Random-walk calculation of conductivity in continuum percolation

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Random walks are used to obtain the diffusion constant for continuum percolation models of composite materials in two and three dimensions. An Einstein relation is then used to find the conductivity. The same calculation gives the dielectric constant for the composite. First-passage-time methods and special boundary conditions are used for systems where both materials have finite conductivity, where one component is a superconductor, and where one component does not conduct. The percolation models consist of randomly placed overlapping spheres in three dimensions or disks in two dimensions. Our results are consistent with known results where applicable and are far better than effective medium theories. Estimates for anomalous diffusion exponents at percolation were also found.

I. INTRODUCTION

We provide a systematic methodology for computing the conductivity and dielectric constant for continuum models of composite materials. Mixtures of two substances are considered where the inhomogeneities are large enough such that within each part of the material its properties are determined by macroscopic constitutive relations. Examples of such systems include porous rocks filled with brine,[1] dielectrics with metallic inclusions separated by insulating material[2] and possibly granular superconductors.[3] The method we use is an extension of the procedure employed by Schwartz and Banavar[4] for the grain consolidation model of porous rocks and later used by Tobochnik *et al.*[5] to model conduction in a plane containing random cuts. The method involves extracting the diffusion constant from a computation of the mean-square displacement of many random walkers. The conductivity is then determined via an Einstein relation. The same calculation gives the dielectric constant of the composite.

Other methods for dealing with composite materials either rely on an underlying lattice or are restricted to low densities of inclusions or inclusions of simple shapes that do not overlap. In the former case the lattice is replaced by a random resistor network and the equivalent resistance is then found using relaxation methods or solving Kirchloff's equations for the network. More recently the $Y-\Delta$ transfermation[6] for two-dimensional (2d) lattices and multigrid techniques[7] have improved the numerical efficiency tremendously. However, it is important to be able to solve these problems in a continuum using more realistic models such that results away from the percolation threshold are meaningful. Also, at the threshold the critical transport properties of some continuum systems are expected to differ from their lattice counterparts, even though the static exponents are the same.[8]

At low densities, effective medium theories (EMT) can be used[9] Two common versions are the symmetric version of Bruggemann's (SB) theory where the composite conductivity σ_c is found from the solution of

$$\phi_1\frac{\sigma_1-\sigma_2}{\sigma_1+(d-1)\sigma_c}+\phi_2\frac{\sigma_2-\sigma_c}{\sigma_2+(d-1)\sigma_c}=0\ , \tag{1}$$

where ϕ_i is the fraction of material with conductivity σ_i and d is the spatial dimensionality. An alternative EMT is the Clausius-Mossotti (CM) approximation derived for dielectrics in most electricity and magnetism texts which gives

$$\frac{\sigma_c-\sigma_1}{\sigma_c+(d-1)\sigma_1}=\phi_2\frac{\sigma_2-\sigma_1}{\sigma_2+(d-1)\sigma_1}\ . \tag{2}$$

In the CM approximation the fraction of material two must be small. In the case where $\sigma_2=0$, these approximations reduce to

$$\sigma_c^{\rm SB}=1-\frac{d}{d-1}\phi_2\ , \tag{3}$$

$$\sigma_c^{\rm CM}=\frac{1-\phi_2}{1+[1/(d-1)/\phi_2}\ . \tag{4}$$

Unlike the SB approximation the CM result does not predict a percolation threshold. The above results assume spherically symmetric inclusions.

In addition to EMT another approach is to do a multipole expansion.[10–12] These expansions work for nonoverlapping inclusions of simple shapes, when the density of inclusions is not too large. However, for high densities and large differences between the dielectric constants (or conductivities) of the components too many multipoles are needed to make the numerical calculation feasible. One advantage of the multipole method, however, is that one can calculate the frequency dependence of the dielectric constant.

