Pair connectedness and mean cluster size for continuum-percolation models: Computer-simulation results

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We devise a new algorithm to obtain the pair-connectedness function $P(r)$ for continuum-percolation models from computer simulations. It is shown to converge rapidly to the infinite-system limit, even near the percolation threshold, thus providing accurate estimates of $P(r)$ for a wide range of densities. We specifically consider an interpenetrable-particle model (referred to as the penetrable-concentric-shell model) in which D-dimensional spheres ($D = 2$ or 3) of diameter σ are distributed with an arbitrary degree of impenetrability parameter λ, $0 \leqslant \lambda \leqslant 1$. Pairs of particles are taken to be "connected" when the interparticle separation is less than σ. The theoretical results of Xu and Stell for $P(r)$ in the case of fully penetrable spheres ($\lambda = 0$) are shown to be in excellent agreement with our simulations. We also compute the mean cluster size as a function of density and λ for the case of 2D, and, from these data, estimate the respective percolation thresholds.

I. INTRODUCTION

The subject of physical clustering and percolation in "continuum" (off-lattice) models of random media has been receiving considerable attention in recent years.[1-12] Continuum-percolation models, although less tractable than their lattice counterparts, are better able to capture the essential physical aspects of real systems. Examples of phenomena in which continuum-percolation models and concepts are expected to be useful include transport and mechanical properties of fluid-saturated porous media,[13] composite materials,[14] microemulsions,[6] gelation,[15] conductor–insulator transition in liquid metals,[16] and the structure of liquid water.[8]

A typical continuum model consists of a system of generally interacting particles at number density ρ. One establishes a criterion for "bound" and "unbound" pairs, as first suggested by Hill,[17] in order to define the physical clusters. From this formulation of physical clusters, one can then obtain the pair-connectedness function $P(\mathbf{r}_1,\mathbf{r}_2)$[1]: a quantity of fundamental importance. The quantity $\rho^2 P(\mathbf{r}_1,\mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2$ represents the probability of finding two particles centered in volume elements $d\mathbf{r}_1$ and $d\mathbf{r}_2$ about $\mathbf{r}_1$ and $\mathbf{r}_2$, respectively, and are physically connected, i.e., belong to the same cluster. For statistically isotropic media (the subject of the present study), the pair-connectedness function just depends upon the relative distance $r = |\mathbf{r}_1 - \mathbf{r}_2|$, i.e., $P(\mathbf{r}_1,\mathbf{r}_2) = P(r)$. Given $P(r)$, one can then determine the mean cluster size S from the relation[1]

$$S = 1 + \rho \int P(r) d\mathbf{r}. \qquad (1)$$

At the percolation density ρ_c, the mean cluster size of course becomes infinite.

Theoretical techniques used to determine $P(r)$ focus on solving the connectedness Ornstein–Zernike integral equation.[1,4,5,8,12] Closure of the integral equation requires one to employ an approximation for the "direct" connectedness function. A commonly employed closure is the Percus–Yevick approximation. This was first used by Chiew and Glandt[4] to study permeable spheres. DeSimone *et al.*[8] employed this approximation to study the penetrable-concentric-shell (PCS) model introduced by Torquato.[18] More recently, Xu and Stell[12] solved the connectedness Ornstein–Zernike equation using a generalized-mean-spherical approximation for the model of fully penetrable (randomly centered) spheres.

Until the pioneering papers of Seaton and Glandt[9] and of Sevick *et al.*,[10] computer simulations of $P(r)$ for continuum-percolation models were nonexistent and hence integral-equation theories could not be tested. Seaton and Glandt obtained $P(r)$ for the "sticky-sphere" model. Sevick *et al.* measured $P(r)$ for the 3D PCS model. They also obtained the cluster size distributions and mean cluster size for this model.

In this study, we develop, among other things, a new algorithm to obtain $P(r)$ from computer simulations for continuum-percolation models. Specifically, we consider the D-dimensional PCS model (where $D = 2$ and 3). In the PCS model[18] (depicted in Fig. 1), each sphere (disk) of diameter σ is composed of an impenetrable core of diameter $\lambda\sigma$, encompassed by a perfectly penetrable-concentric shell of thickness $(1-\lambda)\sigma/2$. The extreme limits of the impenetrability parameter $\lambda = 0$ and 1 correspond to the cases of fully penetrable and totally impenetrable particles, respectively. DeSimone *et al.* have noted that the PCS model may serve as a useful first step in studying percolation in liquid water and

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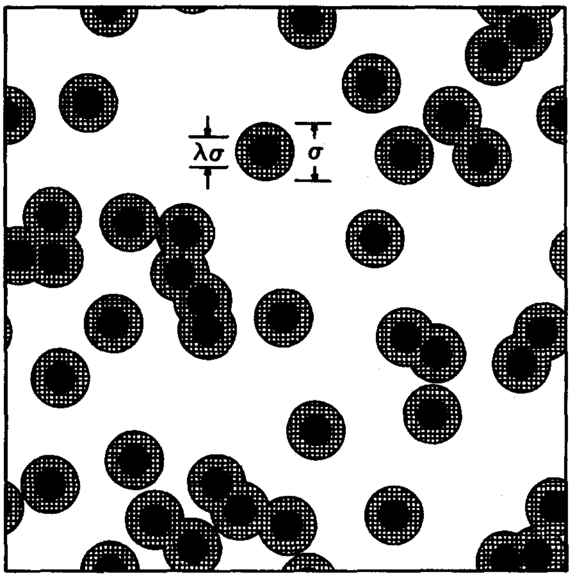

