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Self-Consistent Mode-Coupling Theory of Excitation Transport in Disordered Media: Application to Anderson Localization and Incoherent Transport

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ABSTRACT

We present a novel mode-coupling procedure for constructing self-consistent equations for excitation transport in disordered systems. The present approach allows incoherent hopping models and fully quantum mechanical models, including those expected to display Anderson localization, to be treated in a unified way. This procedure applies both to short-range (e.g. nearest neighbor) and to long-range (e.g. dipole-dipole) interactions. Our formulation is based on self-consistent equations for $P_o(\varepsilon)$ and $D(\vec{k},\varepsilon)$. $P_o(\varepsilon)$ is the Laplace transform of $P_o(t)$, the conditional probability that an excitation that occupies a given spatial position at a given time will be found at that position after a time t has elapsed. $D(\vec{k},\varepsilon)$ is the frequency and wavevector dependent diffusion coefficient. The mode-coupling procedure is applied to an incoherent hopping model and to quantum percolation. For the hopping problem, our procedure correctly reproduces the long-time power law decay of $P_o(t)$. For quantum percolation, we derive an equation that is similar in structure to the equation obtained by Vollhardt and Wölfle for a Fermi gas in a random potential. The frequency dependence of the ac conductivity and the critical exponents determined from this equation agree with the predictions of scaling theory.

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functional of $P_o(\epsilon)$, resulting in a second relation between these two quantities. When the second relation is substituted into the first, we obtain a closed self-consistent equation for $P_o(\epsilon)$. This equation contains an integration over the wavevector $\vec{k}$ that is conjugate to the position variable. This type of equation is characteristic of mode-coupling theories of nonlinear hydrodynamics, in which self-consistent equations are derived that contain similar integrations over wavevector[21-23]. An important feature of the mode-coupling theories is their capacity to correctly predict the long-time asymptotic power-law decays of hydrodynamic variables ("long-time tails"). In a similar fashion, the approach presented here can be used to generate approximations to $P_o(t)$ that reproduce the correct long-time power-law behavior of that quantity, namely $P_o(t) \sim t^{-d/2}$ in d dimensions[1,3,4,7]. The major advantage of the present formulation is its capacity to treat both quantum and incoherent transport with short-range or long-range interactions within the same theoretical framework.

In Sec. II, we present a general formulation of our mode-coupling approach. In Sec. III, this methodology is applied to the incoherent hopping problem on a substitutionally disordered lattice. In Sec. IV, we apply the mode-coupling theory to quantum percolation. We derive a self-consistent equation for the diffusion kernel that is similar in structure to the Vollhardt-Wölfle equation for a Fermi gas in a random potential[18,19]. The results of Sec. III for incoherent transport and the results of Sec. IV for quantum percolation are compared. Section V contains a comparison to previous work.

II. The Mode-Coupling Procedure

In this section, we present the self-consistent procedure for calculating transport properties of dilute excitations (e.g., electrons or Frenkel excitons) on disordered lattices. We shall consider a general class of models involving the motion of a single excitation in a disordered system whose ensemble average is

determined for a general class of models of a disordered lattice. In these models, which will be considered in detail in the subsequent sections, a fraction c of the lattice sites participates in the excitation transport, and the remaining sites are blocked. In the Appendix, we demonstrate that $\tilde{D}$ can be obtained order-by-order in c from the naive density expansions of D and P_o, which are the only microscopic input to the theory[9]. (The naive (unrenormalized) density expansions of these quantities are defined in Eqs.(A2) and (A3).) Substitution of Eq.(4) into Eq.(3) yields

$$P_o(\varepsilon) = \Omega^{-1} \int d\vec{k} \, [\varepsilon + k^2 \tilde{D}(\vec{k},P_o)]^{-1}. \tag{5}$$

We propose the following self-consistent procedure for calculating $P(\vec{k},\varepsilon)$. (i) Approximate the functional $\tilde{D}(\vec{k},P_o)$. (ii) Substitute this expression into the right side of Eq.(5), and solve Eq.(5) for $P_o(\varepsilon)$. (iii) Substitute this expression for $P_o(\varepsilon)$ into the approximate functional $\tilde{D}(\vec{k},P_o)$ to generate $D(\vec{k},\varepsilon)$, (Eq.(4)). This expression for $D(\vec{k},\varepsilon)$, when substituted into the right side of Eq.(2), yields $P(\vec{k},\varepsilon)$.

