The structure of hyperspherical fluids in various dimensions

Marvin Bishop[a)]
Department of Mathematics/Computer Science, Manhattan College, Manhattan College Parkway, Riverdale, New York 10471

Paula A. Whitlock and Dino Klein
Department of Computer and Information Sciences, Brooklyn College, 2900 Bedford Avenue, Brooklyn, New York 11210

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The structure of hard, hyperspherical fluids in dimension one, two, three, four, and five has been examined by calculating the pair correlation function using a Monte Carlo simulation. The pair correlation functions match known results in one, two, and three dimensions. The contact value of the pair correlation functions in all the different dimensions agrees well with the theory of Song, Mason, and Stratt [J. Phys. Chem. **93**, 6916 (1989)]. The decrease in ordering as the dimension is increased is readily apparent in the structure of the pair correlation function. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1848091]

I. INTRODUCTION

The structure of simple fluids is a problem of long standing interest. It is well known[1] that in the gaseous state there is little order and that particles are distributed at random whereas in the solid state the particles pack into long-ranged ordered crystals. The liquid state is characterized by an intermediate short-ranged order. The nature of this order is revealed by the pair correlation function $G(R)$.

This function measures the relative number of particle pairs at a distance R from the center of a reference particle. If N particles are contained in a region of size V (the volume in D dimensions), then the average number density ϱ is N/V. Let the center of one of the particles be fixed at the origin of the appropriate coordinate system. Then the probability of finding a second particle in a region $d\mathbf{R}$, keeping the one particle fixed at the origin, is given by

$$\varrho G(R) d\mathbf{R}. \tag{1}$$

Here, $\varrho G(R)$ behaves as a local density because if $G(R)$ is zero there are no particles in $d\mathbf{R}$ whereas if $G(R)$ is 1 the number of particles in $d\mathbf{R}$ is determined by the average density. The pair correlation function contains information about the system's density fluctuations. In a solid the particles are confined, except for small oscillations around the lattice sites. The change in the shape of the pair correlation function mirrors the underlying particle rearrangements. Indeed, the appearance of split peaks in $G(R)$ has been used as a signature of crystallization.

In two dimensions, for example, the close-packed lattice will be hexagonal.[2,3] If r_0 is the lattice spacing, which is determined by the density, there will be six nearest neighbors at a separation of r_0 from the reference particle, six second neighbors at a separation of $\sqrt{3}r_0$, and six third nearest neighbors at a separation of $2r_0$. Therefore, in the solid phase $G(R)$ will have three sharp peaks centered at these locations. As the density is lowered the particles will become less localized. Then the peak heights are lowered and the peak widths are broadened. Since the difference between $\sqrt{3}$ and 2 is relatively small, the second and third neighbor peaks will begin to merge together as the phase transition is approached. Above the transition point, in the liquid phase, the distinction between the second and third peak will completely disappear. In the gas phase the second neighbor peak displayed by the liquid phase will be gone. The first neighbor peak reflects the order that remains in the system; this order is caused only by the shape of the hard hypersphere potential. Thus, changes in the structure of the pair correlation function directly mirror the changes in the particle ordering as a system alters its state.

The pair correlation function can be calculated for any spatial dimension. As the dimension is increased, particles will be less hindered in their movement and therefore higher densities are required for localization. This localization behavior has been previously observed in analytic calculations of $G(R)$ in one dimension by Salsburg, Zwanzig, and Kirkwood[4] even though there is no crystallization phase in one-dimensional hard rod systems. Localization in two dimensions has already been described above. Alder and Wainwright[5] and Wood[6] observed a fluid-solid transition in two-dimensional hard disks by following the variation of pressure as a function of density along an isotherm. Alder and Wainwright[5] used molecular dynamics (MD) methods whereas Wood[6] employed Monte Carlo (MC) techniques. In three dimensions a phase transition was found for hard spheres by Alder and Wainwright[7] with MD and confirmed by Wood and Jacobson[8] with MC. More recently, four- and five-dimensional MD simulations of hard hyperspheres by Michels and Trappeniers[9] have reported a fluid-solid transition. The transition density (in standard reduced units) increases[10] from $\varrho \approx 0.88$ in two dimensions, to $\varrho \approx 0.95$ in three dimensions, to $\varrho \approx 1.0$ in four dimensions, and then to $\varrho \approx 1.19$ in five dimensions.