FIG. 1. A computer-generated realization of a distribution of disks of radius $\sigma/2$ (shaded region) in the PCS model. The disks have a hard core of diameter $\lambda\sigma$ indicated by the smaller, black circular region. Here $\lambda = 0.5$ and volume fraction of disks is approximately 0.3.

in liquid metals. Our results for $P(r)$ in the 3D case are compared to the data of Sevick *et al.* and it is shown that our algorithm converges to the infinite-system behavior more rapidly than their results do. Our 2D data for $P(r)$ are new. Moreover, in the 2D case, we obtain the mean cluster size S as a function of density (and volume fraction) for fixed λ and, from it, estimate the percolation thresholds for the selected values of λ.

In Sec. II, we describe the simulation technique we develop to determine $P(r)$ and S. In Sec. III, we report our results for these quantities for the D-dimensional PCS model ($D = 2$ and 3) as a function of the density and impenetrability parameter λ. There we compare our data to previous simulation and theoretical results. Finally in Sec. IV, we make some concluding remarks.

II. SIMULATION PROCEDURE

Obtaining statistical measures, such as the pair-connectedness function and mean cluster size, from computer simulations is a two-step process. First, one must generate realizations of the random medium. Second, one samples each realization for the desired quantity and then averages over a sufficiently large number of realizations. In the ensuing discussion, we describe the details of our simulation methods to obtain the aforementioned cluster measures for equilibrium distributions of identical disks or spheres in the PCS model.[18]

Consider each D-dimensional sphere to have a diameter σ and an inner impenetrable core of diameter $\lambda\sigma$. In order to generate random realizations for fixed λ and reduced number density $\eta = \rho\pi\sigma^D/[D(D-1)]$, we employ a conventional Metropolis algorithm.[19] Particles are initially placed in a cubical cell of volume L^D on the sites of a regular array (square and simple cubic lattices for $D = 2$ and 3, respectively). The cell is surrounded by periodic images of itself. Each particle is then moved by a small distance to a new position which is accepted or rejected according to whether or not the inner hard cores overlap. This process is repeated many times until equilibrium is achieved. Each of our simulation consists of 5000–20 000 moves per particle, the first 200–400 of which were discarded before sampling the equilibrium properties. Realizations were selected every 10–20 moves per particle.

Before computing $P(r)$ and S, one must first make use of an algorithm which distinguishes the various clusters in the system. By definition, two particles are assumed to be "directly" connected if their interparticle distance is less than σ. Pairs of particles may be "indirectly" connected, however, i.e., pairs can be connected through chains of other particles. Existing cluster-counting algorithms which can distinguish such clusters include the "cluster-labeling" method[20] and the "connectivity-matrix" method.[10] The former was originally developed for lattice percolation[20] and subsequently adapted for continuum percolation.[3] The latter was originally developed for continuum percolation[10] but can be applied to lattice percolation as well. In this study, we employ a modified cluster labeling method.

A. Pair-connectedness function

Computing the pair-connectedness function is, in principle, straightforward. First, one constructs concentric shells of radii

$$r_n = n\Delta r, \quad n = 1,2,3,... \tag{2}$$

up to $r = L/2$ around each particle in the system (where Δr is a distance which is small compared to σ). One then counts the number of particles in each shell which are connected to each of the central particles. The pair-connectedness function $P(r)$ is readily obtained from the number of connected particles for each n. The most subtle aspect of measuring $P(r)$ from computer simulations is extracting infinite-system behavior from a finite-sized experiment.

In order to minimize the effect of system size in simulations, one typically surrounds the central cell with replicas of itself.[21] Cluster identification depends upon the type of boundary conditions employed. This is a crucial point since determination of which particles are members of the same cluster will affect measurements of $P(r)$. Previous investigators[9,10] in their 3D simulations utilized what we term "simple" periodic boundary conditions over the central cell. The essence of our technique to compute $P(r)$ involves the application of "free boundary conditions" over the central and replicating cells. It shall be shown that our method leads to a pair-connectedness function which converges to the infinite-system behavior more rapidly than previous techniques which employ the straightforward use of periodic boundary conditions.