III. Application to Incoherent Transport: The Master Equation

We consider a lattice with a fraction c of the sites occupied by active particles that are capable of retaining and transferring an excitation. These particles are assumed to be randomly distributed among the lattice sites. The unoccupied sites do not participate in the transport. The dynamics of a particular realization of the random system are governed by a Pauli master equation.

$$\frac{d\vec{p}}{dt} = \underset{\approx}{W} \cdot \vec{p} \tag{6}$$

$\vec{p}$ is an N component vector whose i'th element is the probability that an excitation resides on the particle labelled i. N is the number of active particles. W is an NxN matrix whose elements are given by

$$\tilde{D}^{(n)}(\vec{k},P_o) = \sum_{j=1}^{n} c^j \tilde{D}_j(\vec{k},P_o) \qquad (10)$$

Equation (10) can be substituted into Eq.(5) to yield a self-consistent equation for P_o. We can improve upon approximations of the form given in Eq.(10) by noting that at c=1 (no disorder), D can be calculated exactly, since the system is translationally invariant. In this case, D is independent of ϵ, and hence $\tilde{D}(\vec{k},P_o)$ at c=1 does not depend on $P_o(\epsilon)$. Denoting $\tilde{D}$ at c=1 by $\tilde{D}^*$, we have

$$\tilde{D}^*(\vec{k},P_o) = k^{-2} \sum_{\vec{r}} w(\vec{r}) [1-\exp(i\vec{k}\cdot\vec{r})]. \qquad (11)$$

We now have two pieces of information regarding $\tilde{D}$: a truncated expansion in c (Eq.(10)) that will be accurate for $c < 1$ and the exact form at c=1 (Eq.(11)). By constructing an approximation to $\tilde{D}$ that interpolates between Eqs.(10) and (11), we guarantee that the self-consistent equation will give exact results at c=1. A hierarchy of such approximations can be derived by constructing Padé approximants in the variable $\gamma \equiv c/(1-c)$:

$$\tilde{D}(\vec{k},P_o) = \frac{a_1\gamma+a_2\gamma^2+\cdots a_j\gamma^j}{1+b_1\gamma+b_2\gamma^2+\cdots b_j\gamma^j} \; . \qquad (12)$$

The coefficients a_n and b_n in Eq.(12) are obtained by requiring that Eq.(12) reduce to Eq.(10) in the limit $c\to 0$ and to Eq.(11) at c=1. The first member of the hierarchy is

$$\tilde{D}(k,P_o) = \frac{\gamma\tilde{D}_1(\vec{k},P_o)}{1+\gamma\tilde{D}_1(\vec{k},P_o)/\tilde{D}^*(\vec{k},P_o)} \qquad (13)$$

$$P_o(\varepsilon) = \Omega^{-1} \int d\vec{k} [\varepsilon + k^2 \tilde{D}(\vec{k}, P_o)]^{-1} \tag{15a}$$

$$\tilde{D}(\vec{k}, P_o) = \frac{k^{-2} 2cw(d - \sum_{j=1}^{d} \cos(\vec{k} \cdot \vec{a}_j))}{1 + 2(1-c) P_o w} . \tag{15b}$$