II. GENERAL SIMULATION PROCEDURE

The model we used consisted of a collection of spheres (disks in $2d$) placed randomly in an L^d d-dimensional box. The spheres are allowed to overlap. We define the radius of each sphere to be unity and define the conductivity of the spheres to be σ_2. The conductivity of the re-

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gion between the spheres is denoted by σ_1. In all cases either σ_1 or σ_2 is defined to be unity. We will define the overall conductivity of the composite system to be σ_c. We have considered cases for which $\sigma_i = 0$, 2, 5, 10, and ∞. The volume fraction of the background, ϕ_1, is related to the number density of the spheres n by

$$\phi_1 = e^{-nV} , \tag{5}$$

where V is the volume of a sphere or in $2d$, V would be replaced by the area of a disk.

There are three cases to consider for the random-walk algorithm depending on whether the conductivity for one component is zero, finite, or infinite. Here we will discuss those parts of the algorithm that are common to all cases. In the standard random walk the mean-square displacement of a walker from its original position is

$$\langle r^2 \rangle = N \langle s^2 \rangle , \tag{6}$$

where N is the number of steps and $\langle s^2 \rangle$ is the mean-square step size. In a continuum a Brownian particle would move such that

$$\langle r^2 \rangle = 2dDt , \tag{7}$$

where D is the diffusion constant. Thus the random walk is equivalent to Brownian motion if we identify the time for one step to be equal to $\langle s^2 \rangle / (2dD)$. We are always interested in the diffusion constant of the composite system relative to that of a pure system with no inclusions. We denote this relative diffusion constant by D_c. We are free to choose our diffusion constant in the pure system and our units of time such that

$$\langle r^2 \rangle = D_c t \tag{8}$$

in the composite system, where $D_c = 1$ for the pure system with $\sigma = 1$.

Most of the data reported in this paper are based on averages over 1000 walkers. A new configuration of spheres or disks was created for each walker to ensure that each walk was totally independent of the others. The computer time to create the configurations is generally much less than that to simulate the walks.

The algorithm initially used for this problem was to allow the random walkers to move with a small step size s with the boundaries treated in a special way. Using only small steps, this approach is very inefficient since much of a walker's time is spent in regions far from any boundaries. Since only in the limit as $s \to 0$ is the continuum result reached, one can run into severe computational costs. Thus it is advantageous to treat the part of the walk away from boundaries in a different manner. To do this we determine the largest sphere (or disk in $2d$) centered at the walkers position that does not intersect any boundaries. Then we move the walker immediately to a random point on the surface of this sphere. Finally, we update the time according to the first-passage-time distribution which is stored in an array. This distribution can be determined numerically or analytically.[13] In the analytical calculation one solves the diffusion equation for the probability of a particle being a distance r away from its initial position at a time t, with absorbing boundary conditions at the surface of a sphere ($r = 1$) centered at the particle's initial position. From this one can compute the probability of being absorbed in a time t to $t + dt$. In the numerical approach one simply starts with many walkers at the origin and computes the number of walkers which take a time t to $t + dt$ to reach the surface of a sphere centered at the origin. Once these probabilities are known it is straightforward to create an array of times such that a random choice among the array elements gives a first-passage time with the correct probability of occurring. These times are then scaled by the square of the radius of the first-passage sphere. This kind of move is not constrained to any lattice. When a walker is close to a boundary, typically we use a distance of 0.001, the next move must be determined by the relative conductivities on each side of the boundary. In the next section we discuss what the walker does near a boundary.

An alternative method of computing the diffusion constant is to put on a small bias on the walk and compute the mean distance moved in the direction of the bias. This mean distance will be proportional to the bias and the diffusion constant. This method has been used for calculations in macroscopically inhomogeneous systems.[14] We have also done some preliminary calculations and have found that because weak biases are necessary, there is no improvement in computation time. In addition, it would be much harder to use the first-passage algorithm for speeding up the calculation since one could not move the walkers to a random spot on the surface of a sphere and the distribution of times would depend on where you moved the walker.

