A priori prediction of diffusion cage-trapping exponent in the dynamic Ising model

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ABSTRACT

We address the problem of cage-trapping in diffusion through network structures exhibiting dynamic disorder using kinetic Monte Carlo Ising simulations to generate evolving network configurations. Diffusion is studied using blind random walkers (RWs) and we partition the net displacement of the RWs into two terms that represent contributions from transport through neighboring conducting clusters and self-diffusion of the site on which the RW finds itself, respectively. We propose a new scaling analysis for the diffusion cage-trapping time in this system and provide a theoretical analysis for estimating the trap-time scaling exponent that appears to universalize data obtained from computer simulations in both 2d and 3d Ising lattices.

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1. Introduction

The analysis of diffusion through disordered structures is a problem of widespread interest to many areas of science and engineering [1]. Many of the most successful theories in this area have used static percolation concepts, and applications have been made to a variety of interesting problems. These various studies have generally studied diffusion through “frozen” (quenched) structures consisting of randomly distributed conducting-sites surrounded by non-conducting host material [2]. Computer simulations of this process usually employ particles called random walkers (RWs) that diffuse throughout the system by “hopping” from one conducting-site to another. The rate of diffusion throughout the system depends upon the structure’s fixed topological connectivity, as well as kinetic rules governing the RW hopping transition rates. These types of theories have introduced many useful ideas to the field: critical exponents that govern the percolation transition, scaling ideas, and the notion of critical thresholds for the appearance of incipient spanning conducting clusters in a given lattice geometry [1]. In the case of classical percolation theory, however, the diffusion of the RWs is restricted to the conducting-site cluster in which the walker initially belongs. This implies that below the static percolation threshold, the mean-square displacement of a walker in the system approaches a finite value after some time, with the concomitant result that the conductivity becomes zero in the long-time limit. This is not the case if the conducting structure is allowed to change its configuration over time [3,4]. This latter situation is often referred to as the dynamic or stirred percolation problem and is the focus of the present work. A good example of a relevant physical problem is the analysis of electrical conduction in dynamic microemulsion mixtures [5–9]. Conductivity measurements in such systems have revealed the onset of sudden conductivity increases when increasing either the density of conducting microemulsion droplets, or the system temperature, above certain thresholds – which are not the same as the static threshold values [9]. In many network materials dynamics can slow dramatically, a phenomenon sometimes caused by confinement effects or what is often called cage-trapping. Trapping is the term used generically to refer to the situation where significant structural changes in the system are required to facilitate continuing diffusion [10,11]. Many open, interesting problems in statistical physics involve dynamical phenomena of this kind and here we study a simple statistical mechanical model where cage-trapping plays a significant role in network diffusion. In this Letter we present scaling equations for this model that involve a trap-time scaling exponent and we also provide a theoretical analysis showing how it can be estimated a priori from cluster size distribution arguments in the network model.

2. Computational realization of the network structure

We developed dynamic network structures using kinetic Monte Carlo (KMC) simulations, consistent with Kawasaki dynamics (i.e. constant conducting-site density) [8,12–14] on Ising lattice models [8,15]. At any point during the simulations conducting-site pathways (with density \(\phi\)) are taken to be given by the network of up spins, using the Ising terminology, with the non-conducting-sites represented by the down spins. The thermodynamic properties of this system are well established in terms of the reduced Ising lattice temperature \(T_c\), where \(T\) is the system temperature and in 2d, for example, \(T_c = \frac{1}{\ln 2}\) is the critical temperature in which k\(b\)
is Boltzmann’s constant and \( \Gamma \) the spin (site)–spin (site) coupling parameter \([16,17]\).

For the 3d system. Given a lattice of size \( L \times L \times L \) we pre-equilibrate the system by doing a number of Monte Carlo Steps (MCS), where a MCS consists of a complete sweep of spin exchanges, i.e., \( L^3 \) updates. In addition to the ‘usual’ Ising parameters another feature of our simulation model is the ability to update only a fraction of the conducting-sites during any step of the simulation. After pre-equilibration we perform the diffusion simulations as follows: a RW is placed on a randomly selected conducting-site and one of its neighboring sites selected randomly. If the selected site is a conducting-site the RW moves to it otherwise the RW remains fixed at its current position. The number of RW steps attempted between equilibration we perform the diffusion simulations as follows: a RW moves to it otherwise the RW remains fixed at its current position. The number of RW steps attempted between consecutive structure updates is defined by the symbol \( n_w \) and the fraction of conducting-sites updated per lattice sweep by \( q \).

