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Correlation Effects on Hopping Transport in a Disordered Medium

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ABSTRACT

Correlated hopping transport through a disordered system is discussed in terms of a random walk model with memory correlations on a bond disordered lattice. Correlations will in general result in a difference between the transition rate to the previously occupied site and the rate for transitions to any other nearest neighbour site. Such a correlated process corresponds exactly to Fürth's model for a random walk with a finite memory. This paper establishes a first order master equation for Fürth's random walk on a bond disordered lattice. The equation is found to be equivalent to a symmetrized second order equation which was used previously as the starting point for an effective medium treatment.

Atomic diffusion processes in condensed systems are often described by lattice gas models. The dynamics in such systems is given by many particle master equations in which the transition rates depend on the local field of the instantaneous configuration. In the simplest case one allows only hard core repulsions for

the interactions between particles. The system can then be described as a gas of mutually selfavoiding random walkers on a lattice.

Many studies have investigated the dynamic properties of such lattice gases. For the case of hard core repulsions (i.e. the "noninteracting lattice gas") detailed information on the tracer diffusion constant has been obtained both analytically[1−3] and by simulation[4]. Recently[5] it was observed that the problem can also be approached via dynamic disorder theory[6,7]. This approach[5] is based on the idea that the tracer particle moves in a disordered background which changes its configuration as a function of time.

Dynamic and static correlations between the particles play an important role in actual materials. In superionic conductors, for example, such correlations arise from non-hard-core interactions (e.g. Coulomb forces) between particles, from lattice relaxation effects, or from cooperative motion of groups of particles[8,9].

Given the importance of such correlation effects in real materials it is of interest to incorporate them into the dynamic disorder approach to lattice gas models. This requires as a first step to study the effect of correlations in the presence of frozen disorder, i.e. when the disordered geometry does not change with time. Such a study has been carried out[10], and the results have been utilized in the dynamic disorder approach[5]. These investigations were based on the correlated random walk model of Fürth[11,12]. However in Ref. 10 the disorder was introduced into a symmetrized second order equation for Fürth's random walk and not directly into the first order formulation of the problem. This deficiency will be remedied in the present paper in order to make the model more transparent.

Let me begin by recalling Fürth's model for correlated random walks[11,12]. In this model the random walker has a finite memory. Instead of choosing between all possible directions for a jump with equal probabilities the walker chooses the new direction with a probability which depends on the direction of the previous hop.

In the simplest case, called the backward jump model, the walker has a transition rate w_b for returning to its previously visited site, and a transition rate w for transitions to all other nearest neighbour sites. In general correlations will give rise to a difference, $w \neq w_b$, between them. For example for Coulomb interactions one expects[13,14] $w_b > w$, because each transition disturbs the previous local equilibrium configuration. Thus the particle has a higher than average probability to return to its previous site at the next jump attempt.

To formulate the model mathematically it is convenient to choose a markovian description by suitably enlarging the state space[15]. In the present case the state of a particle is given by its "history" (i,j) which consists of the currently occupied site i and the previously occupied site j. The quantity of interest is then $P(i,j,t)$ the probability density to find the particle at site i at time t if it arrived at i via a transition from site j and if it started from some origin at time 0. On a regular lattice $P(i,j,t)$ obeys the following master equation[12]:

$$\frac{d}{dt}P(i,j,t) = w_b[P(j,i,t) - P(i,j,t)] + w\sum_{k \neq i}[P(j,k,t) - P(i,j,t)] \qquad (1)$$

The summation runs over all nearest neighbour sites k of site j except i.

Consider now the case of bond disorder on the same underlying regular lattice as before. Each bond has probablity p of being present, and probability $1 - p$ of being absent. The particle can only jump across those bonds which are present. Transitions across absent bonds have transition rate 0.

It is not straightforward to generalize Eq. (1) to this case. The reason is that in the formulation of Eq. (1) it is implicitly assumed that the length of memory is given by the length of the history, and thus the walker has a memory of roughly length 1 in units of $\frac{1}{w}$. For the disordered lattice difficulties arise from the presence of dangling ends containing sites with coordination number $z_i = 1$. In the limit $w_b \to 0$ the particle will be trapped at such sites. Thus the particle acquires a

site dependent infinite memory contrary to the model assumption. In Ref. 10 this problem was circumvented by introducing the disorder only after symmetrizing Eq. (1). Here it will be shown that the same results can be obtained by establishing directly the analogue of Eq. (1) for the disordered case.