III. BOUNDARY CONDITIONS

The key notion in using random walkers when there are media of different conductivities is that (i) either the time for each step or the step size varies with the conductivity and (ii) when one is at the border between two regions the probabilities of moving in different directions depends on the relative conductivities. Various algorithms for random walks on lattice systems have already been discussed in the literature,[15] but the continuum situation has not been discussed in detail.

A. $\sigma = 0$

In the case where one material has zero conductivity clearly the walker cannot enter the zero conductivity regions. The only issue is what is to be done when the walker is very close to the border. The two possible solutions are called the "blind ant" and the "myopic ant" boundary conditions. In the blind ant case the walker moves in any direction and if it crosses a forbidden zero conductivity region, the walker is returned to its original place and the time is incremented. In the myopic ant case, the walker walks with equal probability in only those directions which do not cross the forbidden region. We chose the blind ant boundary condition. One way of seeing this as preferable is to imagine infinitesimal steps. In this case the walker would sometimes move in the direction forbidden by the myopic ant and bounce off the forbidden region returning to its original position, just as

is prescribed by the blind ant boundary condition.

An alternative way to see the correctness of the blind ant boundary condition is to relate it to the boundary condition that the normal component of the electric field at the surface of a sphere should be zero. In our case the electrostatic potential would correspond to the density of walkers if one imagined a very large number of walkers diffusing simultaneously. For simplicity consider the one-dimensional case where we wish to find the density of walkers next to the boundary V_0 and one step away from the boundary V_1. We want our boundary condition to lead to $V_0=V_1$ in the steady-state limit. For the blind ant case the time rate of change of V_0 is given by $dV_0/dt \propto -\frac{1}{2}V_0+\frac{1}{2}V_1$, because half the time a walker right at the boundary will move one step away and half the time it stays still, and half the time a walker at one step away will move to the boundary and half the time it moves to two steps out. In the steady-state limit $dV_0/dt=0$, and this clearly leads to $V_0=V_1$ as desired. In the myopic ant case $dV_0/dt \propto -V_0+\frac{1}{2}V_1$ since all the walkers move away from the surface in the next time step. In the steady-state limit this leads to $V_0=V_1/2$, and thus the normal component of the field is not zero as desired.

We have also examined the effect of having a uniformly distributed random step size near the boundary, and found that within our error bars there was no change in the results. We have used step sizes of 0.02 in $2d$ and 0.04 in $3d$. The advantage of a random step size is that there may be very narrow regions in which it is impossible to move with a finite step size. Our results show that these regions are not significant enough to change the results at our present level of accuracy.

B. $\sigma=\infty$

Here walkers can be in any region. However, if the walker is in the superconducting region, then assuming fixed steps, time does not change. Thus the walker can move anywhere within the superconducting region in zero time. Near the border between two regions of conductivity σ_i the probability for taking a step in the ith direction into a region where the conductivity is σ_i is

$$p_i=\sigma_i/\sum_j \sigma_j \ , \tag{9}$$

where the sum is over all possible steps. If one is just outside the superconducting region, then this implies the next step must be inside that region. If one is inside the superconducting region, then it appears that there is no way to step out since from Eq. (9) only steps inside the superconductor have a finite probability of occurring. However, since time does not change when one is inside the superconducting region, the walker can try to escape an infinite number of times, and there is thus a finite probability of stepping out somewhere. Thus our algorithm consists of taking a walker that is close to a superconducting region and moving it in one step a small distance, typically around $a_s=0.05$, outside of this region at random anywhere on the connected cluster of superconducting spheres. In practice this means pick one of the spheres in the connected cluster at random and move the walker to a random position outside this sphere. If one is still in the cluster, repeat this process until one is in a region of finite conductivity. A small time $t_s=da_s^2$ is added to the time. This is the mean first-passage time to move a distance a_s from a flat surface. This algorithm was checked on regular arrays of non-overlapping superconducting disks and compared with multipole expansions.[10] The results agreed well within the statistical errors.