[a)]Electronic mail: marvin.bishop@manhattan.edu

TABLE I. Selected results in one, two, and three dimensions.

D	ρ	PreEq	PostEq	Maximum displacement	Acceptance ratio	$G(1)$	Theory[a]
1	0.20	8000	20 000	2.60	0.75	1.243±0.275	1.249
	0.40	8000	30 000	1.00	0.73	1.655±0.072	1.665
	0.60	8000	50 000	0.60	0.66	2.487±0.075	2.498
	0.80	8000	50 000	0.10	0.83	4.909±0.591	4.995
2	0.20	10 000	20 000	1.00	0.69	1.304±0.205	1.31
	0.40	10 000	20 000	0.30	0.72	1.832±0.040	1.80
	0.60	10 000	20 000	0.10	0.77	2.808±0.147	2.70
	0.80	10 000	20 000	0.08	0.61	5.059±0.689	4.62
3	0.20	5000	20 000	0.50	0.68	1.324±0.169	1.339
	0.40	5000	20 000	0.20	0.65	1.817±0.068	1.811
	0.60	5000	20 000	0.10	0.63	2.615±0.194	2.561
	0.80	5000	20 000	0.05	0.62	4.011±0.601	3.971

[a]In one dimension the theory result is Eq. (7), in two dimensions it is the PY results of Ref. 23, and in three dimensions it is the values reported by Barker and Henderson (Ref. 24).

This paper presents a systematic examination of the variation of $G(R)$ as the dimension is increased from one to five. Information about $G(R)$ allows one to calculate the equation of state and provides idealized models for testing the universality of theories in different dimensions.

II. METHOD

The number of particles, the number density of the state, and the dimensionality of the system of interest are preselected input parameters. These determine the side L of the system simulation hyperbox:

$$L = \left(\frac{N}{\varrho}\right)^{1/D}. \tag{2}$$

The hyperspheres are initially arranged in a lattice and are moved using the standard Metropolis Monte Carlo technique[11] to form an equilibrated fluid. However, for the case of hard hyperspheres the usual energy check becomes a simple test for overlaps since the pair potential of D dimensional hyperspheres with diameter σ separated by the D dimensional distance R is given by

$$U(R) = \begin{cases} \infty & R < \sigma \\ 0 & R \geqslant \sigma \end{cases}. \tag{3}$$

The computer simulation employs the standard system of reduced units in which the hypersphere diameter is set to be 1.0. It proceeds by attempting to move, in turn, each of the particles in the simulation hyperbox. One attempted move for each of the N hyperspheres is called a pass. Each particle is first moved a different random amount in each of the D directions within a test hyperbox. Thus, a new trial position is randomly selected from a hyperbox surrounding the current location of the center of mass of the hypersphere. If the attempted displacement is too large, the chance of an overlap with another particle will be great and too many moves will be rejected whereas if the displacement is too small, the simulation will not adequately sample phase space. If the new position is not accepted, the hypersphere remains at its current location. The acceptance ratio, the number of accepted moves divided by the number of total moves, is monitored. This ratio as well as the maximum magnitude of an allowed displacement for each density and dimension is listed in Tables I–III. The values of the maximum allowed displacement have been determined with short

TABLE II. Four-dimensional results.

ρ	PreEq	PostEq	Maximum displacement	Acceptance ratio	$G(1)$	LM[a] $G(1)$
0.05	3000	10 000	1.00	0.81	1.064±0.082	
0.10	3000	10 000	0.90	0.67	1.134±0.071	
0.20	3000	10 000	0.30	0.70	1.291±0.041	1.292
0.30	3000	10 000	0.20	0.66	1.473±0.031	1.476
0.40	3000	10 000	0.10	0.73	1.690±0.049	1.692
0.50	3000	10 000	0.10	0.64	1.943±0.054	1.947
0.60	3000	10 000	0.08	0.61	2.244±0.128	2.251
0.70	3000	10 000	0.05	0.65	2.604±0.223	2.617
0.80	3000	10 000	0.05	0.57	3.034±0.361	3.060
0.90	3000	10 000	0.05	0.48	3.552±0.612	3.590

[a]Reference 10.

TABLE III. Five-dimensional results.

