Random walks in percolating networks with two jump frequencies

Joseph P. Straley
Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506-0055
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The time dependence of the mean-squared displacement of a random walker on a random network is studied, for the case that there are two very different jump probabilities and that the concentration of the better conducting bond is close to percolation. A scaling theory is proposed and discussed, and numerical simulations of the three-dimensional bond problem are used to verify the scaling predictions.

I. SCALING LAWS

How are the laws of diffusion and transport modified when the medium in question is a random AB mixture of good- and poor-conducting regions? Hong *et al.*[1] have given a scaling theory and qualitative discussion in response to this question. The purpose of this paper is to present a slightly different interpretation of this scaling theory, and to give some simulation results that demonstrate that this alternate discussion is correct.

The system under consideration is modeled as a lattice[2] with a random distribution of jump rates assigned to its bonds. Specifically there is a concentration p of good-conductor bonds with (large) jump rate B, and a concentration $1-p$ of poor-conductor bonds with (small) jump rate A. At long times the mean-squared displacement of a random walk on this lattice will in general grow linearly with time T, at a rate determined by the diffusion constant. The behavior of the diffusion constant near the percolation threshold is the same as that of the conductivity in the corresponding network, which exhibits singular behavior as $p \to p_c$ and $A/B \to 0$. Building upon the scaling theory for that problem[3,4] and the scaling theory for the $A=0$ case,[5] we consider a scaling law

$$\begin{aligned}\langle R^2 \rangle &\equiv T\, D(\epsilon, A, B, T^{-1}) \\ &= \mu T\, D(\epsilon/\lambda, \lambda^{-s} A/\mu, \lambda^t B/\mu, d^2 \mu^{-1} \lambda^{(\beta-2\nu)} T^{-1}) ,\end{aligned} \tag{1}$$

where $\epsilon=(p-p_c)/p_c$, d is the lattice spacing (or any other microscopic length), and λ and μ are arbitrary parameters. This equation only holds in the critical region, which restricts its applicability to the limits that ϵ is small, A is much less than B, and T is large. It asserts an internal structure to D, such that it holds constant value along curves in the ϵ, A, B, T parameter space. The scaling in μ states how the diffusion constant and the rate coefficients transform under a change of units for T and is a simple consequence of dimensional analysis. Hong *et al.*[1] have attempted to make something more out of it, claiming that it links two limiting problems. However, the only difference between an "ant" problem (such as $A=0.001$, $B=1$) and a "termite" problem ($A=1$, $B=1000$) is a scale factor (here 1000) in the diffusion constant and time scale.

The scaling in λ is nontrivial, and implies that for smaller ϵ the same value of $\langle R^2 \rangle / T$ will be observed if A is decreased while B and the time scale are increased in specified ways. The form of the function is determined by matching to the standard special cases, which will now be verified.

The diffusion function D is only singular when the three arguments ϵ, A/B, and T^{-1} are simultaneously zero; if we identify the largest of these, we can perform expansions in the other variables. Thus for $|\epsilon| > (A/B)^{1/(s+t)}$ and long times, expanding in the small parameters gives

$$\begin{aligned}\langle R^2 \rangle \approx{}& \epsilon^t B T\, D(\mathrm{sgn}\epsilon, 0, 1, 0) \\ &+ \epsilon^{-s} A T\, D(\mathrm{sgn}\epsilon, 0', 1, 0) \\ &+ \epsilon^{\beta-2\nu} d^2\, D(\mathrm{sgn}\epsilon, 0, 1, 0') + \cdots ,\end{aligned} \tag{2}$$

where the prime indicates a partial derivative on the indicated variable, and values $\mu = B\epsilon^t$ and $\lambda=\epsilon$ been chosen to simplify the representation.

For $\epsilon > 0$, $D(1,0,1,0)$ is nonzero and the leading term describes the diffusion in the percolating phase. The last term is an offset due to small time behavior, and the second term is unimportant in this case.

