# Computer investigation of long-range correlations and local order in random packings of spheres

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Random packings containing 8192 hard spheres in three dimensions have been built with an efficient computer algorithm for various packing fractions up to $c=0.643$, a value close to the upper limit $c_b \simeq 0.649$. Long-range correlations and local order have been investigated via the calculation of the two-point correlation function $g(r)$ and the Voronoi tessellation, respectively. The $g(r)$ curve exhibits large-$r$ damped oscillations characterized by a correlation length $\xi$ that increases with $c$ and whose extrapolation for $c>c_b$ diverges at $c_0=0.754$, which would be the volume fraction of an ideal icosahedral order. When they are extrapolated in the same manner, most of the geometrical characteristics of the Voronoi cells converge to their corresponding values for the perfect dodecahedron circumscribed around a sphere. [S1063-651X(96)01012-4]

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## I. INTRODUCTION

Random packings of hard spheres in three dimensions have been used throughout the past decades to represent the structure of liquids, amorphous solids, or glasses [1–6]; to provide structural models to investigate phenomena such as electrical conductivity [7–9], fluid flow [10–12], and stress distribution [13]; and to investigate processes such as sintering [9,14,15] or particle-size segregation [16,17]. One of the most fascinating features of random packings is that there is an upper limit of the volume fraction $c_b$ that cannot be surpassed and is significantly smaller than the one $c_m=0.7405$ of the ordered close packings (hexagonal-closed-packed and face-centered-cubic). Since the pioneering work of Bernal [1], who realized a close random packing with real spheres in a laboratory, much effort has been devoted by both experimentalists and theorists to obtain the most compact random packing [18,19]. To our knowledge, the best efficient algorithm to build random close packings on a computer remains the one of Jodrey and Tory [20], which leads to $c_b \simeq 0.649$. We have used it in the present work. It is now believed that the existence of an upper limit for the volume fraction is related to the degeneracy between hexagonal-closed-packed and face-centered-cubic structures leading to the so-called geometrical ''frustration'' associated with the impossibility to tile the space with perfect tetrahedra only [22–24].

One of the most widely used tools to investigate the structure of isotropic random packings is the pair-correlation function $g(r)$, which is the Fourier transform of the experimental wavelength-dependent x-ray- (or neutron-) scattering intensity $I(q)$ [21]. This quantity gives interesting information on the sphere correlations, but is too much average to be able to describe in detail the topology of the local structure. A powerful way to get local information is to determine the Voronoi cells, a method that was initiated by Finney [2] and has a wide range of applications, especially for studying froths and foams [22,25]. A Voronoi cell is the generalization of a Wigner-Seitz cell for disordered structures. It is the polyhedron that contains the ensemble of points closer to a given sphere center than to any other. Apart from a few studies [26–28], the Voronoi tessellation was not performed very often on three-dimensional random packings. A recent work, however, reports on a study of Voronoi cell statistics for a few random packings of limited volume fractions (up to $c=0.58$) [29].

In this paper, we have built random packings containing 8192 spheres, in a wide range of concentrations, up to volume fractions very close to the upper limit $c_b$, using the efficient Jodrey-Tory (JT) algorithm [20]. For each computer generated packing we have calculated the pair-correlation function and performed a complete Voronoi tessellation. An important result of our paper is that all the calculated characteristics vary with $c$ in such a way that when they are extrapolated above $c_b$, they apparently approach their corresponding values for a perfect tetrahedral local order, with dodecahedra as Voronoi cells, for the ideal packing fraction $c_0=0.754$; this value corresponds to the ratio of the volume of a sphere over the volume of its circumscribed perfect dodecahedron. Our results strongly suport the idea that the perfect tetrahedral order (with fivefold symetry) would be an unreachable fixed point as postulated in a recent theory for the glass transition [30]. In Sec. II we provide a short description of the algorithm used to build the packings, in Sec. III we give the results on the pair-correlation function, and in Sec. IV we present the numerical results for the Voronoi cell characteristics.