w is the magnitude of the transfer rate between nearest-neighbor sites on the lattice. $\vec{a}_j$ is the lattice vector in the jth direction. Equations (15) constitute our final self-consistent equations for P_o and $\tilde{D}$ for incoherent transport with nearest-neighbor interactions. They may be solved iteratively. The long-time behavior can be obtained analytically by the following procedure. We postulate that $P_o(\varepsilon) \sim A \varepsilon^{\alpha-1}$ as $\varepsilon \to 0$, where A is a function of c. If this form is substituted into Eqs.(15), A and α can be determined analytically. The Tauberian theorem for Laplace transforms can then be used to infer that $P_o(t) \sim A t^{-\alpha}$ for $t \to \infty$[27]. With this procedure, we obtain the following results. For a cubic lattice (d=3) there exists a critical concentration c*, such that for $c^* < c \leq 1$, transport becomes diffusive in the limit of long times and large distances, and $P_o(t)$ decays as $t^{-3/2}$. This power law decay of $P_o(t)$ agrees with scaling arguments, which predict that $P_o(t)$ in d dimensions should decay as $t^{-d/2}$ at long times[3,4,7]. c* is predicted to be ~ 0.325, which agrees well with the currently accepted numerical estimate of 0.310 for the site percolation threshold on a simple cubic lattice[28]. The diffusion constant D(0,0), which is defined to be the $\vec{k} \to 0$ and $\varepsilon \to 0$ limit of $D(\vec{k}, \varepsilon)$ vanishes as c approaches c* from above as $(c-c^*)^s$ with s=1. This "mean-field" value for the critical exponent s has also been obtained in effective medium theories of dynamical percolation[10,11]. This exponent has been estimated from Monte Carlo calculations to be 1.75 ± 0.1[29]. For $c < c^*$, $P_o(t)$ decays to a finite value, which indicates that the excitations are localized on clusters of finite size.

particles. For a given configuration of the random system, the dynamics are governed by the tight-binding Hamiltonian

$$H = \sum_{i<j} \hbar J_{ij}(\vec{r}_{ij}) [|i><j| + |j><i|]. \tag{19}$$

The indices i and j in Eq.(19) label the active particles. $P(\vec{r},t)$, the configuration-averaged probability that an excitation propagates a distance $\vec{r}$ in time t, is given by

$$P(\vec{r},t) = (N-1) < [\exp(-iLt)]_{22,11} \delta_{\vec{r},\vec{r}_{12}} > + \delta_{\vec{r},\vec{0}} < [\exp(-iLt)]_{11,11} >. \tag{20}$$

Equation (20) is the fully quantum mechanical analog of Eq.(8), which defines $P(\vec{r},t)$ for the master equation. L is the Liouville operator corresponding to the Hamiltonian of Eq.(19). (The action of L on an operator M is defined by LM = [H,M].) We have adopted tetradic notation[30], in which $[\exp(-iLt)]_{\ell\ell,mm}$ is the conditional probability that an excitation propagates from a particle at position $\vec{r}_m$ to a particle at position $\vec{r}_\ell$ in time t. The diffusion kernel is related to $P(\vec{r},t)$ by Eq.(2).

In order to apply the mode-coupling equation (Eq.(5)) to this model, we need to calculate the functional $\tilde{D}$. To that end, we expand $\tilde{D}(\vec{k},P_o)$ in a power series in c, as in Eq.(9). The coefficients $\tilde{D}_n(\vec{k},P_o)$ can be calculated according to the procedure set forth in the Appendix. (See Eq.(A7).) In our treatment of incoherent transport in Sec. III, we proposed a hierarchy of approximations to $\tilde{D}$ that are obtained by truncating Eq.(9) to yield Eq.(10) and then using a Padé approximant to interpolate between this expression and the exact result at c=1 (Eq.(11)). In the incoherent hopping model, the diffusion kernel assumes a simple, frequency-independent form at c=1, as shown in Eq.(11). Since the diffusion kernel is independent of ε, it is also independent of $P_o(\varepsilon)$, and we can immediately identify

excitation may still be localized. An intuitive picture of this situation is that a cluster may have infinite extent but may be sufficiently ramified (i.e., contains many holes) that it cannot be completely explored by a quantum excitation. In fact, it is found that an excitation in the quantum percolation model undergoes a transition from being localized at long time to being delocalized at long time at a critical concentration that lies above the usual percolation threshold[32-35]. We will refer to this concentration as the "quantum percolation threshold", although it should be noted that the usual percolation threshold is a static property of the lattice, while the quantum percolation threshold is a dynamical property of the model. The relationship between the quantum threshold and the static percolation threshold is particularly interesting in two dimensions. According to the scaling theories of Anderson localization, the quantum excitation should be localized in two dimensions in the presence of a finite amount of disorder[15-17]. The quantum percolation threshold, c*, should be unity. In contrast, it is well known that the static percolation threshold for two-dimensional lattices is less than unity[26].