For $\epsilon < 0$, there is no diffusion for $A=0$, so that $D(-1,0,1,0)$ vanishes. The second term now describes the small diffusion below the percolation threshold, and the last term is the contribution from finite clusters. The prefactor of the last term is $\epsilon^{\beta-2\nu}$ rather than $\xi^2 \approx \epsilon^{-2\nu}$ because this is the average over walks rather than the average over clusters:[6] the ensemble weights differ in that the probability that the random walker is on any given site of a cluster is inversely proportional to the size of the cluster.

In both of these cases "large T" is defined relative to the time scale

$$\tau \approx \epsilon^{\beta-2\nu-t} d^2/B . \tag{3}$$

Hong *et al.*[1] define a second time constant

$$\tau_H \approx \epsilon^{\beta-2\nu+s} d^2/A . \tag{4}$$

They identify τ as being the time required for the random walker to explore the typical finite cluster and τ_H as the

time required to escape from it, and give a sketch in which $\langle R^2 \rangle$ is increasing for $T < \tau$, has a plateau for $\tau < T < \tau_H$, and exhibits a linear (diffusive) increase for $\tau_H < T$. This picture is based on the assertion that most of the sites on the perimeter of a finite cluster are interior to it. However, with the exception of the two-dimensional square bond problem, this is generally not true. In three dimensions ($p_c = 0.2492$) the poorly conducting region occupies three fourths of the lattice and is quite well linked; almost every perimeter site can be used to escape from the cluster.

It is contended here that there is only one relevant time scale, just as there is only one length scale. This time scale characterizes the length of time required to adequately sample the various environments of the percolating system; on time scales larger than τ the inhomogeneity has been averaged out allowing the linear-in-T behavior (which is already present at the earliest times) to be clearly perceived. Note that for $A = B$, the linear increase of $\langle R^2 \rangle$ starts immediately at $T = 0$ with no time constant required. The time penalty to the random walker to finally escape from a finite cluster of B links (as distinct from leaving the cluster for a region interior to it) is already included in the conductivity exponent t. The time interval τ_H can be retrieved from the theory as being the time required for a mean-squared displacement of order $\epsilon^{\beta-2\nu}$ (a characterization of the size of the finite clusters) for a diffusion process with diffusion constant of order $A\epsilon^{-s}$; however, nothing interesting happens on this time scale. Numerical evidence for this contention will be given below.

The scaling law (1) can also be used to discuss the large-time behavior for $|\epsilon| < (A/B)^{1/(t+s)}$. In this region the A/B is the controlling parameter and we can expand in ϵ and $1/T$:

$$\begin{aligned}\langle R^2 \rangle \approx & (A/B)^{t/(t+s)} BT\, D(0,1,1,0) \\ & + \epsilon (A/B)^{1/(t+s)} BT\, D(0',1,1,0) \\ & + (A/B)^{(\beta-2\nu)/(t+s)} D(0,1,1,0') \end{aligned} \tag{5}$$

for small A the range of ϵ is quite small. The first term introduces the characteristic conductivity[3]

$$\sigma_c \approx A^{t/(s+t)} B^{s/(s+t)} \tag{6}$$

and the characteristic length scale is

$$\xi \approx (A/B)^{-\nu/(t+s)} . \tag{7}$$

For short times, the fourth argument of D is controlling, and the scaling law becomes

$$\begin{aligned}\langle R^2 \rangle \approx & d^2 \left[\frac{BT}{d^2} \right]^{(2\nu-\beta)/(t+2\nu-\beta)} D(0,0,1,1) \\ & + \frac{Ad^2}{B} \left[\frac{BT}{d^2} \right]^{(t+s+2\nu-\beta)/(t+2\nu-\beta)} \\ & \times D(0,0',1,1) + O(\epsilon) . \end{aligned} \tag{8}$$

At the percolation threshold and for $A = 0$, the "small-time" regime extends to infinity, and $\langle R^2 \rangle$ has a pure power-law behavior for all but the shortest times (where nonuniversal behavior is most likely to be encountered).

