kenkre, P. E. Parris, and D. Schmid, *Phys. Rev. B* **32**:4946 (1985).
32. P. E. Parris and V. M. Kenkre, *Chem. Phys. Lett.* **107**:413 (1984); P. E. Parris and V. M. Kenkre, *J. Lumin.* **31**:612 (1984).

33. B. Movaghar, *J. Phys. C: Solid State* **13**:4915 (1980); B. Movaghar, G. W. Sauer, and D. Wurtz, *J. Stat. Phys.* **27**:473 (1982).
34. H. Scher and C. H. Wu, *Proc. Natl. Acad. Sci. USA* **78**:22 (1980); *J. Chem. Phys.* **74**:5366 (1981).
35. R. Grassberger and I. Procaccia, *J. Chem. Phys.* **77**:6281 (1981).
36. J. Anlauf, *Phys. Rev. Lett.* **52**:1845 (1984).
37. F. Kayser and J. B. Hubbard, *Phys. Rev. Lett.* **51**:79 (1983).
38. P. E. Parris, *Phys. Lett. A* **114**:250 (1986); P. E. Parris, V. M. Kenkre, and S. M. Phatak, *J. Stat. Phys.* **35**:749 (1986); V. M. Kenkre, P. E. Parris, and S. M. Phatak, *Physica* **128A**:571 (1984).
39. T. Lubensky, *Phys. Rev. A* **30**:1845 (1984).
40. B. Ya. Balagurov and V. G. Vaks, *Zh. Eksp. Teor. Fiz.* **65**:1939 (1973) [*Sov. Phys. JETP* **38**:968 (1974)].
41. M. D. Donsker and S. R. S. Varadhan, *Commun. Pure Appl. Math.* **28**:525 (1975).
42. M. Bramson and J. L. Lebowitz, *Phys. Rev. Lett.* **61**:2397 (1988).
43. Th. M. Nieuwenhuizen, *Phys. Rev. Lett.* **62**:357 (1989).
44. K. Lakatos-Lindenberg, R. P. Hemenger, and R. M. Pearlstein, *J. Chem. Phys.* **56**:4852 (1972); R. P. Hemenger, K. Lakatos-Lindenberg, and R. M. Pearlstein, *J. Chem. Phys.* **60**:3271 (1974); R. M. Pearlstein, *J. Chem. Phys.* **56**:4852 (1972); R. P. Hemenger and R. M. Pearlstein, *Chem. Phys.* **2**:424 (1973).
45. V. M. Kenkre, *Phys. Status Solidi B* **89**:651 (1978).
46. D. L. Huber, *Phys. Rev. B* **22**:1714 (1980); **24**:1083 (1981).
47. P. E. Parris, *Phys. Rev. Lett.* **62**:1392 (1989).
48. P. E. Parris, *Phys. Rev. B* **40**:4928 (1989).
49. J. W. Edwards and P. E. Parris, *Phys. Rev. B* **40**:8045 (1989); *J. Lumin.* **45**:279 (1990).
50. H. Benk and R. Silbey, *J. Chem. Phys.* **79**:3487 (1983).
51. U. Seiferheld, H. Bassler, and B. Movaghar, *Phys. Rev. Lett.* **51**:813 (1983); I. G. Hunt, D. Bloor, and B. Movaghar, *J. Phys. C* **18**:3497 (1985).
52. I. M. Lifshitz, *Adv. Phys.* **13**:483 (1964).
53. J. Luttinger and R. Tao, *Ann. Phys.* (N.Y.) **145**:185 (1983); R. Freidberg and J. M. Luttinger, *Phys. Rev. B* **12**:4460 (1975).
54. R. J. Elliot, J. A. Krumhansl, and P. L. Leath, *Rev. Mod. Phys.* **46**:465 (1974).
55. E. N. Economou, *Green's Functions in Quantum Physics* (Springer-Verlag, New York, 1979).
56. A. J. Van Strien, J. F. C. van Kooten, and J. Schmidt, *Chem. Phys. Lett.* **76**:7 (1980); A. J. Van Strien, J. Schmidt, and R. Silbey, *Mol. Phys.* **46**:151 (1982).
57. J. W. Haus and K. W. Kehr, *Phys. Rep.* **150**:265 (1987).
58. P. E. Parris, unpublished results.
59. D. L. Huber and W. Y. Ching, presented at the 7th Conference on Dynamical Processes in Excited States of Solids, Athens, Georgia (August 1989), and unpublished results.