TABLE I. Times for walkers to move a unit distance along a line from the boundary between two regions of conductivity $\sigma=1$ and h. The time to move into the former region is t_1 and the latter is t_h.

h	t_1	t_h
1	0.997	0.973
2	0.783	0.601
3	0.649	0.448
4	0.614	0.347
5	0.553	0.289
6	0.514	0.247
7	0.487	0.212
8	0.488	0.189
9	0.464	0.171
10	0.451	0.156

C. $\sigma_1=1$ and $\sigma_2=h$

Here we consider the case where the conductivity both inside (component two) and outside (component one) the inclusions is finite and nonzero. In this case we move the walkers a finite distance a_f typically equal to 0.02 or 0.04 toward or away from the center of the sphere or disk surface. The probability of moving toward the center is $h/(1+h)$ and the probability of moving away is $1/(1+h)$. The times for such a move equal $dt_i a_f^2$, where the t_i are the mean first-passage times for moving a distance a_f into the region with conductivity σ_i in one dimension. The times t_i were determined numerically by averaging over 10 000 walkers moving from the origin to ± 1 in steps of 0.01 where the conductivity equals 1 for negative positions and h for positive positions. These times are listed in Table I.

IV. EINSTEIN'S RELATIONS

A. $\sigma=0$

In our reduced units Einstein's relation between conductivity and diffusion for percolation clusters becomes[16,17]

$$\sigma_c=P_\infty D_c \ , \tag{10}$$

where P_∞ is the probability of being on the infinite conducting cluster. However, we put walkers down in regions that have nonzero conductivity but are totally enclosed by nonconducting regions and thus cannot contribute to the conductivity. It is numerically impossible to determine whether or not a walker is in one of these

regions. We will now show that $\sigma_c = \phi D_m$, where D_m is the slope at long times of $\langle r^2 \rangle$ versus t and ϕ is the fraction of material with unit conductivity. Imagine we distributed walkers uniformly in our system. Then there would be three types of walker. There will be n_0 walkers that land on the zero conductivity regions, n_e walkers that land on the enclosed regions, and n_∞ walkers that land on the infinite conducting region. Their mean-square displacements after a long time will be, respectively, $0, c = \text{const}$, and $D_c t$. In measuring D_m we only use the $n_e + n_\infty$ walkers, thus in the long time limit we will measure

$$\langle r^2 \rangle = \frac{n_e c + n_\infty D_c t}{n_e + n_\infty} , \tag{11}$$

and thus the slope is

$$D_m = \frac{n_\infty D_c}{n_e + n_\infty} . \tag{12}$$

Multiplying this by

$$\phi = \frac{n_e + n_\infty}{n_0 + n_e + n_\infty} , \tag{13}$$

gives the desired result

$$\sigma_c = \frac{n_\infty D_c}{n_0 + n_e + n_\infty} = P_\infty D_c . \tag{14}$$

B. $\sigma = \infty$

For the case when one component is superconducting Einstein's relation leads to $\sigma_c = \phi_n D_m$, where ϕ_n is the fraction of the material that is normal metal with conductivity equal to unity. There are many ways to see this, but perhaps the most physical is the following. Imagine a d-dimensional cube of side length L. We are interested in comparing the conductance g of this cube with superconducting inclusions with the same size cube without inclusions for which we define the conductance $g_0 = 1$. To do so we will change the composite material to a new equivalent contracted cube with no inclusions. Since each point on the superconducting regions is shorted to every other point connected to it via a superconducting region, we can replace the extended superconducting regions by points with no volume and still have the same conductance. If we measured the diffusion constant on the original composite material using our algorithm, we obtain D_m. Now the diffusion constant on the contracted system would just be $D_m \phi_n^{2/d}$, since the diffusion constant scales as length squared and the linear dimensions of the contracted cube have changed by a factor of $\phi_n^{1/d}$. Now the contracted system and the original system without inclusions are both pure systems, thus their conductivity ratio is just given by the ratio of their diffusion constants, which is just $D_m \phi_n^{2/d}$, since the diffusion constant of the original pure system is defined to be unity. Now we find the ratio of conductances by multiplying this result by the ratio of the cross-sectional areas $\phi_n^{2/d}$ divided by the ratio of the lengths $\phi_n^{1/d}$. The result is $g/g_0 = \phi_n D_m$. This then provides the correct ratio of the conductivities of composite to pure system.