Fitting this power law is an alternative route to the determination of the exponent t, which has both advantages and disadvantages relative to the Kirchhoff law approach. The greatest advantage is that the simulation program is very simple, and trivially vectorized (by considering a set of noninteracting walkers). A major drawback is that a large number of walks needs to be considered to extract the diffusion constant from the statistical noise; but this is partly discounted by the observation that what is wanted is not the diffusion constant for any one network but the diffusion constant for an ensemble of them: the Kirchhoff's law approach gives too much information (at great expense) about one realization and encounters the statistical fluctuation problem in the ensemble averaging; the random walk method gives incomplete information about one realization quite cheaply, allowing a large number of configurations to be averaged in. This approach has been used[7-9] to determine the ratio t/ν to good accuracy.

The foregoing discussion assumes that one of the three parameters ϵ, A/B, or $1/T$ is controlling. However, in the crossover region where, for example, $\epsilon^{t+2\nu-\beta}$ and T^{-1} are comparable, both scaling laws apply and the corresponding terms match.

Gefen *et al.*[10] have given a different result for short times ($A = 0$):

$$\langle R^2 \rangle \approx T^{2\nu/(t+2\nu-\beta)} . \tag{9}$$

Their theory differs from the present one in that $\langle R^2 \rangle$ is calculated using only the walkers which are on the infinite cluster. Close to the percolation threshold the density of the infinite cluster is vanishing (with exponent β); the behavior is different since it is a miniscule and rather anomalous subset of all walkers that is being considered. The usefulness of this alternate definition is debatable: we observe that the restriction to the infinite cluster is only meaningful when $A = 0$, and that their result is not obtained as the $A \to 0$ limit. In a transport measurement, the current resulting from a finite frequency driving field (which would be sensitive to the value of the time constant) would include contributions from finite clusters intersected by the electrodes, and there would be response currents in internal finite clusters, which also experience the driving field. The Gefen *et al.* definition has the attractive feature that it studies the most interesting clusters, but it not been used for numerical simulations, since it requires identifying the infinite cluster (i.e., one that extends beyond the boundaries of the simulation box); close to p_c in a large box it will be relatively uncommon that the "infinite" cluster includes the origin, making it difficult to get good statistics.

II. SIMULATION RESULTS

The foregoing ideals were tested by generating random walks on percolating clusters on the simple cubic lattice, for which[11] $p_c = 0.2492$. The three-dimensional percolation exponents are believed to have the values[12-15] $\beta = 0.454 \pm 0.008$, $\nu = 0.88 \pm 0.01$, $t = 1.94 \pm 0.1$, and

$s=0.75\pm0.04$. Each bond was assigned a jump probability (A or B=unity) at random so that the density of B bonds was the specified p. The random walkers were started from the center of a 31^3 (or larger) cube with reflecting boundaries. At each time step each random walker chose a new bond along which to move; the move was successful according to the jump probability for the chosen bond (otherwise the walker stayed at its site). At least 5000 configurations were generated for each p, and 128 walks of 1000 (or more) steps were performed on each configuration (this made optimal use of the vector capability of the IBM 3090). It was verified that few walks ever reached the boundary for $p \leq 0.27$. The figures to be shown below display the averages for 1000 equally spaced times without any smoothing. The sample size is large enough that the statistical fluctuations cannot be seen on these figures. Multiply sampling each configuration shares with the "exact enumeration" approach[16] the possible defect that the curves, though smooth, are erroneous because too few configurations have been studied to fully characterize the ensemble. This can be checked for by repeating the simulation with a different random number sequence; on the data to be presented, the error seems to be on the order of 1% in the worst case.

Figure 1 shows $p=0.20$ and various values of A. As predicted by Eqs. (2) and (8), the effects of finite A are seen at the earliest times and are manifested by a nearly constant divergence of $\langle R^2 \rangle$ from its $A=0$ form. For all these curves τ, as defined by Eq. (3), has the common value 196 time steps consistent with the curves being linear for times of this order [Eq. (3) seems to overestimate the time scale by a factor of 10]; $\tau_H \approx 2.47/A$ varies from 500 (for $A=0.005$) to 25 000 (for $A=0.0001$). According to the interpretation of Hong *et al.*,[1] these curves should coincide for $T<\tau_H$; the lower curves should be indistinguishable.