Substituting a transfer matrix element $J(\vec{r})$ that has the value J for nearest neighbors on the lattice and is zero otherwise into Eqs.(21) and (22) and making use of Eq.(5), we obtain a self-consistent equation for quantum percolation with nearest neighbor interactions:

$$P_o(\varepsilon) = \Omega^{-1} \int d\vec{k}[\varepsilon + k^2 \tilde{D}(\vec{k},P_o)]^{-1} \tag{23a}$$

$$\tilde{D}(\vec{k},P_o) = \frac{4 k^{-2} c J^2 P_o}{1 + 4(JP_o)^2} [d - \sum_{j=1}^{d} \cos(\vec{k}\cdot\vec{a}_j)]. \tag{23b}$$

$\vec{a}_j$ is the lattice vector in the jth direction. $D(\vec{k},\varepsilon)$ can now be calculated self-consistently for the quantum transport problem from

$$\frac{D(0,\epsilon)}{D_o} = 1 - \left(\frac{2\,a^{d-2}}{c\,\pi^d}\right) \int_0^{k_o} \frac{k^{d-1}\,dk}{k^2 + \epsilon/D(0,\epsilon)} \tag{27}$$

$$D_o = c\,a^2/[2\bar{P}_o(0)]. \tag{28}$$

In Eq.(27), the $\epsilon \to 0$ limit of $\bar{P}_o(\epsilon)$, the non-critical contribution to $P_o(\epsilon)$, is assumed to be finite in the vicinity of a critical point. We treat D_o as an additional input to the calculation. It can be estimated with a theory that is valid at concentrations greater than and far from the critical point.

Equation (27) is our final self-consistent equation for quantum percolation with nearest-neighbor interactions. It is similar in structure to the equation derived by Vollhardt and Wölfle (VW) for a gas of noninteracting fermions at T = 0 in the presence of randomly placed static scatterers[18-20]. The VW equation can be obtained from Eq.(27) by replacing the factor $2a^{d-2}/(c\pi^d)$ with $d\lambda k_F^{2-d}$, where k_F is the Fermi wavevector, and λ is a dimensionless parameter specifying the degree of disorder. Since the blocked sites in the quantum percolation model can be regarded as randomly placed static scatterers, the types of disorder in the two models are very similar. However, there are fundamental differences between the two approaches. First, in the present work we consider the dynamics of a single quantum particle, while the resummation procedure used by VW to derive their self-consistent equation is closely connected to the many-body properties of the Fermi gas. The present treatment can be applied to a variety of physical problems that are not described by the model of VW, such as the motion of dilute electronic or vibrational excitations[36]. Second, the starting point for the VW resummation procedure is a perturbation expansion in scattering events, in which the zeroth order problem is the delocalized Fermi gas. Our resummation procedure is based on an expansion in c, the concentration of conducting sites, and hence, the zeroth order problem in the present treatment is a localized electron. Finally, in the VW

The frequency dependence of the ac conductivity calculated from Eq.(27) agrees with the predictions of scaling theories, as discussed by Shapiro[20]. The present mode-coupling calculation, which is based on Eq.(24), can thus be used to calculate critical exponents. It does not yield numerical values for the critical concentration c*. This is not surprising, since c* depends on the lattice geometry as well as the dimensionality, unlike the critical exponents which are universal and are expected to depend only on the dimensionality. In Eq.(24) we focus on long wavelength properties which will be sensitive to the dimensionality, but not the lattice type.

We shall now comment on the significance of the partitioning introduced in Eq.(24). A self-consistent equation for $P_o(\varepsilon)$, which does not involve the division of P_o into contributions from long and short wavelengths, is given by Eqs.(23), which yield the following results. In one and two dimensions, Eq.(29) holds, and the excitation is always localized. In three dimensions, a transition occurs at $c^* \approx 0.48$. For $c<c^*$, Eq.(31) holds with $\nu=1$. However, for $c>c^*$, $s=1/2$ in Eq.(32). According to the scaling theories, s should equal 1 if $\nu=1$. It is, therefore, clear that the modified self-consistent calculation based on Eqs.(24) and (25) is preferable to the calculation based on Eqs.(23). Both calculations are based on an approximation to $\tilde{D}$ (Eq.(23b)) that is valid for small values of c. There is no reason to expect such an approximation to work well at concentrations <u>above</u> the critical point. By introducing the cut-off wavevector k_o in Eq.(24), we introduce a scaling ansatz that holds in the vicinity of the critical point, both from above and from below.