## II. ALGORITHM USED TO BUILD THE PACKINGS

Since we have reproduced the JT algorithm [20] with only slight differences, we will not provide too many details here. The algorithm proceeds by an iterative sequential resorption of overlaps of imagined spheres, which consists of successive displacements of pairs of nearest-neighboring points (sphere centers) starting at iteration $i=0$ from a set of $N$ points randomly located in a cubic box of edge length $B$. Periodic boundary conditions (PBC's) are used at the box

 

edges. At each iteration $i$ the set of points (the sphere centers) is characterized by the list of coordinates and also by a list of distances (between pairs of points) in increasing order together with some other tables necessary to identify the points in the list. A minimum packing fraction $c_m^i = N(\pi/6)(d_m^i/B)^3$ corresponds to the minimum distance $d_m^i$. Along the iterative procedure, one also carries a maximum distance $d_M^i$ related to a maximum packing fraction $c_M^i = N(\pi/6)(d_M^i/B)^3$, which is set to $d_M^0 = B(6/\pi N)^{1/3}$ (i.e., $c_M^0 = 1$) at $i=0$. After the identification of the pair of points $M_1^i$ and $M_2^i$ realizing the minimum distance $d_m^i = M_1^i M_2^i$ these points are spread apart symmetrically along the $M_1^i M_2^i$ line to new positions $M_1^{i+1}$ and $M_2^{i+1}$ such that $M_1^{i+1} M_2^{i+1} = d_M^i$. Then, before going to the next iteration, the list of distances and the related tables are updated, the new minimum distance $d_m^{i+1}$ is determined, and the maximum distance is set to a lower value given by

$$d_M^{i+1} = d_M^i - \frac{R}{N}(c_M^i - c_m^i)^\alpha, \tag{1}$$

where the ''rate'' $R$ and the exponent $\alpha$ are two input parameters of the algorithm in addition to $B$, $N$, and $d_M^0$. Note that formula (1) is slightly different from (simpler than) the original one used by Jodrey and Tory [20] and consequently our definition of the rate $R$ is different. The process stops at iteration $n$ when one finds $d_M^n < d_m^n$. Then the final minimum distance $d_m^n$ is taken as the particle diameter for the resulting packing.

Note that the value taken for the box edge length $B$ does not play any role as it only fixes the unit of length. In practice, $B$ has been set to an integer value $B=20$ because we have used an underlying cubic lattice of 8000 cells to label the spheres in order to accelerate the search for their neighbors. The number $N$ of spheres has been set to $N=8192$, almost an order of magnitude larger than the JT value $N=1000$ [20]. We have checked on a few other $N$ values that the results are size insensitive. Reaching large $N$ is interesting here in our attempt to get information about correlations on distances larger than previously reported. While the parameter $d_M^0$ is not very important (it should be taken sufficiently large, however) the remaining parameters $R$ and $\alpha$ are essential not only to fix the final packing fraction, but also to determine the overall speed to reach it. Here we have taken $\alpha = 0.33$ and we have varied only $R$. We provide in Table I the values of $R$ considered with the resulting packing fractions. When extrapolating these packing fractions to $R=0$, we obtain $c_b \simeq 0.645$, in good agreement with previous estimates. To build the most dense packing, of volume fraction $c=0.643$, we used a few days of IBM RISC 6000/580 computer time.

As soon as they are built, all the packings are rescaled to get a particle diameter of 1 and, consequently, in the following, the distances will be expressed in diameter units. To give an idea of the linear sizes of our packings we have listed the rescaled box edge lengths $b = B/d_m^n$ in Table I.

TABLE I. Packing fraction $c$ and box edge length $b$ (in diameter units) as a function of the input parameter $R$ defined in the text.

| $R$ | $c$ | $b$ |
|---|---|---|
| 0.1 | 0.4235 | 21.64 |
| 0.05 | 0.4850 | 20.68 |
| 0.033 | 0.5177 | 20.23 |
| 0.020 | 0.5512 | 19.82 |
| 0.010 | 0.5850 | 19.43 |
| 0.004 | 0.6130 | 19.13 |
| 0.002 | 0.6260 | 18.99 |
| 0.001 | 0.6336 | 18.92 |
| 0.0005 | 0.6392 | 18.86 |
| 0.0002 | 0.6430 | 18.82 |