In order to illustrate these two different techniques, consider a 2D realization shown in Fig. 2. Figure 2(a) shows clusters as obtained by the use of periodic boundary conditions over the central cell.[9,10] Figure 2(b) depicts clusters as obtained by the use of free boundary conditions over central and replicating cells. Clearly, even though the same realiza-

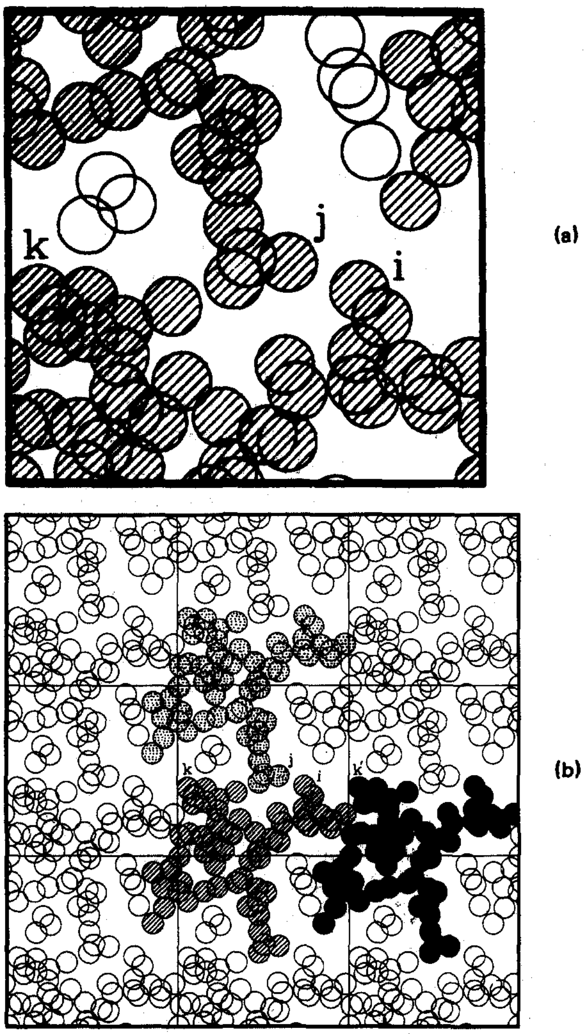

FIG. 2. A computer generated 2D realization for the penetrable-concentric-shell (PCS) model for $\lambda = 0.3$ and $\eta = 0.81$: (a) central cell in which clusters are obtained by the use of periodic boundary conditions over the central cell; (b) central and replicating cells in which clusters are obtained by the use of free boundary conditions over the central and replicating cells.

tion is involved, cluster identification is different. Consider two specific cases: (i) the particle pairs i and j and (ii) the particle pairs i and k. In case (i) ($r<L/2$), the pairs of particles are in the same cluster in Fig. 2(a), while in Fig. 2(b) they are not in the same cluster. In case (ii) the particles i and k are in the same cluster; however, since the interparticle distance $r>L/2$, this pair is not counted at all as an event in determining $P(r)$.[22] Instead, one must consider the "image" particle k', assuming particle i is the central particle. Clearly, particles i and k' are in different clusters. The two examples (i) and (ii) were assumed to contribute to $P(r)$ in Ref. 10 [in case (ii), particles i and k' were taken to be connected because particles i and k are connected; they use the distance between i and k']. Thus, the $P(r)$ presented in Ref. 10 is, in general, overestimated (especially as the threshold is approached from below), as shall be shown in the subsequent section. In the present work, the pairs i and j, and i and k' are not considered to be connected to one another. Our technique to obtain $P(r)$ shall be shown to converge to the infinite-system behavior relatively rapidly. Of course the two different methods become identical and "error-free" in the limit of an infinite system. Application of free boundary conditions over the central and replicating cells requires considerable computer time; it is essential, however, to do so in order to appreciably reduce finite-size errors in $P(r)$. It should be noted that the "connectivity-matrix" method[10] for cluster counting can be modified to incorporate the free boundary conditions employed in this study, albeit at the expense of additional computer time.

B. Mean cluster size and percolation threshold

The mean cluster size S is alternatively defined as the second moment of the cluster size distribution normalized by the first moment,[1]

$$S=\frac{\Sigma_s s^2 n_s}{\Sigma_s s n_s}, \tag{3}$$

where n_s is the mean number of clusters of size s. We employ this definition to compute S from our simulation as opposed to the alternative definition in terms of $P(r)$, Eq. (1). Unlike the determination of $P(r)$, the mean number of clusters of size s, n_s, and hence S, will be the same for both periodic boundary conditions applied to the central cell [cf. Fig. 2(a)] and free boundary conditions applied to the central and replicating cells [cf. Fig. 2(b)]. Since the former boundary condition (used in Ref. 10) is much less computer intensive, it is the one we utilize to compute S. Because Sevick *et al.* have already accurately determined S for the 3D PCS model, we shall present results for S only for the 2D case. In light of the fact that the mean cluster size S becomes infinite at $\eta=\eta_c$, we can estimate η_c by extrapolating the data for the inverse mean cluster size as a function of η to the $S^{-1}\to 0$ limit.

III. RESULTS AND DISCUSSIONS

We have carried out a number of simulations for the PCS model for both $D=2$ and 3. Two values of the impenetrability parameter λ were selected ($\lambda=0$ and 2/3) for $D=3$ in order to compare our results for $P(r)$ (which were obtained using the new algorithm described in Sec. II) with previous data reported by Sevick *et al.*[23] and with the theoretical results of Xu and Stell.[12] For $D=2$, we present new results for $P(r)$ at $\lambda=0$, 0.5, and 0.8 and for S at $\lambda=0$, 0.5, 0.8, and 0.9. In order to study the dependence of our results upon the size of system, we have carried our simulations with 64, 125, 216, and 512 particles for $D=3$ and with 100, 225, 400, and 625 particles for $D=2$.