Figure 2 removes the power law part for $A=0$ and small T by dividing by the $A=0$ result, which reduces the data to lines which (according to the scaling laws presented above) are straight for large T and approximated for smaller T by $T^{(t+s)/(t+2\nu-\beta)} \approx T^{0.83}$, which is almost straight.

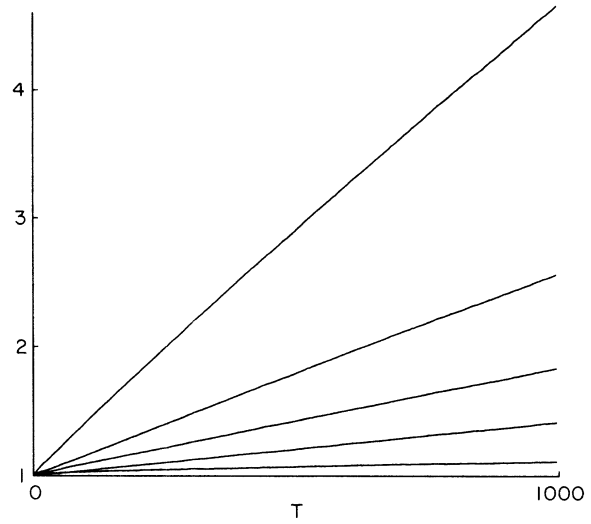

FIG. 2. The results of dividing the data of Figure 1 by $\langle R^2 \rangle$ for $A=0$. This largely removes the time dependence on time scale τ and shows that there is no other time scale in the problem.

Figure 3 displays $\langle R^2 \rangle$ for fixed $A=0.0005$ and various p spanning the percolation threshold. The time constant τ, which determines the time of the onset of linear growth of $\langle R^2 \rangle$, is short for the outside curves and longer but still finite at p_c. None of these curves exhibits a plateau.

Figure 4 illustrates the case $p=p_c=0.2492$ for various values of A. For $A=0$, $\langle R^2 \rangle$ is a pure power law in T

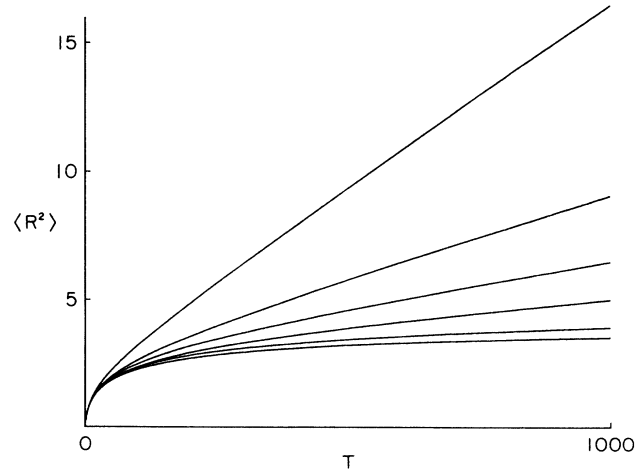

FIG. 1. $\langle R^2 \rangle$ as a function of time for $p=0.20$. The values of A are 0.0, 0.0001, 0.0005, 0.0010, 0.0020, 0.0050. Each curve represents $128 \times 10\,000$ walks of 1000 steps on a 31^3 cube.