In this work we have used the mode-coupling ideas of Sec. II to derive approximate self-consistent equations for incoherent transport (Eqs.(15)) and for fully quantum mechanical transport (Eq.(27)). Because of the complexity of the quantum problem, Eq.(27) was derived under more restrictive conditions than Eqs.(15). Equations (15) can be used to calculate $P(\vec{r},t)$ for all concentrations, times, and displacements in the incoherent hopping problem. Equations (5), (11), (13), and (14) can be used to perform calculations for any excitation

$$I_d(x) = \begin{cases} (2x)^{-1/2} & d=1 \\ (2\pi)^{-1}\ln(x^{-1}) & d=2 \\ I_3(0) - (4/\pi)(2x)^{1/2} & d=3. \end{cases} \qquad (37)$$

The constant $I_3(0)$ is 0.4822 to four digits[38]. In order to facilitate comparison between Eq.(27) (for the quantum problem) and Eq.(34) (for the master equation), we consider the limiting behavior of the wavevector integral on the right side of Eq.(27) for small values of $a^2\varepsilon/D(0,\varepsilon)$.

$$K_d(x) \equiv \int_0^{q_o} \frac{q^{d-1}\, dq}{q^2 + x} . \qquad (38)$$

For small values of x,

$$K_d(x) = \begin{cases} (\pi/2)x^{-1/2} & d=1 \\ [\ln(x^{-1})]/2 & d=2 \\ q_o - (\pi/2)x^{1/2} & d=3 . \end{cases} \qquad (39)$$

For small values of x, $K_d(x)$ has the same x dependence as $I_d(x)$ in Eq.(37). Substitution of Eq.(39) into Eq.(27) and of Eq.(37) into Eq.(34) shows that the self-consistent equations for $D(0,\varepsilon)$ for the quantum and incoherent transport problems have the identical structure. For example, consider Eq.(34) in three dimensions. From Eq.(37), we have

$$\frac{D(0,\varepsilon)}{D_o} = 1 - \left(\frac{1-c^*}{1-c}\right)\left(\frac{c^*}{c}\right) + \left(\frac{(1-c)}{c}\right)\left(\frac{4}{\pi}\right)\left(\frac{\varepsilon}{D(0,\varepsilon)}\right)^{1/2} , \qquad (40)$$

where $c^* = I_3(0)/(1+I_3(0)) \approx 0.325$. Equation (40) should be compared with Eq.(30). The two equations have the same structure and yield the same critical exponents.

Another self-consistent resummation procedure for the master equation was developed by Gochanour, Andersen, and Fayer, who treated incoherent excitation transport among particles randomly distributed in a continuum[5]. Their approach was generalized to substitutionally disordered lattices by Loring, Andersen, and Fayer[8]. This formulation is based on a self-consistent equation for $P_o(\varepsilon)$ that was derived diagrammatically and has the capacity to treat both short-range and long-range interactions. Within this approach the predicted $D(\vec{k},\varepsilon)$ is not self-consistent with the predicted $P_o(\varepsilon)$, in that these quantities do not obey Eq.(3). A consequence of this feature is that the $P_o(t)$ predicted with this formulation decays exponentially at long times ~ $\exp(-\Gamma t)$, rather than as ~ $t^{-d/2}$ as predicted by scaling theory.[6,9,41]

The treatment of Anderson localization by Vollhardt and Wölfle is based on the derivation of self-consistent equations for $D(0,\varepsilon)$ and $M(\vec{k},\varepsilon)$, where $M(\vec{k},\varepsilon)$ is the current relaxation kernel[18]. They consider a Fermi gas in a random potential, and their method cannot be easily generalized to incoherent or quantum excitation transport arising from interactions of arbitrary distance dependence.

The present approach, like other mode-coupling procedures, can be formulated in the time domain rather than in the frequency domain, as in Eq.(5)[23,42]. A comparison of these procedures will be presented in a future work[43]. The present mode-coupling procedure is the only approach that has the capacity to treat both the Anderson localization and the incoherent transport problems with short-range or long-range interactions.