## III. PAIR-CORRELATION FUNCTION

The pair-correlation function $g(\vec{r})$ is defined such that $g(\vec{r})d^3r$ is proportional to the probability of finding a sphere center inside a volume $d^3r$ at a distance $\vec{r}$ from a given sphere center. Consequently, for an isotropic packing, the number of sphere centers $dN$ located between distances $r$ and $r+dr$ from a given sphere center is proportional to $g(r)4\pi r^2 dr$. Knowing that, on average, the number of sphere centers per unit volume is $6c/\pi$, one can normalize $g(r)$ to unity when $r$ goes to infinity by writing

$$dN = \frac{6c}{\pi} g(r) 4\pi r^2 dr = 24 c g(r) r^2 dr. \tag{2}$$

We have used this formula to compute $g(r)$ in all the packings listed in Table I. For each sphere in the box, we counted the number of sphere centers located between distances $r$ and $r+dr$ from its center, taking care of PBC's when investigating regions outside of the box. Then we averaged the results over the $N$ particles in the box and divided it by $24cr^2dr$. In all the calculations, we have taken $dr=0.05$. We have checked that our $g(r)$ curves exhibit all the well-known short-range features [21]. In particular, a double peak, characteristic of close packings, appears in the range 1.7–2 for $c$ larger than about 0.6. But here we would like to focus on the behavior for larger distances. Such behavior is well reflected in Fig. 1, where we have plotted $\log_{10}|g(r)-1|$ versus $r$ for two packing fractions $c=0.551$ and $c=0.643$. The curves have been limited to the maximum value $r_M = b/2$ since correlations due to artifacts from PBC's appear for $r > r_M$. The dashed curves correspond to the pair-correlation function calculated in the same manner, but for the same number of points distributed randomly (without taking care of hard core conditions) in the same box. When the solid curve reaches the dashed curve, this means that the interparticle distance is so large that the two particles of the pair become uncorrelated. It clearly can be seen on these figures that the range of $r$ values where the spheres are significantly correlated increases with concentration. More quantitatively, for $c > 0.4$, our data can be well fitted by

$$g(r) = 1 + A \exp\left(-\frac{r}{\xi}\right)\sin\left(2\pi\frac{r}{a}\right), \tag{3}$$

where $A$ is an amplitude parameter, $a$ is twice the period of the oscillations seen in Fig. 1, and $\xi$ is a typical correlation length inversely proportional to the slope of the straight-line fit of the maxima. From a standard nonlinear fitting proce-

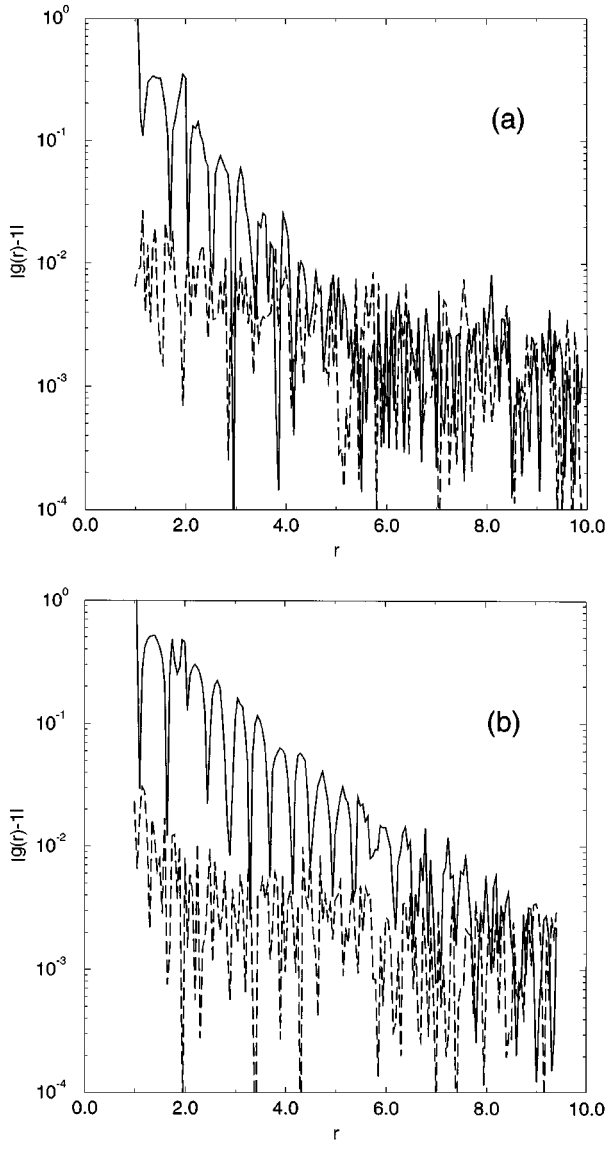


FIG. 1. Pair-correlation function presented as a linear log plot of $|g(r)-1|$ versus $r$ for (a) $c=0.551$ and (b) $c=0.643$. The dashed curve corresponds to the same number of points randomly disposed in the box.