A. Pair-connectedness function

1. 3D media

Our results are compared to the previous simulations of Sevick *et al.*[10] and the accurate theoretical results recently obtained by Xu and Stell.[12] It shall be shown that this theory is in excellent agreement with our simulations.

Figures 3 and 4 show our 3D data for $P(r)$ at $\lambda=0$ and 2/3, respectively. For the purposes of comparison, we in-

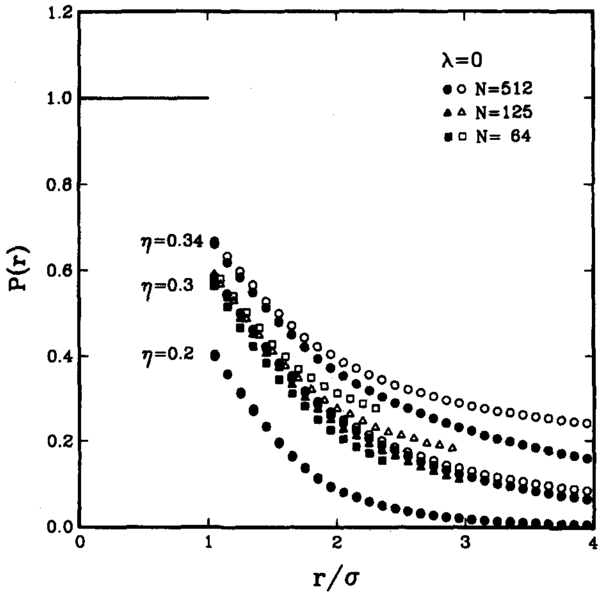

FIG. 3. The pair-connectedness function $P(r)$ from Monte Carlo simulations of the 3D PCS model for $\lambda = 0$ (fully penetrable spheres) with the number of particles $N = 64$, 125, and 512. Filled symbols are our data and unfilled symbols are from the algorithm of Ref. 10.

clude the corresponding results obtained in Ref. 10. The Sevick *et al.* data were generated by us using their boundary conditions and found to reproduce their results extremely well. The pair-connectedness function determined from these two algorithms differ in three key ways.

First, for a particular size of the system, our data always lie below those of Ref. 10. For smaller systems, such as $N = 64$ and 125, the data of Ref. 10 appear to have been generally overestimated. As N increases or η decreases, the difference between the two algorithms becomes unimportant. For $\eta = 0.2$ and $N = 512$, e.g., the two results nearly overlap one another, indicating that they provide consistent results. However, as $\eta \to \eta_c$, deviations are appreciable even for relatively large systems. For example, for $\eta = 0.3$, marked deviations are apparent already for $N = 512$. For $\eta = 0.34$ in the case of fully penetrable spheres,[24] i.e., very close to percolation point ($\eta_c = 0.35$, cf. Refs. 2, 4, 6, and 10), the data obtained from the algorithm of Ref. 10 deviate significantly from our results. It should be noted that Sevick *et al.* were already aware of the fact that they would need a large number of particles to diminish finite-size effects very near η_c.

Second, the dependence of $P(r)$ on the size of system in our study is opposite to the trend they found. Whereas $P(r)$ decreases as N increases in the work of Sevick *et al.*, our data indicates that finite-size effects result in a slight underestimation in $P(r)$. This trend found in Ref. 10 is inconsistent with their finding that the mean cluster size is underestimated (i.e., S increases as N increases). In other words, an underestimation (overestimation) of either $P(r)$ or S generally implies an underestimation (overestimation) of the other [cf. Eq. (1)].

Last, our data converge to the infinite-system limit more rapidly than the data of Ref. 10. For example, $P(r)$ for $\eta = 0.3$ and $N = 125$ is already very close to the data for 512-particle system in either the case $\lambda = 0$ or 2/3. The differences between the 216-particle system (not shown) and the 512-particle system essentially vanish. This is in contrast to the results of Ref. 10 where appreciable differences are still apparent between 216- and 512-particle system.

We now compare our 3D simulation results for $P(r)$ to theory. In particular, we consider comparing our data for $N = 512$ in the case of fully penetrable spheres ($\lambda = 0$) to the Percus–Yevick (PY) approximation obtained by Chiew and Glandt[4] and the generalized mean-spherical (GMS) approximation developed by Xu and Stell.[12] The agreement between our simulation and the PY approximation is in general poor except at very low densities. As the percolation threshold is approached from below, the PY approximation

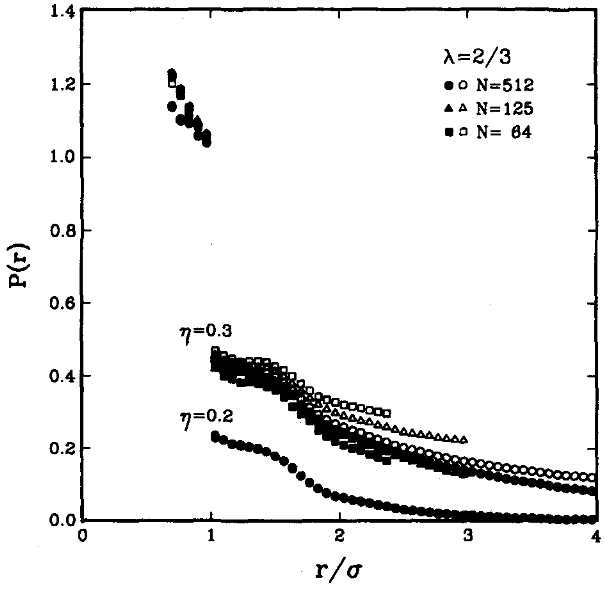

FIG. 4. As in Fig. 3, with $\lambda = 2/3$.