C. σ is finite and nonzero

Since all regions have a finite nonzero conductivity walkers can enter any region and spend time in any region. The mean time spent in each type of conductor is correctly achieved by our boundary conditions. Hence the ratio of conductivities equals the ratio of the diffusion constants.

V. SIMULATION RESULTS

A. Two-dimensional models

1. *Nonconducting disks*

Results in this case, sometimes called the "Swiss cheese" model, have already been published,[13] and are reproduced in Fig. 1 along with the EMT approximations and an interpolation formula due to Xia and Thorpe.[18] The horizontal axis is ϕ_1, which is the porosity or fraction of conducting region between the disks. The interpolation formula incorporates the correct behavior near $\phi_1 = 1$, the correct percolation threshold $\phi_1^{(c)} \approx 0.33$, and the expected lattice critical exponent[19,20] $t \approx 1.3$ defined by $\sigma_c \sim (\phi_1 - \phi_1^{(c)})^t$ near the threshold. In two dimensions it is expected that the transport exponents will be the same for the continuum and lattice cases. The resulting interpolation formula is

$$\sigma_c = \left[1 - m\phi_2/t - \phi_2^2 \frac{t - m(1 - \phi_1^{(c)})}{t(1 - \phi_1^{(c)})^2} \right]^t , \tag{15}$$

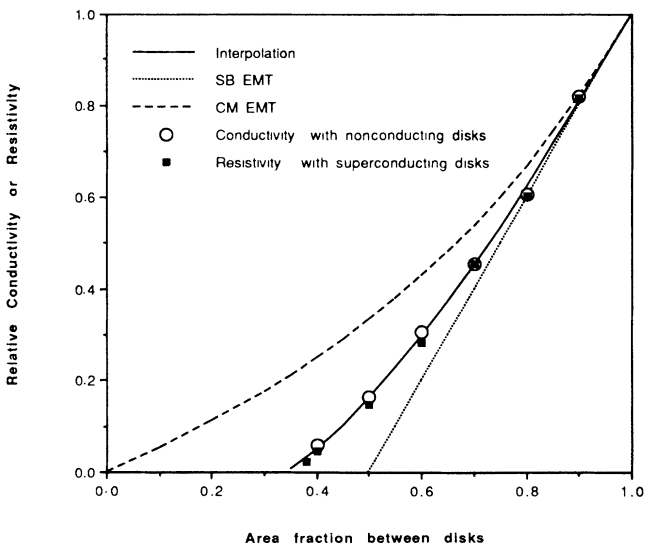

FIG. 1. Conductivity for a model composite of random overlapping nonconducting disks embedded in a medium of unit conductivity material and resistivity of superconducting disks in the same medium. The horizontal axis is the porosity or area fraction of the unit conductivity material ϕ_1. The curves are the symmetric Bruggemann (SB) and Clausius Mossotti (CM) effective medium theories and an interpolation formula due to Xia and Thorpe (Ref. 18).

where m is the initial slope determined by the EMT approximations and $\phi_2=1-\phi_1$. As can be seen the interpolation formula works very well, but the EMT approximations are not very good.

At the percolation threshold we can also compute the anomalous diffusion exponent. We expect that for walks diffusing less than the correlation length the mean-square displacement should go as

$$\langle r^2\rangle \sim t^{\alpha}, \tag{16}$$

where scaling arguments[16] (including walks on finite as well as infinite clusters) lead to

$$\alpha=(2\nu-\beta)/(2\nu+t-2\beta). \tag{17}$$

Using the known 2d lattice results $\beta=\frac{5}{36}$ and $\nu=\frac{4}{3}$, we expect $\alpha\approx 0.69$. At $\phi_1=0.33$ we found $\alpha=0.70\pm 0.03$ for a range of time from 1 to 1000.