ρ	PreEq	PostEq	Maximum displacement	Acceptance ratio	$G(1)$	LM[a] $G(1)$	PY[b,c] $G(1)$
0.05	1000	10 000	0.90	0.79	1.055±0.081		1.055
0.10	1000	10 000	0.80	0.64	1.114±0.064		1.113
0.20	1000	10 000	0.30	0.64	1.241±0.020	1.241	1.233
0.30	1000	10 000	0.18	0.63	1.379±0.042		1.363
0.40	1000	10 000	0.10	0.68	1.536±0.041	1.537	1.502
0.50	1000	10 000	0.09	0.62	1.708±0.042		1.654
0.60	1000	10 000	0.07	0.61	1.899±0.092	1.906	1.818
0.70	1000	10 000	0.05	0.63	2.112±0.131		1.997
0.80	1000	10 000	0.04	0.63	2.352±0.216	2.368	2.192
0.90	2000	10 000	0.03	0.64	2.619±0.324		2.405

[a]Reference 10.
[b]Reference 28.
[c]Reference 29.

runs. The move may or may not be accepted but is always counted in the averaging. Standard periodic boundary conditions[12] are employed in testing for overlaps and in maintaining a constant number of particles in the simulation hyperbox.

The inefficiencies of a "direct" algorithm which examines all possible $O(N^2)$ pair overlaps are greatly reduced by partitioning[13] the simulation hyperbox into subcells. After a new trial position is generated for the selected particle, a determination is made to detect whether or not the particle has moved out of its original cell. If it has, the possibility of overlaps with other particles in the new cell must be considered. If the selected particle is still in its original cell one needs to examine possible overlaps with particles in that cell and, depending upon the magnitude of the move, the neighboring cells as well. The cell dimensions are selected so that on average four particles are in each cell. Timing experiments indicate that the run times are decreased by approximately a factor of 6 over the times of a "direct" algorithm.

Since the successive positions of the hyperspheres are not independent, it takes many passes to converge from the initial state to an equilibrated one. Hence, some number of passes must be discarded; we refer to these discarded passes as the preequilibrium stage (PreEq in Tables I–III). Typically, on the order of 10^3-10^4 passes are needed in order to reach the equilibrated state. Then an additional ten to fifty thousand passes are generated (PostEq in Tables I–III) for analysis.

Even after the asymptotic regime is attained there is still serial correlation between each step in the generation process. Here, we have handled this problem by two methods. First, we sample $G(R)$ only at a save interval of ten passes. This procedure allows some of the serial correlation to dissipate from the previously sampled value of $G(R)$. Second, and considerably more effectively, statistically independent sets of simulations are carried out. These individual results are then averaged together.

The MC method is easy to implement using the small Web computing[14,15] (SWC) framework software. The SWC framework is a Master-Worker Multiple-Instruction, Multiple-Data (MIMD) parallel programming model implemented in Java. While the SWC system is intended to be used as Web-based collaborative software, it can also be utilized to run a multithreaded process on a symmetric multiprocessing (SMP) machine or to run distributed, independent processes on separate machines without the necessity for additional programming. All of these platforms were employed in the current simulations. Detailed comparisons of a C++ serial version of the code and the SWC parallel Java version have been previously reported upon.[16] It was found that all the calculated quantities agreed within the precision of the statistical error. Moreover, the SWC parallel version was considerably more efficient in both CPU and wall-clock time.

An order parameter[17] O, appropriate for a generalized closest packed lattice, was monitored to decide when the system had reached equilibrium:

$$O = \frac{\sum_{j=1}^{D}\sum_{i=1}^{N} \cos[4\pi X_i(j)\varrho^{1/D}]}{DN}. \tag{4}$$

Here, $X_i(j)$ is the jth position component of the ith hypersphere. The order parameter has a value of 1 for a completely ordered lattice and randomly oscillates about 0 when the system has equilibrated. At this point all preequilibrium passes were discarded and the accumulators for the estimators of interest were reinitialized. The simulation was continued until the statistical error of the result was satisfactory.

$G(R)$ is calculated on a grid with spacing ΔR by making a histogram of the number of pair separations as a function of separation distance. This count is normalized by dividing by two factors; one is the differential volume occupied by the pairs, $V_D[(R+\Delta R)^D - R^D]$, where V_D is related to the surface area of a D dimensional hypersphere,[18]

$$V_D = \frac{\pi^{D/2}}{\Gamma(1+D/2)}, \tag{5}$$

and Γ is the standard Gamma function. The other factor is the actual number density of pairs $0.5N(N-1)/V$ or $0.5\varrho(N-1)$.