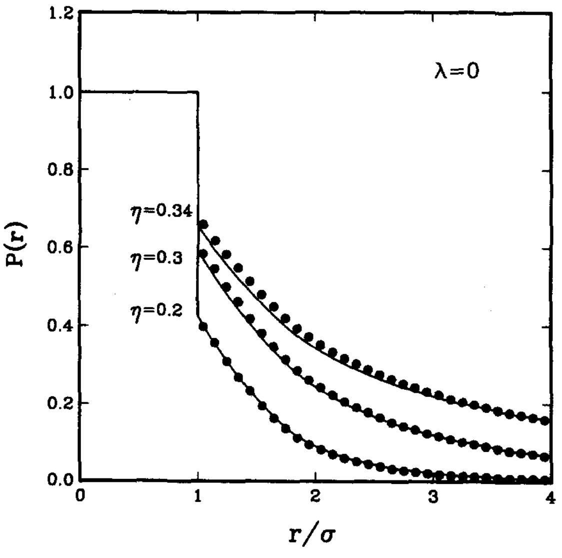

FIG. 5. The pair-connectedness function $P(r)$ for fully penetrable spheres ($\lambda = 0$). Our simulation results with 512 particles (circles); theoretical results of Ref. 12 (solid line).

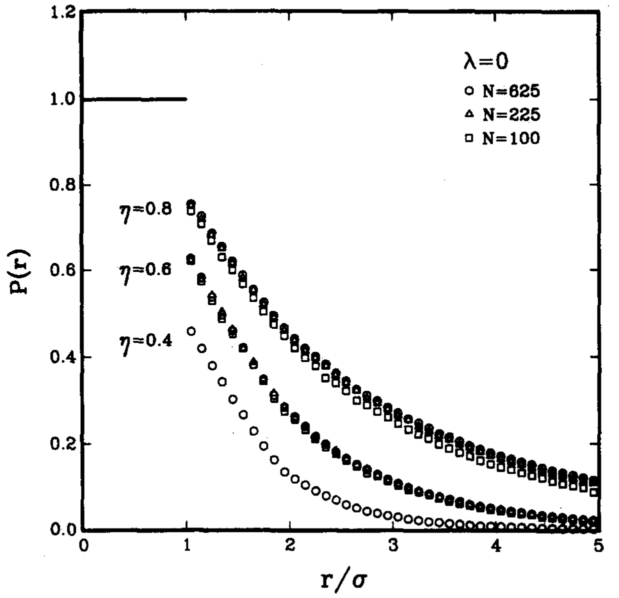

FIG. 6. The pair-connectedness function $P(r)$ from our Monte Carlo simulation of the 2D PCS model for $\lambda = 0$ (fully penetrable disks) with $N = 100$, 225, and 625.

considerably underestimates $P(r)$ and hence significantly overestimates the percolation transition. This conclusion regarding the accuracy of the PY approximation was first reached by Sevick *et al.* On the other hand, the GMS approximation agrees remarkably well with our simulations as shown in Fig. 5. As is well known, the PY approximation leaves out important cluster integrals from the exact expression for $P(r)$.[5,8,12] The GMS theory of Xu and Stell approximately but accurately corrects these deficiencies.

For $\lambda = 2/3$, the only theoretical result for $P(r)$ is the PY approximation obtained by DeSimone *et al.*[8] Again the PY approximation is seen to considerably underestimate $P(r)$, especially as $\eta \rightarrow \eta_c$. This conclusion was also first reached by Sevick *et al.* The critical density η_c is estimated to be about 0.33 (cf. Refs. 6 and 10).

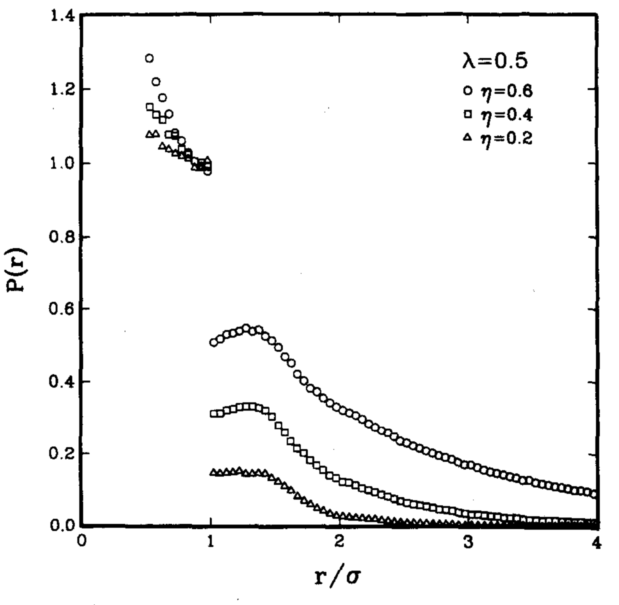

FIG. 7. As in Fig. 6, with $\lambda = 0.5$.