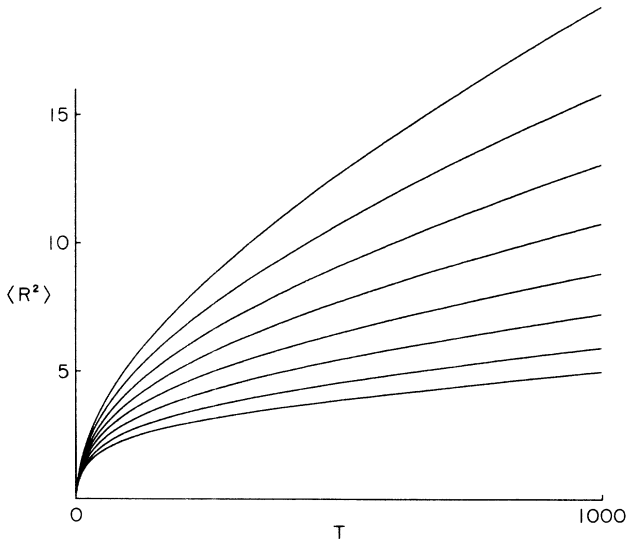

FIG. 3. Dependence of $\langle R^2 \rangle$ on time for $A=0.0005$. The values of p (starting with the lowest curve) are 0.20, 0.21, 0.22, 0.23, 0.24, 0.25, 0.26, and 0.27. Each curve represents $128 \times 10\,000$ walks of 1000 steps on a 31^3 cube.

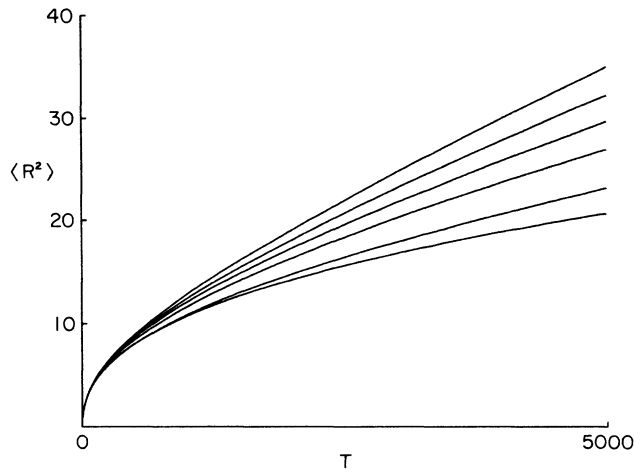

FIG. 4. $\langle R^2 \rangle$ as a function of time, for $p = p_c = 0.2494$. The values of A considered (starting from the lowest curve) are 0.0, 0.0001, 0.0002, 0.0003, 0.0004, and 0.0005. Each curve represents 128×5000 walks of 5000 steps on a 61^3 cube.

[according to Eq. (8)] with the exponent $(2\nu-\beta)/(t+2\nu-\beta) \approx 0.40$; it was verified that the bottom curve in this figure can be reasonably fit using the exponent values given previously.

The scaling laws (8) and (5) can be combined to show that the family of curves shown in Fig. 4 should coalesce into a single universal curve if $(A/B)^{(2\nu-\beta)/(t+s)}\langle R^2 \rangle$ is plotted as a function of $z = (A/B)^{(t+2\nu-\beta)/(t+s)} BT/d^2$. Figure 5 shows that the curves do superimpose when plotted this way. The result is one not very sensitive to the value of s. The resulting curve can be fairly successfully represented by the function $a + bz + cz^{0.4}$.

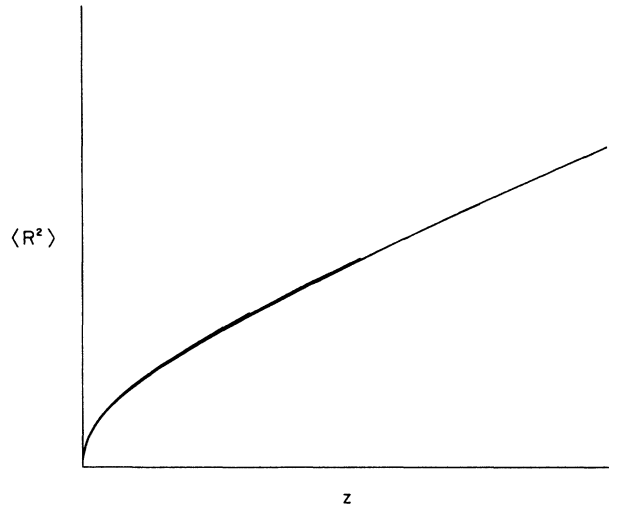

FIG. 5. $(A/B)^{(2\nu-\beta)/(t+s)}\langle R^2 \rangle$ vs $z = (A/B)^{(t+2\nu-\beta)/(t+s)} \times BT/d^2$, using the same data as Fig. 4.