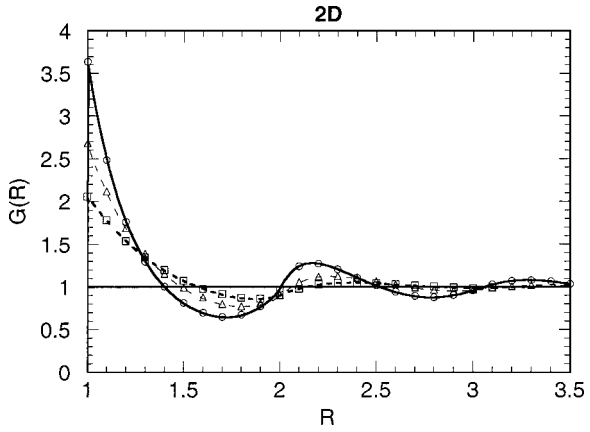

FIG. 1. Comparison of MC two-dimensional data to the results of Chae, Ree, and Ree. $\rho=0.462$….MC □; $\rho=0.577$---MC △; $\rho=0.693$—MC ○.

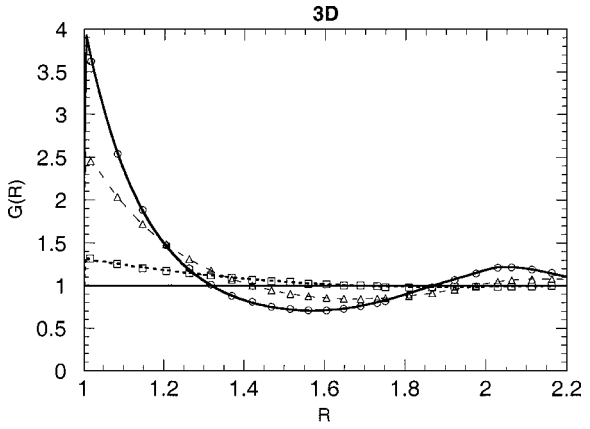

FIG. 2. Comparison of MC three-dimensional data to the results of Barker and Henderson. $\rho=0.20$….MC □; $\rho=0.60$---MC △; $\rho=0.80$—MC ○.

III. RESULTS

A. Comparison with known results in one, two, and three dimensions

The one-, two-, and three-dimensional studies have been used mainly to check the methods and computer codes since these results are well known in the literature. The major new findings are for four and five dimensions. In all the simulations we have used the shuffled, nested Weyl random number generator.[19,20] Empirical tests have shown that this generator gives reasonable behavior in parallel MC calculations.[20]

In one dimension Salsburg, Zwanzig, and Kirkwood[4] derived the exact expression for the pair correlation function in the thermodynamic limit ($N\rightarrow\infty$). Their equation predicts a contact value of $G(R)$:

$$G(1)=\frac{1}{1-\varrho}. \tag{6}$$

Bishop and Berne[21] extended the result of Salsburg, Zwanzig, and Kirkwood[4] to the case of periodic boundary conditions and obtained an N-dependent correction to $G(1)$,

$$G(1)=\frac{1-1/N}{1-\varrho}. \tag{7}$$

The one-dimensional hard rod system has been studied for a variety of densities from $\varrho=0.10$ to 0.8 where $N=1000$. The contact value of the pair correlation function has been determined by fitting a least-squares line to the first five data points for which the separation R is larger than 1.00. The equation of this line is then evaluated at the point $R=1.00$ to find the contact value. The error bar on the contract value has been determined by the error bar on the fitted coefficients of the straight line. This reported error also contains a statistical component in the calculation of $G(R)$ and is an upper bound to the actual error. The results for a selection of the densities examined are listed in Table I. When the MC values are compared to the exact values predicted by Eq. (7), one sees excellent agreement.

In two dimensions the MC simulations have been checked by comparison with the earlier work of Chae, Ree, and Ree[22] which used 90 or 208 particles. Here we have employed systems with 625 particles. Figure 1 presents a comparison of $G(R)$ determined by our parallel MC simulation with their data at densities of 0.462, 0.577, and 0.693. The agreement is excellent. We have also performed two-dimensional MC calculations for a range of densities of ϱ from 0.10 to 0.80.

The $G(1)$ values are compared to a numerical solution of the Percus–Yevick (PY) equation of Lado[23] in Table I. Note that the PY solution has been reported with only three significant figures. The agreement between the MC simulations and the PY solution is excellent until a density of about 0.6. Of course it is well known that the PY equation is not adequate at higher densities.