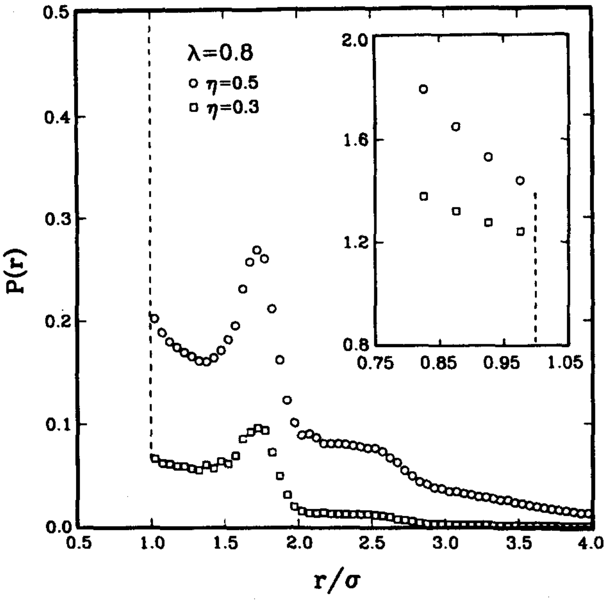

FIG. 8. As in Fig. 6, with $\lambda = 0.8$.

2. 2D media

Our simulation results for the 2D PCS model are new. In Figs. 6–8, we present data for $P(r)$ at $\lambda = 0, 0.5$, and 0.8, respectively. For the case $\lambda = 0$, data are given for various system sizes. For $\eta = 0.6$, the data depend weakly on system size, i.e., results for $N = 100$, 225, and 625 are not very different from one another. We find, as in the 3D case, that $P(r)$ is not very sensitive to system size provided that the system is not too small ($N < 100$).

For $\lambda = 0.5$ and 0.8, the effect of system size is similar to the case of $\lambda = 0$, and thus we present $P(r)$ in Figs. 7 and 8 for $N = 625$ only. In the case of fully penetrable disks, $P(r)$

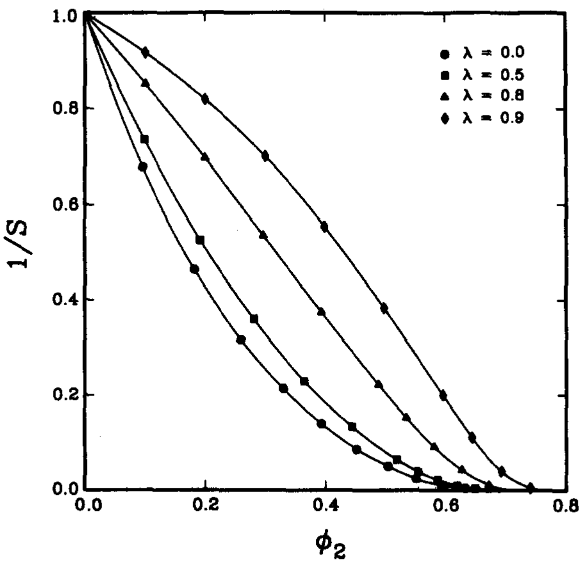

FIG. 9. The inverse mean cluster size S^{-1} as a function of the particle phase volume fraction ϕ_2 for $\lambda = 0$, 0.5, 0.8, and 1 in the 2D PCS model. The symbols are our simulation results and the solid lines are spline fits of the data.

TABLE I. The inverse mean cluster size S^{-1} for fully penetrable disks ($\lambda = 0$) extrapolated to the $N^{-1} \rightarrow 0$ limit. The error bounds are determined from the linear regression. From these data, the critical volume fraction of the particle phase ϕ_2^c is estimated to be 0.68.

η	ϕ_2	S^{-1}
0.10	0.0952	0.6770 ± 0.0011
0.20	0.1813	0.4644 ± 0.0025
0.30	0.2592	0.3152 ± 0.0010
0.40	0.3297	0.2125 ± 0.0012
0.50	0.3935	0.1382 ± 0.0014
0.60	0.4512	0.0854 ± 0.0008
0.70	0.5034	0.0497 ± 0.0008
0.80	0.5507	0.0229 ± 0.0006
0.90	0.5934	0.0094 ± 0.0002
1.00	0.6321	0.0028 ± 0.0002

TABLE III. The inverse mean cluster size S^{-1} for the PCS model for $\lambda = 0.8$ extrapolated to the $N^{-1} \rightarrow 0$ limit. The error bounds are determined from the linear regression. From these data, the critical volume fraction of the particle phase ϕ_2^c is estimated to be 0.71.