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APPENDIX

In this work, we have made the ansatz that the diffusion kernel could be expressed as a functional of P_o. For the models of Sections III and IV, this ansatz (Eq.(4)) takes the form

$$D(\vec{k},\varepsilon,c) = \tilde{D}\,(\vec{k},\, P_o(\varepsilon,c),c). \tag{A1}$$

In this Appendix we write P_o as $P_o(\varepsilon,c)$, D as $D(\vec{k},\varepsilon,c)$, and $\tilde{D}$ as $\tilde{D}(\vec{k},P_o,c)$ to emphasize the dependence of these quantities on concentration. Equation (A1) is a definition of the functional $\tilde{D}$. In this Appendix, we present a systematic prescription for evaluating $\tilde{D}$. We begin by expanding D and P_o in powers of c.

$$D(\vec{k},\varepsilon,c) = \sum_{j=1}^{\infty} c^j D_j\,(\vec{k},\varepsilon^{-1}) \tag{A2}$$

$$P_o(\varepsilon,c) = \varepsilon^{-1} + \sum_{j=1}^{\infty} c^j Q_j\,(\varepsilon^{-1})\,. \tag{A3}$$

D_j and Q_j can be calculated from the exact solution of the transport problem for j or fewer particles. We shall take $\{D_j\}$ and $\{Q_j\}$ as given and use them to construct an expression for $\tilde{D}$. $\tilde{D}(\vec{k},P_o,c)$ depends explicitly on c, as well as depending implicitly on c through the c dependence of P_o. We expand $\tilde{D}$ in a power series with respect to its explicit c dependence.

$$\tilde{D}(\vec{k},P_o,c) = \sum_{j=1}^{\infty} c^j \tilde{D}_j(\vec{k},P_o). \tag{A4}$$

The function $D_n(\vec{k},\varepsilon^{-1})$ in Eq.(A2) is given by

$$D_n(\vec{k},\varepsilon^{-1}) = \frac{1}{n!}\frac{\partial^n}{\partial c^n} D(\vec{k},\varepsilon,c)\Big|_{c=0}$$

$$= \frac{1}{n!}\frac{\partial^n}{\partial c^n} \tilde{D}(\vec{k},P_o,c)\Big|_{c=0}. \tag{A5}$$

The second equality in Eq.(A5) follows from the definition of $\tilde{D}$ in Eq.(A1). Substitution of Eq.(A4) into Eq.(A5) and application of Leibnitz' Rule for the nth derivative of a product yields

$$D_n(\vec{k},\varepsilon^{-1}) = \sum_{j=1}^{n} \frac{1}{(n-j)!}\frac{\partial^{n-j}}{\partial c^{n-j}} \tilde{D}_j(\vec{k},P_o)\Big|_{c=0}. \tag{A6}$$

An expression for $\tilde{D}_n(\vec{k},P_o)$ is obtained by solving Eq.(A6) for $\tilde{D}_n(\vec{k},\varepsilon^{-1})$ and replacing ε^{-1} with P_o.

$$\tilde{D}_n(\vec{k},P_o) = D_n(\vec{k},P_o) - \sum_{j=1}^{n-1} \frac{1}{(n-j)!}\frac{\partial^{n-j}}{\partial c^{n-j}} \tilde{D}_j(\vec{k},P_o(x,c))\Big|_{\substack{c=0\\x=P_o}} \tag{A7}$$

Equation (A7) relates $\tilde{D}_n$ to D_n and to $\{\tilde{D}_j\}$ with $j<n$. Using Eqs.(A2), (A3), and (A7), we can relate the unknown function $\tilde{D}_n$ to the known functions D_j and Q_j with $j<n$. In this manner, we may calculate each term in the series for $\tilde{D}$ in Eq.(A4). For illustrative purposes, we will calculate $\tilde{D}_n(\vec{k},P_o)$ explicitly for n=1 and n=2. For n=1, Eq.(A7) becomes

$$\tilde{D}_1(\vec{k},P_o) = D_1(\vec{k},P_o). \tag{A8}$$

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