In three dimensions our MC pair correlation functions for $N=1000$ have been verified by comparing with the earlier MC data of Barker and Henderson[24] who used systems of 108 hard spheres. Figure 2 presents a comparison for three typical densities: 0.2, 0.6, and 0.8. Fine agreement is obtained. Our values of $G(1)$ are compared to the Barker and Henderson findings in Table I. The earlier work provides further validation of the current simulation approach.

B. Results in four and five dimensions

Our four-dimensional Monte Carlo calculations employed 4096 particles. Typical order parameters are presented for densities of $\rho=0.50$ and 0.80 in Fig. 3. This figure indicates that the lattice structure is erased after about 100

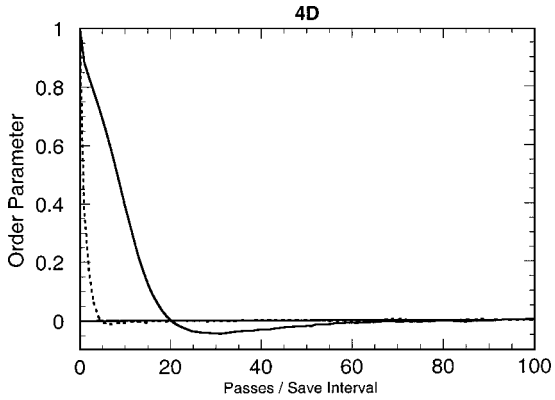

FIG. 3. The order parameter at $\rho=0.50$….and $\rho=0.80$—in four dimensions.

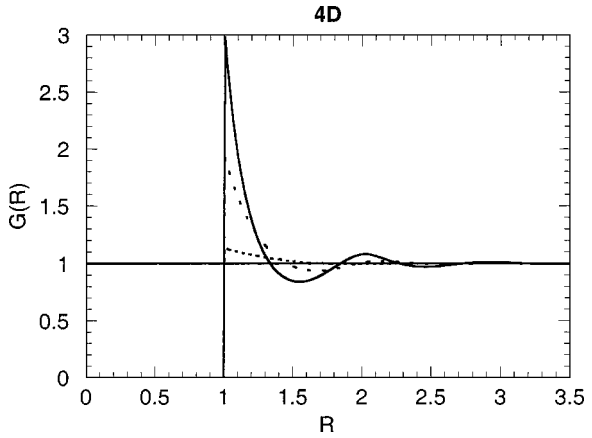

FIG. 4. The pair correlation function in four dimensions for $\rho=0.10$…., $\rho=0.50$----, and $\rho=0.80$—.

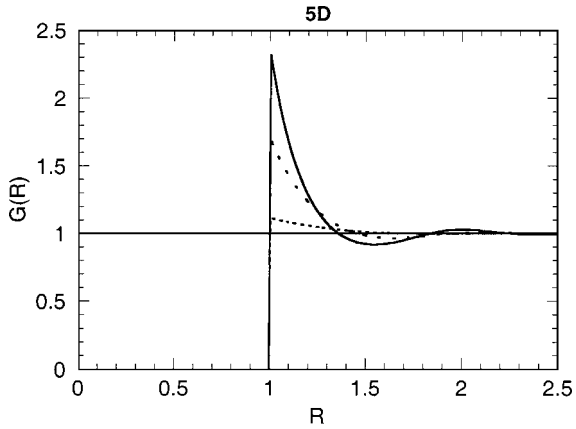

FIG. 6. The pair correlation function in five dimensions for $\rho=0.10$…., $\rho=0.50$----, and $\rho=0.80$—.

passes per save interval, even at the highest density. The $G(R)$ values for three different densities, $\rho=0.10$, 0.50, and 0.80, are presented in Fig. 4. Note the increased ordering as the density is increased. A measure of the statistical error in the average values of $G(R)$ at a density of 0.80 is shown in Fig. 5. This standard deviation corresponds to the scatter in the values of $G(R)$ obtained by averaging ten independent distributed simulations. These errors are extremely small.