2. *Superconducting disks*

In two dimensions there is a duality relation between a mixture of superconducting regions embedded in a medium of unit conductivity and nonconducting regions embedded in a medium of unit conductivity. It can be proven that if the geometry is identical then the resistivity of the former composite equals the conductivity of the latter composite.[21] Thus in Fig. 1 we have plotted the resistivity of the superconducting disk composite on the same graph as the nonconducting disk composite. As can be seen the results agree quite well. The advantage of using the superconducting disks to do the problem is that the walkers cover a much larger fraction of the configuration space than they would with nonconducting disks. Thus we expect to sample the geometry better. The disadvantage is that we may not use the method once there exists clusters of disks spanning our system, and periodic boundary conditions are ambiguous so we do not use them. Instead we start our walkers at the center of our system and consider only times such that no walker has reached the edge. We were able to go down to $\phi_1=0.38$ and a time equal to 5 with a simulation cell of size 300×300. Even though the time here seems very short, the walkers can go a long distance by hopping along the superconducting clusters, and can very effectively sample the geometry.

3. *Disks with finite conductivity*

We considered the cases where the disk conductivity is $h=2$, 5, and 10. The results are shown in Fig. 2 along with the EMT approximations. We note that, in general, our results typically fall between the SB and CM approximations. Also, we note that there is not yet any hint that as h becomes large, σ_c will diverge at $\phi_1^{(c)}$.

B. Three-dimensional models

1. *Nonconducting spheres*

Results are shown in Fig. 3 along with the EMT approximations and two interpolation formulas. The percolation threshold is about 3% as found by previous workers and is consistent with experimental results on brine filled porous rocks, which are known to conduct down to close to zero porosity. This effect is known as Archie's law.[22] Our results are also similar to those found by Schwartz and Banavar[4] for the grain consolidation model.

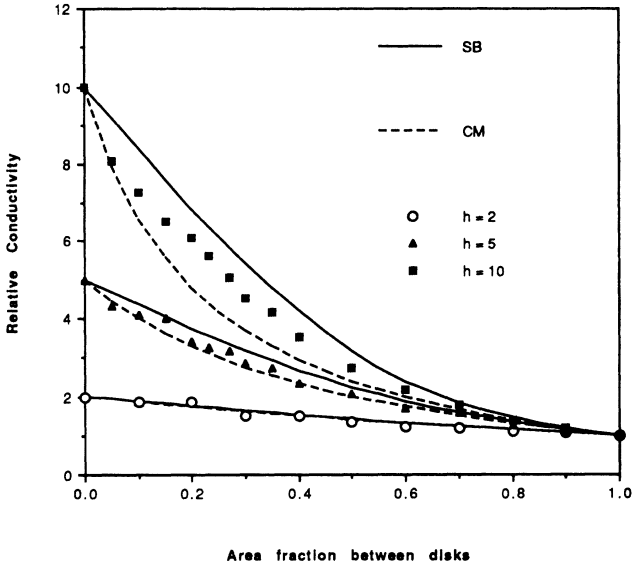

FIG. 2. Conductivity for a model composite of random overlapping disks with conductivity h embedded in a medium of unit conductivity of area fraction ϕ_1. Also shown are the SB and CM effective medium theories.