III. FREQUENCY DOMAIN

Several different definitions have been given for a frequency-dependent conductivity or diffusion constant, based on different models for the transport mechanism.

Gefen and Goldhirsch[17] have discussed the response of a diffusion model to time-dependent sources. They find that the ratio of current to driving field depends strongly on sample dimension, so that conductivity is no longer a useful definition. This model takes the equivalence of dc conductivity and the diffusion constant too literally; in general currents are driven by fields, not concentration gradients.

In another model for the inhomogeneous conductor at finite frequency, the resistors become impedances.[18,19] Capacitance between grains provides a significant reactive part to the poor conductances A. This model is most appropriate to the low-frequency domain, below the plasma frequency of the good conductor. Note that in the low-frequency limit, there is no field in dead-ended branches: it is screened out very quickly by the electron gas. The theory for this model is a simple generalization (making all conductivity variables complex) of the theory of the two-component conductor. Thus at p_c, the ac conductivity is expected to have a $\omega^{t/(t+s)}$ ($\approx \omega^{0.7}$) frequency dependence[20] [compare Eq. (6)].

A different model considers particles that do not interact or interact via a short-ranged force and which are driven by a uniform external field.[21] This model might also apply to the low concentration limit of the diffusion of charged particles (for which the plasma frequency is very low). Now particles are carried into the dead-end branches by the field, and the time required for a diffusing particle to sample the various environments becomes significant to the dynamics. Linear response theory applies to this case, and the frequency-dependent conductivity is proportional to the Fourier transform of $\langle R^2 \rangle$:

$$\sigma(\omega) \propto -\omega^2 \int_0^\infty e^{-i\omega T - \eta T} \langle R^2(T) \rangle dT \ , \tag{10}$$

where η is a positive convergence factor. A scaling theory similar to Eq. (1) can be given for σ:

$$\sigma(\epsilon, A, B, \omega) = \mu \sigma(\epsilon/\lambda, \lambda^{-s} A/\mu, \lambda^t B/\mu, d^2 \mu^{-1} \lambda^{\beta-2\nu} \omega) \ , \tag{11}$$

which can be discussed exactly as has been discussed previously. For example, at p_c the frequency dependence of the conductivity is predicted to be $\sigma \approx \omega^{t/(t+2\nu-\beta)} \approx \omega^{0.6}$. $\sigma(\omega)$ can also be related to a network problem, but now the capacitances are between the sites and a ground plane, rather than between sites.[5]

There is a very good reason for pursuing this approach: The function $\langle R^2(t) \rangle$ contains information about all frequencies, and constructing its Fourier transform is equivalent to solving the network equations many times. In this respect the random walk method gives more information than the Kirchhoff law approach, which only gives the conductivity for one frequency at a time; the counterbalancing difficulty is that to get the true zero-frequency conductivity, the random walks must extend to very long times.

Attempting to perform the Fourier transform on the simulation results, one runs into several problems. For nonzero A or $p > p_c$, the integral in Eq. (10) should be divergent (as ω^{-2}) for small ω, but this will only be reproduced by the numerical integration if the range of integration is several times larger than $1/\omega$. Integrating Eq. (10) by parts eliminates this problem but requires differentiation of the numerical data, which amplifies the statistical fluctuations in it. The coefficient of the divergence (which is the dc conductivity) is determined by the asymptotic slope of $\langle R^2(T) \rangle$, which cannot be accurately determined for most of the curves shown in Figs. 1 and 2 (τ is too large). The statistical fluctuations also become prominent for frequencies comparable to the reciprocal of the time step. The needed extrapolation and smoothing of $\langle R^2 \rangle$ can be provided by fitting it to a suitable functional form. Thus, the ability to fit the data for $p = p_c$ to the function $a + bT + cT^{0.4}$ demonstrates that $\sigma(\omega)$ is well represented by the scaling form $\sigma \approx b + c\omega^{0.6}$.

IV. ACKNOWLEDGMENTS

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