The pair correlation function at contact can be obtained from the MD equation of state as reported by Luban and Michels.[10] The compressibility factor Z is defined as

$$Z=\frac{P\beta}{\varrho}, \tag{8}$$

where β is $1/k_BT$ (k_B is Boltzmann's constant and T is the absolute temperature), and P is the pressure. In the case of hard hyperspheres Z is independent of the temperature. The equation of state, Z as a function of ϱ, in all dimensions[25] is related to $G(1)$ by

$$Z=1+\varrho B_2 G(1). \tag{9}$$

Here, B_2 is the second virial coefficient. B_2 has the value of $\pi^2/4$ and $4\pi^2/15$ in four and five dimensions,[26] respectively.

The $G(R)$ MC values at contact are contained in Table II. The agreement with the MD calculations of Luban and Michels[10] is excellent. They used 648 particles and continued their simulations for at least 3.2×10^5 collisions after equilibration. They report an accuracy of about one part in 10^3. The equation of state data of Luban and Michels was confirmed by Gonzalez-Melchor, Alejandre, and Lopez de Haro[27] who also performed MD calculations with 648 particles as part of a study of mixtures of hyperspheres in four and five dimensions.

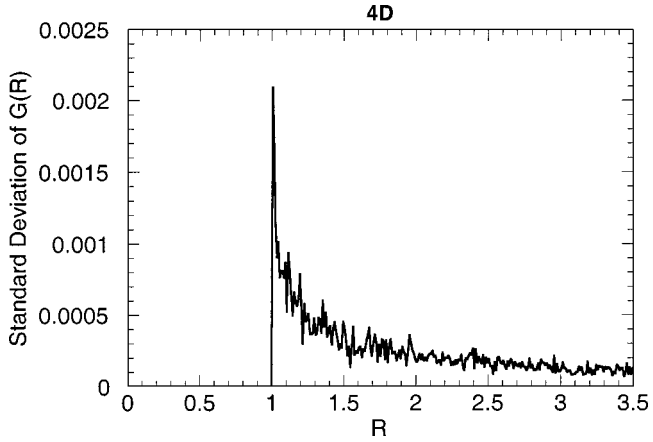

FIG. 5. The standard deviation of $G(R)$ in four dimensions at $\rho=0.80$.

In five dimensions N was equal to 7776 and $G(R)$ for three typical densities ($\rho=0.10$, 0.50, and 0.80) are presented in Fig. 6. Now hardly any ordering is visible, even at the highest densities. The statistical accuracy of $G(R)$ in five dimensions is similar to that observed in four dimensions.[16] The contact pair correlation function results are contained in Table III. We find excellent agreement with the MD simulations of Luban and Michels[10] who employed 512 particles and report errors of about one part in 10^3. Their results were also confirmed by Gonzalez-Melchor, Alejandre, and Lopez de Haro,[27] who used 512 and 3888 particles. The pair correlation function at contact has been obtained in closed form from an exact solution of the Percus–Yevick approximation in five dimensions by Freasier and Isbister[28] and by Leutheusser.[29] Two different expressions have been published. Freasier and Isbister[28] have

$$\eta=\pi^2\rho \tag{10}$$

with

$$G(1)=-\left[\frac{\alpha}{2}+\beta+\gamma\right]. \tag{11}$$

Here,

$$\alpha=\frac{-\left[\left(2\eta+\frac{\eta^2}{20}\right)\Delta+3\left(1+\frac{3\eta}{20}\right)\right]}{d}, \tag{12}$$

$$\beta=\frac{\left(\frac{\eta}{2}+\frac{\eta^2}{30}\right)\Delta+\frac{\eta}{4}}{d}, \tag{13}$$

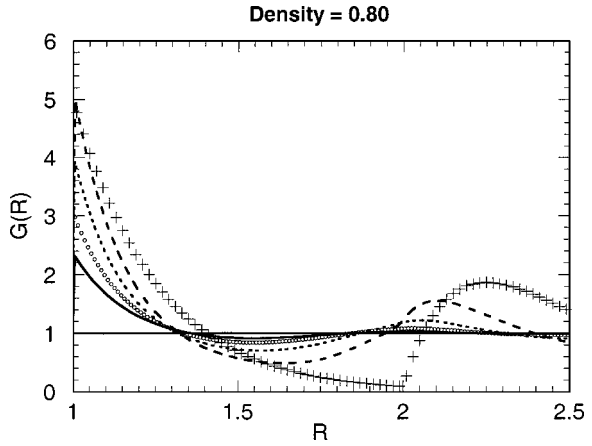

FIG. 7. A comparison of $G(R)$ at dimension one, +; two,----; three,….; four, ○; and five,—at $\rho=0.80$.