Thus, the number of conducting-sites updated each lattice sweep is \( N_w = qL^3 \phi \). Furthermore, we define characteristic time constants for the RW and structure evolution dynamics by the variables \( T_w \) and \( T_x \), respectively. It follows in a straightforward fashion that \( T_w \sim q^{-1} \) and \( T_x \sim n_w \) with \( n_w \), normalized to the value one. Therefore, \( T_x(q) \) represents the relative time scales of structure and RW dynamics. We partition the RW walker displacement into two contributions: (1) those that arise from self-diffusion of a site on which it may be resident and (2) displacements that take place purely because of its hopping to neighboring sites. It turns out that these respective transport mechanisms contribute unique characteristics to the diffusion trajectory of theRWs.

3. Simulations and scaling results

In our previous papers \([18–20]\) we proposed the following scaling equation:

\[
\langle r^2(t) \rangle \sim \frac{1}{T_x} \left( \frac{t}{T_x} \right)^{(\phi_c - \phi)^{\theta_x}},
\]

where \( \phi_c \) is the concentration at the dynamic percolation transition which we find from finite-size scaling arguments. The value for \( \phi_c \) depends upon dimensionality \([20]\) and \( Q \) is a general scaling function that can be found empirically from simulation data by collapsing data using this scaling relationship in Eq. (1).

<table>
<thead>
<tr>
<th>( n^* )</th>
<th>( d_1 )</th>
<th>( d_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2d</td>
<td>1.949</td>
<td>1.8958</td>
</tr>
<tr>
<td>3d</td>
<td>2.037</td>
<td>2.5234</td>
</tr>
</tbody>
</table>

Using Eq. (5) to find \((s^2/\theta_x)\) we get that,

\[
\frac{1}{(s^2/\theta_x)} \sim \int_0^\infty s^{2(\delta_1 - 1)} \exp(-(\phi - \phi_c)^2s)ds \sim (\phi_c - \phi)^{2 + 2/(\delta_1 - 1)}.
\]

From Eq. (6) we can immediately find an estimate for \( \theta_x \),

\[
\theta_x = 2\left( \frac{2}{\delta_1 - 1} \right).
\]

where \( \delta_1 \) in Eq. (7) is known independently for various lattice types (it depends upon the lattice dimension) and \( \tau \) is usually found to be less than 3 in static percolation models. However, in our simulation model we were able to find \( \tau \) directly from simulations where we employed a cluster counting algorithm to find the distribution of cluster sizes. These results are presented in Table 1 for 2 and 3d, respectively, and also in Table 1 we provide our theoretical estimates of \( \theta_x \) in our 2d and 3d dynamic Ising lattice models, based upon the values of \( \tau \). In Fig. 1 we show an initial set of network

![Scaling for diffusion mean-square displacement using theoretically predicted values for \( \theta_x \) exponent for 2d (left) and 3d (right) systems, respectively.](image)
4. Conclusions

In this paper we investigated a novel conjecture for estimating the diffusion trapping-time exponent $h_z$ introduced in a previous paper from our group [19] where we addressed the problem of diffusion through network structures exhibiting dynamic disorder. Here we showed that the trapping-time exponent is given by the equation

$$h_z = \frac{2^{2 + d_f}}{C_0^{s/C_16/C_17}}$$

where $d_f$ is the fractal dimension of a typical conducting-site cluster and $s$ is the power law scaling exponent for these cluster size distributions which were found from simulation results in our network system. These results showed excellent scaling collapse of the simulation data in both 2d and 3d network systems and are the first results, that we are aware of, providing a theoretical route for estimating a priori diffusion trapping-time exponents in dynamic disordered network materials.

Appendix A. Condition under which the conjecture holds

We need to show that

$$\langle r_0^2 \rangle = \kappa \langle r_c^2 \rangle, \quad \text{(8)}$$

where $r_0$ and $r_c$ are the displacements in the non-conducting and conducting-site space, respectively, and $\kappa$ is a constant. Since

$$\text{Var}(r_0, r_c) = E[\langle r_0^2 \rangle, \langle r_c^2 \rangle] - \mu_{r_0,r_c}^2, \quad \text{(9)}$$

where $\mu_{r_0,r_c}$ is the mean of the random variable that is product of the $r_0$ and $r_c$. Now if $r_0$ and $r_c$ are independent random variables as we should expect them to be in our simulation model then,

$$E[\langle r_0^2 \rangle, \langle r_c^2 \rangle] = \langle r_0^2 \rangle \cdot \langle r_c^2 \rangle. \quad \text{(10)}$$

Therefore,

$$\langle r_0^2 \rangle \cdot \langle r_c^2 \rangle = \text{Var}(r_0, r_c) + \mu_{r_0,r_c}^2, \quad \text{(11)}$$

We now postulate that $r_0$ and $r_c$ are both independent binomial random variables since they represent independent displacement steps taken with 'success' probabilities $p$ and $q$, respectively. In this case it immediately follows that

$$\langle r_0^2 \rangle \cdot \langle r_c^2 \rangle = pq$$

i.e. $\kappa = pq$.

References