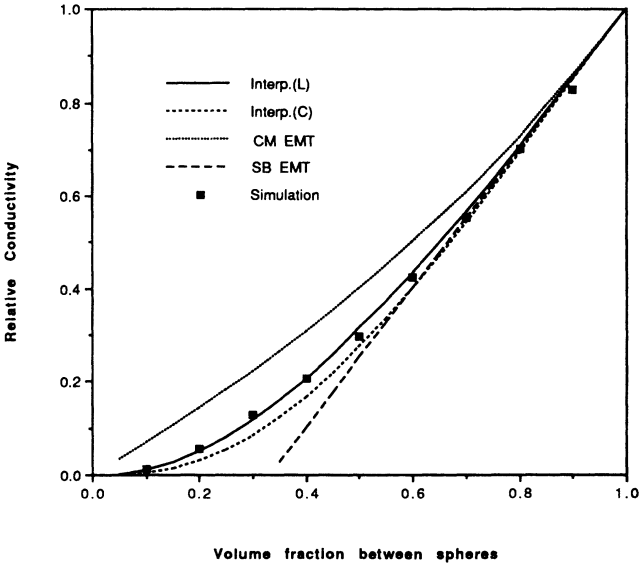

FIG. 3. Conductivity for a model composite of random overlapping nonconducting spheres embedded in a medium of unit conductivity of volume fraction ϕ_1. The curves are the SB and CM effective medium theories and two interpolation formulas Interp. (L) based on lattice exponents and Interp. (C) based on continuum exponents.

The random overlapping sphere model is one example of a three-dimensional model with narrow conducting necks which are expected to lead to a change in the transport exponents from their lattice values. The first interpolation formula uses the lattice value $t_{\rm lat} \approx 1.9$, and the second the value expected theoretically for a continuum $t_{\rm cont} = t_{\rm lat} + 0.5 \approx 2.4$.[8] The lattice interpolation formula actually seems to fit the data better. This is probably a coincidence, although it may indicate that the narrow necks responsible for the difference in the continuum exponents are only measurable at porosities very close to the threshold.

We have tried very hard to estimate t from the anamolous diffusion exponent α. Using the lattice values $\nu \approx 0.89$ (Ref. 23) and $\beta \approx 0.42$,[24] one predicts that $\alpha \approx 0.48$ for the lattice and $\alpha \approx 0.41$ for the continuum. Our results at the percolation threshold[25] $\phi_1 = 0.032$ for 2000 walkers averaged over a time of 2000 are $\alpha \approx 0.45 \pm 0.04$. Thus we are unable to distinquish between the two possibilities. We estimate at least another order of magnitude in running time is needed to pin down the anamolous diffusion exponent. (The present results for this data point took about one month of CPU time on a SUN 3/75 computer.)

2. *Nonconducting region between spheres*

Now we consider the case where the spheres are conducting, but the region between them is not conducting. Thus the walkers move only on the spheres. If we continue to call the volume fraction of the spheres ϕ_2 and the volume fraction of the background ϕ_1 then the percolation threshold is at about $\phi_1^{(c)} = 0.71$.[26] In Fig. 4 we show the random-walk results along with EMT approximations. Note that in this case the EMT do not have the correct behavior for a low density of zero conductivity material. This is no doubt because the geometry of space between spheres is very complicated and does not at all approximate nice, simple, widely separated spheres.

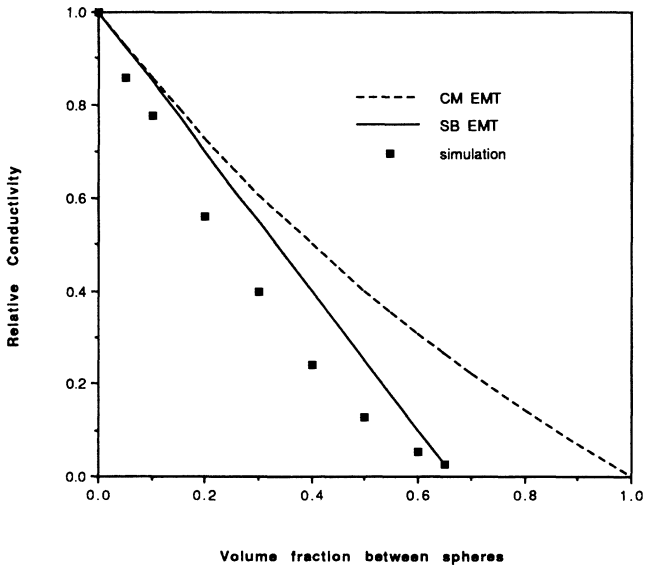

FIG. 4. Conductivity for a model composite of random overlapping unit conductivity spheres embedded in a nonconducting medium of volume fraction ϕ_1. The curves are the SB and CM effective medium theories.