The generalization of Eq. (1) to the case of a bond disordered lattice is obtained by introducing the memory length explicitly. To do this it is necessary to allow transitions from a site to itself (i.e. $i = j$ in Eq. (1) is now allowed). Then the equivalent of Eq. (1) for the disordered case can be written as

$$\frac{d}{dt}P(i,j,t) = \sum_k A_{ij}w_{ijk}A_{jk}P(j,k,t) - \sum_l A_{li}w_{lij}A_{ij}P(i,j,t).\qquad (2a)$$

The first summation runs over all k that are nearest neighbours of j and over $k = j$. Similarly the second sum runs over $l = i$ as well as all l that are nearest neighbours of i. The disorder is represented by the quantities A_{ij} defined as $A_{ii} = 1$ for all i, and

$$A_{ij} = A_{ji} = \begin{cases} 1 & \text{if bond } (ij) \text{ is present,} \\ 0 & \text{if bond } (ij) \text{ is absent.} \end{cases}\qquad (2b)$$

The transition rates w_{ijk} depend on the history of traversed bonds and are defined as

$$w_{ijk} = \begin{cases} w & \text{for } i \neq j \neq k \\ w_b & \text{for } i = k, i \neq j \\ w_b & \text{for } i = j = k \\ w & \text{for } j = k, i \neq j \\ (M + z - z_i)w & \text{for } i = j, i \neq k \\ 0 & \text{otherwise}. \end{cases}\qquad (2c)$$

Here w_b is the transition rate for transitions to the previously visited site, w is the transition rate to all other nearest neighbour sites, z is the coordination number of the underlying regular lattice, and z_i is the coordination number of site i. The

constant M is related to the memory length. It regulates the transitions from a site to itself, i.e. from state (i, j) with $i \neq j$ to state (i, i). Such a transition corresponds to a loss of memory of site j while waiting at site i. The memory length in units of $\frac{1}{w}$ is given by $\frac{1}{M}$. In the limit $M = 0$ the walker forgets the previously visited site only by jumping to a nearest neighbour. For $M = 0$ and no disorder, i.e. if $A_{ij} = 1$ for all i, j and $z_i = z$, Eq.(2) reduces to Eq. (1).

Equation (2) can be symmetrized by introducing the probability density $P(i, t)$ to find the walker at site i at time t if it started from the origin at time 0. It is defined as

$$P(i, t) = \sum_j P(i, j, t)$$

where the sum runs over $j = i$ and all nearest neighbours j of site i. One finds the result

$$\frac{d^2}{dt^2} P(i, t) + (\gamma + w_b + w(z_i - 1)) \frac{d}{dt} P(i, t) + \gamma w z_i P(i, t)$$

$$= w \sum_{j\{i\}} A_{ij} \frac{d}{dt} P(j, t) + w\gamma \sum_{j\{i\}} A_{ij} P(j, t).$$

Here $\gamma = w_b + w(M + z - 1)$ and the summations run only over nearest neighbour sites of site i. Note that the equation is of second order in the time variable. The equation can be rewritten as

$$\frac{d^2}{dt^2} P(i, t) + (\gamma + w_b - w) \frac{d}{dt} P(i, t) = w \sum_{j\{i\}} A_{ij} \frac{d}{dt} \left(P(j, t) - P(i, t) \right)$$

$$\tag{3}$$

$$+ w\gamma \sum_{j\{i\}} A_{ij} \left(P(j, t) - P(i, t) \right).$$

This symmetrized form is analogous to the master equation for an uncorrelated random walk in a disordered system, and it has been used as the starting point for an effective medium treatment[10].

Combining the effects of correlations and disorder as in Eq. (3) has several interesting effects on the frequency dependent conductivity $\sigma(\omega)$ as discussed in Ref. 10. For example there exists the possibility of a maximum in $\mathrm{Re}\sigma(\omega)$ for certain values of the parameters p and $w_b < w$. For $w_b > w$ it is possible to find crossover behaviour in $\mathrm{Re}\,\sigma(\omega)$ which resembles the power law behaviour $\mathrm{Re}\,\sigma(\omega)$ of universal dielectric response over almost two decades in frequency. This is very reminiscent of experimental results[16] on Na-β-alumina where correlations and disorder are known to be important.

In summary this paper has reconsidered Fürth's correlated random walk for a bond disordered lattice. It was shown that the problem can be formulated using a first order master equation if the memory length is introduced explicitly into the equations. The first order formulation and a previously used second order equation for the symmetrized quantities $P(i,t)$ were found to be equivalent.

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