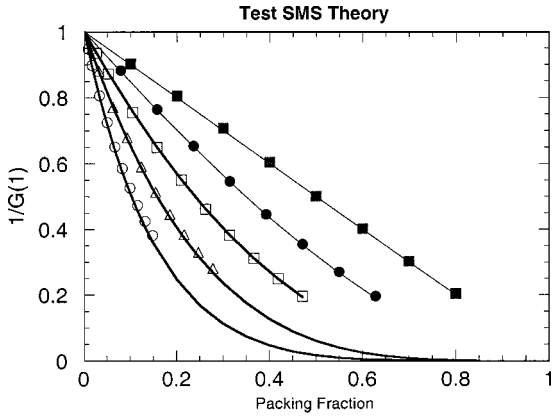

FIG. 8. Comparison of the MC data to the theory of Song, Mason, and Stratt (SMS) (Ref. 30). MC in dimension one, solid square; two, solid circle; three, □; four, △; and five, ○. Theory—.

$$\gamma = \frac{\left(-2+\frac{\eta}{5}\right)\Delta + 1}{2\left(1-\frac{\eta}{60}\right)}, \tag{14}$$

$$d = 1 - \frac{7\eta}{60} + \frac{\eta^2}{600}, \tag{15}$$

$$\Delta = \frac{-\left(1+\frac{\eta}{20}+\frac{7\eta^2}{1200}\right)+\left(1-\frac{\eta}{10}\right)\left(1+\frac{3\eta}{10}+\frac{\eta^2}{600}\right)^{1/2}}{\eta\left(1-\frac{\eta}{60}\right)^2}. \tag{16}$$

Leutheusser[29] gives the packing fraction as

$$\eta = \frac{\pi^2\rho}{60} \tag{17}$$

with

$$G(1) = \frac{-[1-33\eta-87\eta^2-6\eta^3-(1+18\eta+6\eta^2)^{3/2}]}{60\eta(1-\eta)^3}. \tag{18}$$

Both approaches give the same numerical values which are reported in Table III. When we compare the two-dimensional PY results in Table I with those in five dimensions, Table III, we notice that the PY approximation is valid at higher densities for higher dimensions. Moreover, the PY approximation is in excellent agreement at low to moderate densities with both the MD calculations of Luban and Michels[10] and the present MC simulations.

A comparison of the behaviors of the pair correlation functions in dimension one to five shows that the ordering of the particles is less pronounced, at a given density, as the dimension is increased. As illustrated in Fig. 7, this effect is particularly noticeable at higher dimensions. The well-defined second peak in one dimension indicates a nearly "solidlike" ordering which is not surprising given the confinement in one dimension. Note that there is still a well-defined second peak in two dimensions whereas that peak substantially decreases as the dimension increases from three to four and then to five.

Gonzalez-Melchor, Alejandre, and Lopez de Haro[27] have published MD $G(R)$ data in the solid phase for binary mixtures of hard hyperspheres in four and five dimensions. Their self-pair-correlation functions reveal the structure expected for the solid phase. Song, Mason, and Stratt[30] have also considered the pair correlation function at contact for arbitrary dimension and density. They have derived a theoretical equation, using mean field theory, for $G(1)$ based on the Carnahan–Starling equation of state:[31]

$$G(1) = \frac{1-\alpha\eta}{(1-\eta)^D}, \tag{19}$$

where η is the packing fraction

$$\eta = \frac{B_2\rho}{2^{D-1}} \tag{20}$$

and

$$\alpha = D - 2^{D-1}\frac{B_3}{B_2^2}. \tag{21}$$

Here, B_3 is the third virial coefficient. Figure 8 compares their predictions for $1/G(1)$ vs η with all of our MC simulation data. They have previously compared their predictions with Luban and Michels[10] and the closed form of PY. As would be anticipated our MC results confirm this previous investigation.

IV. CONCLUSION

A parallel Monte Carlo Java program has been developed which allows us to efficiently compute the pair correlation in any dimension. We obtain excellent agreement with previously published results in one to three dimensions and report new findings for four and five dimensions. The decrease in ordering as the dimension increases is readily apparent. This is a result of the hyperspheres being able to easily avoid each other at higher dimensions. The MC data also confirm the theory of Song, Mason, and Stratt.

We plan to extend these calculations to even higher dimensions and to investigate the equation of state of hard hyperspheres in detail.

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