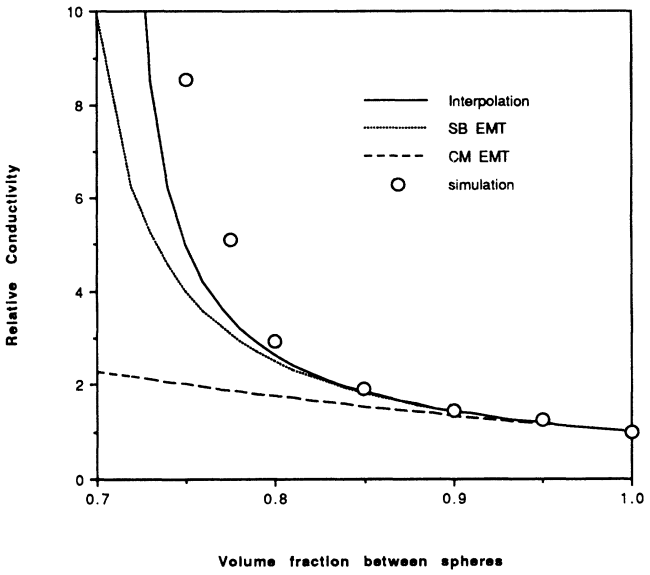

FIG. 5. Conductivity for a model composite of random overlapping superconducting spheres embedded in a medium of unit conductivity of volume fraction ϕ_1. The curves are the SB and CM effective medium theories and an interpolation formula.

We also measured the anomalous diffusion coefficient and found $\alpha = 0.51 \pm 0.03$ for a walk time of 1000. This is consistent with the lattice result, ≈ 0.48, which is expected since the narrow conducting necks, which are responsible for changing the conductivity exponent from the lattice result, do not exist in this 3D model.

3. *Superconducting Spheres*

Figure 5 shows the results for superconducting spheres. These data were obtained using a $60 \times 60 \times 60$ simulation cell with the walkers starting at the center of the cell. Because of the small cell size, we cannot obtain results close to the percolation threshold and we could only allow the walkers to walk for times between 20 at $\phi_1 = 0.95$ and 2.4 at $\phi_1 = 0.75$. The interpolation formula is based on the same idea as before, except here we used the resistivity exponent defined by $\rho \sim (\phi_1 - \phi_1^{(c)})^s$, where lattice calculations give $s \approx 0.76$.[19] The most important feature of our results is that we are able to see the beginning of the divergent behavior. Currently, work is in progress to look at a similar system where the spheres cannot overlap and configurations are generated by Monte Carlo simulations for dense liquids.

VI. SUMMARY

In this paper we have shown that the random-walk algorithm can be used successfully to determine the conductivity of a wide variety of model composite systems. Using on the order of 1000 walkers, one can expect to ob-

tain results at about the 5% accuracy level. The advantage of our method is that it does not rely in any way on an underlying lattice. Given a particular geometry the major sources of approximation are the finite number of walkers, the finite size moves made at the boundary between two materials, and the limited time during which the walkers move. Within our statistical accuracy, changing the parameters that control these limitations by a factor of 2 or 3 does not change the final results. Thus, at least another order of magnitude in numerical effort is needed to significantly reduce the statistical uncertainty of our results.

The method used in this paper should be applicable to any continuum model where it is possible to determine the boundaries between the regions of different conductivity. The method can also be used for more than two components, and in any dimension.

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