η	ϕ_2	S^{-1}
0.10	0.09972	0.8515 ± 0.0025
0.20	0.1987	0.6966 ± 0.0015
0.30	0.2966	0.5349 ± 0.0003
0.40	0.3933	0.3729 ± 0.0006
0.50	0.4880	0.2216 ± 0.0020
0.55	0.5344	0.1509 ± 0.0023
0.60	0.5803	0.0907 ± 0.0008
0.65	0.6262	0.0421 ± 0.0019
0.70	0.6721	0.0100 ± 0.0004

monotonically decreases with increasing r and shows behavior which is qualitatively similar to the corresponding 3D instance. For cases in which the impenetrable core is nonzero ($\lambda > 0$), the 2D pair-connectedness function again monotonically decreases with increasing r, provided that η is small. In such instances, however, if η is sufficiently large, the maximum in $P(r)$, for $r > \sigma$, occurs for r slightly greater than σ (say $r = r_1$) rather than at $r = \sigma$ as in the 3D analog. This implies that the probability of finding a connected particle at $r = r_1$ is greater than at $r = \sigma$ for $D = 2$. This is due simply to the topological difference between 2D and 3D.

B. Mean cluster size and percolation threshold

Here we present new results for S as a function of η and estimate η_c for the 2D PCS model. The data are obtained by extrapolating S^{-1} for various system sizes ($N = 100$, 225, 400, and 625) to the $N^{-1} \rightarrow 0$ limit. Following Sevick *et al.* for the 3D case, we estimate η_c by extrapolating these data to the $S^{-1} \rightarrow 0$ limit. Tables I–IV show our results for S^{-1} as a function of reduced number density η and of the volume fraction of particle phase ϕ_2. The volume fraction ϕ_2 was estimated for each λ and η from our previous work.[25,26] The error bounds given in the tables are determined from errors associated with the linear regression.

In general, the mean cluster size S depends upon the size of system for the entire range of volume fractions. For low concentrations, the slope of the extrapolation is smaller, indicating that finite-size effects are small. As η increases, the slopes increase significantly, demonstrating that finite-size effects are indeed important near the threshold, as expected. Thus, for any finite-sized system, η_c is considerably overestimated. This is expected since the mean cluster size in a simulation cannot be greater than the total number of particles, even for η close to η_c. In contrast, $S \rightarrow \infty$ as $\eta \rightarrow \eta_c$ for an infinite system. This underestimation of S implies that the pair-connectedness function will be underestimated, which is consistent with the findings of our simulations. In Fig. 9, our extrapolated results are plotted as a function of ϕ_2. Percolation points are estimated from this plot. For $\lambda = 0$, 0.5, 0.8, and 0.9, we find $\phi_2^c = 0.68$, 0.68, 0.71, and 0.75, respectively (where ϕ_2^c is the critical volume fraction of the particle phase).[26] The corresponding number densities are 1.13, 0.85, 0.75, and 0.76, respectively. These estimates are reasonably close to recent simulation results.[6]

The percolation thresholds could have been estimated using the scaling law for the mean cluster size and finite-size scaling analysis.[27] This procedure is more accurate than the one used in this study. We discovered, however, that for $\lambda > 0$ (i.e., for finite-sized hard cores) the region in which the scaling law holds is extremely narrow. Hence, it is quite nontrivial to accurately measure thresholds for $\lambda > 0$ using

TABLE II. The inverse mean cluster size S^{-1} for the PCS model for $\lambda = 0.5$ extrapolated to the $N^{-1} \rightarrow 0$ limit. The error bounds are determined from the linear regression. From these data, the critical volume fraction of the particle phase ϕ_2^c is estimated to be 0.68.

η	ϕ_2	S^{-1}
0.10	0.0979	0.7347 ± 0.0010
0.20	0.1915	0.5255 ± 0.0008
0.30	0.2806	0.3583 ± 0.0019
0.40	0.3649	0.2282 ± 0.0020
0.50	0.4441	0.1323 ± 0.0015
0.60	0.5183	0.0636 ± 0.0010
0.65	0.5583	0.0387 ± 0.0004
0.70	0.5867	0.0206 ± 0.0003
0.75	0.6187	0.0080 ± 0.0002
0.80	0.6493	0.0024 ± 0.0002

TABLE IV. The inverse mean cluster size S^{-1} for the PCS model for $\lambda = 0.9$ extrapolated to the $N^{-1} \rightarrow 0$ limit. The error bounds are determined from the linear regression. From these data, the critical volume fraction of the particle phase ϕ_2^c is estimated to be 0.75.

η	ϕ_2	S^{-1}
0.10	0.0999	0.9169 ± 0.0003
0.20	0.1998	0.8185 ± 0.0016
0.30	0.2994	0.6995 ± 0.0017
0.40	0.3986	0.5538 ± 0.0027
0.50	0.4973	0.3825 ± 0.0021
0.60	0.5950	0.1995 ± 0.0026
0.65	0.6437	0.1099 ± 0.0022
0.70	0.6925	0.0396 ± 0.0013
0.75	0.7413	0.0034 ± 0.0004

such an analysis. Elsewhere we have carried out such detailed calculations.[28]

IV. CONCLUDING REMARKS

We have devised a new algorithm which enables us to accurately measure the pair-connectedness function both for 2D and 3D continuum percolation models. Our results for $P(r)$ converge rapidly to the infinite-system limit, thus providing accurate numerical estimates of $P(r)$ for the PCS model in both 2D and 3D. We have also determined that the GMS approximation for $P(r)$ developed by Xu and Stell provides good estimates of it for the case of fully penetrable spheres. Lastly, we have presented numerical estimates of mean cluster size as a function of ϕ_2 for fixed λ in 2D, and used this information to estimate percolation thresholds for the selected values of λ.

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[1]A. Coniglio, U. De Angelis, A. Forlani, and G. Lauro, J. Phys. **10**, 219 (1977); A. Coniglio, U. DeAngelis, and A. Forlani, J. Phys. A **10**, 1123 (1077).

[2]S. W. Haan and R. Zwanzig, J. Phys. A **10**, 1547 (1977).

[3]E. T. Gawlinski and H. E. Stanley, J. Phys. A **14**, L291 (1981).

[4]Y. C. Chiew and E. D. Glandt, J. Phys. **16**, 2599 (1983).

[5]G. Stell, J. Phys. A **17**, L855 (1984); J. Xu and G. Stell, J. Chem. Phys. **89**, 2344 (1988).

[6]A. L. R. Bug, S. A. Safran, G. S. Grest, and I. Webman, Phys. Rev. Lett. **55**, 1986 (1985); A. L. R. Bug, S. A. Safran, G. S. Grest, and I. Webman, Phys. Rev. B **33**, 4716 (1986).

[7]A. J. Post and E. D. Glandt, J. Chem. Phys. **84**, 4585 (1986); A. K. Sen and S. Torquato, *ibid.* **89**, 3799 (1988).

[8]T. DeSimone, R. M. Stratt, and S. Demoulini, Phys. Rev. Lett. **56**, 1140 (1986); T. DeSimone, S. Demoulini, and R. M. Stratt, J. Chem. Phys. **85**, 391 (1986).

[9]N. A. Seaton and E. D. Glandt, J. Chem. Phys. **86**, 4668 (1987).

[10]E. M. Sevick, P. A. Monson, and J. M. Ottino, J. Chem. Phys. **88**, 1198 (1988).

[11]S. Torquato, J. D. Beasley, and Y. C. Chiew, J. Chem. Phys. **88**, 6540 (1988).

[12]J. Xu and G. Stell (preprint).

[13]S. Feng, B. I. Halperin, and P. N. Sen, Phys. Rev. B **35**, 197 (1987).

[14]S. Torquato, Rev. Chem. Eng. **4**, 151 (1987).

[15]A. Coniglio, H. E. Stanley, and W. Klein, Phys. Rev. B **25**, 6805 (1982).

[16]L. A. Turkevich and M. H. Cohen, J. Phys. Chem. **88**, 3751 (1984).

[17]T. L. Hill, J. Chem. Phys. **23**, 617 (1955).

[18]S. Torquato, J. Chem. Phys. **81**, 5079 (1984); **84**, 6345 (1986).

[19]N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. N. Teller, and E. Teller, J. Chem. Phys. **21**, 1087 (1953).

[20]J. Hoshen and R. Kopelman, Phys. Rev. B **28**, 5323 (1983).

[21]Use of free boundary conditions in a single unit cell for continuum-percolation models generally underestimates $P(r)$ and thus overestimates the percolation threshold. This effect is of course magnified as the threshold is approached from below.

[22]Periodicity requires that one sample $P(r)$ for $r<L/2$. As indicated earlier it is computationally convenient to construct concentric shells around each of the particles contained in the central cell. In most cases, the outermost shell of radius $L/2$ will lie in at least one replicating cell; in such instances one must consider the image particles which lie within a radius $L/2$ of the central particle and then sample for $P(r)$. In the present study, the particles i and k' (the image of k) of Fig. 2(b) are not considered to be connected.

[23]In Ref. 10, each particle was defined to have a hard core diameter σ' and a diameter $\sigma' + \lambda'$. Thus, λ' is related to λ by the relation $\lambda' = (1-\lambda)\sigma$. $\lambda' = \sigma'$ and $0.5\sigma'$ correspond to $\lambda = 0$ and 2/3, respectively, in our definition.

[24]Sevick *et al.* did not actually present $P(r)$ for $\eta = 0.34$ and $\lambda = 0$.

[25]S. B. Lee and S. Torquato, J. Chem. Phys. **89**, 3258 (1988).

[26]The relation between the particle-phase volume fraction ϕ_2 and the reduced number density η for fixed impenetrability parameter λ is, in general, nontrivial. For the extreme limits $\lambda = 0$ and 1, one has the simple relations $\phi_2 = 1 - \exp(-\eta)$ and $\phi_2 = \eta$, respectively. For intermediate λ, the relations are complex, and hence for such λ we employ the accurate simulation results reported by us in Ref. 25 to obtain ϕ_2 in terms of η and λ.

[27]D. Stauffer, *Introduction to Percolation Theory* (Taylor and Francis, London, 1985).

[28]S. B. Lee and S. Torquato (to be published).