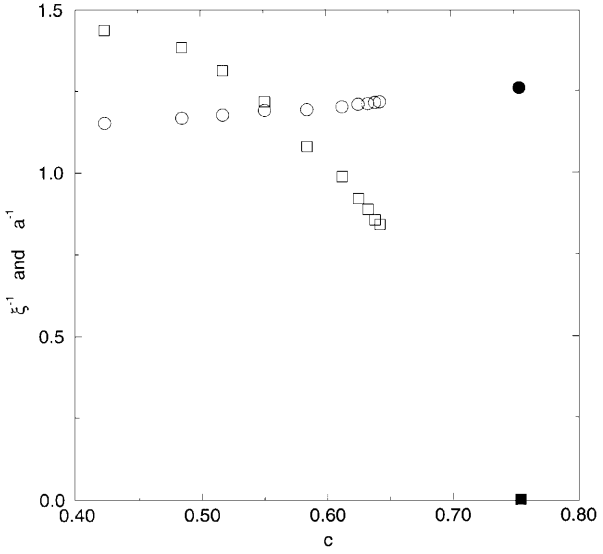


FIG. 2. Parameters of the fit of $g(r)$ to Eq. (3), $\xi^{-1}$ (squares) and $a^{-1}$ (circles), as a function of $c$. The filled symbols indicate their expected extrapolations for $c=c_0$ (see the text).

dure performed in the range $2.5<r<r_M$, we have found that $A$ ($\simeq 1.7$) is roughly independent on $c$, while $a$ decreases and $\xi$ increases when $c$ increases. The numerical results for $a^{-1}$ and $\xi^{-1}$ are reported as a function of $c$ in Fig. 2. These results do not depend much on the fitting procedure. We have checked that the same results can be obtained from estimations of the position and the width of the Fourier transform $S(q)$ of $g(r)$, but we prefer to perform a direct fit of $g(r)$ to get better control on the error bars on both $a^{-1}$ and $\xi^{-1}$, which turn out to be smaller than the thickness of the symbols used in the figure.

The data of Fig. 2 can be analyzed as the functional dependence that $\xi$ would diverge at some $c$ value $c_0$ larger than $c_b$. One can determine this particular value of $c$ in coherence with the analysis of the Voronoi cells, which will be done in Sec. IV. We suppose that in such an ideal packing of volume fraction $c_0$, a given sphere would be surrounded isotropically by 12 spheres all at a diameter distance, forming a perfect icosahedral arrangement. The Voronoi cell, which is the polyhedron whose faces are the bisector planes of the segments joining the central sphere to all its neighbors, would then be a perfect dodecahedron with faces tangent to the sphere. The corresponding value of the volume fraction is obtained by dividing the volume of the sphere of diameter 1, which is $\pi/6$, by the volume of a dodecahedron such that the center-to-face distance is equal to a half unit. Knowing, from a geometry textbook [32], the expressions for the volume $V_d$ and the center-to-face distance $H_d$ as a function of the edge length $l$ for a perfect tetrahedron

$$V_d=\frac{15+7\sqrt{5}}{4}\,l^3, \tag{4a}$$

$$H_d=\frac{1}{5^{1/4}}\left(\frac{1+\sqrt{5}}{2}\right)^{5/2}\frac{l}{2}, \tag{4b}$$

one finds $l=0.449$ and $c_0=0.754$, a value slightly larger than the one of the most compact packing of equal spheres $c_m=\pi/3\sqrt{2}=0.7405$.

The large-$r$ oscillations of $g(r)$ are due to a pseudoperiodic arrangement of the spheres around a given one. Since the correlation is more effective the denser the packing is, one expects that the period of the oscillations should be associated with the smallest distance. Aside from the hard-sphere radius (1 in our units) one can look for what we call the most compact parallel planes locally. The most compact plane closest to a given sphere is defined by the centers of its three nearest neighbors. Therefore, in the ideal arrangement described above, this period $a$ should be identified with the distance between the center of the icosahedron and the center of one of its faces, knowing that the distance from the center to a vertex is here equal to unity. Getting, again from a geometry textbook [32], the expressions of the center-to-face

and center-to-vertex distances $H_i$ and $L_i$ as a function of the edge length $l$ for a perfect icosahedron,

$$H_i=\frac{1}{\sqrt{3}}\left(\frac{1+\sqrt{5}}{2}\right)^2\frac{l}{2}, \tag{5a}$$

$$L_i=5^{1/4}\left(\frac{1+\sqrt{5}}{2}\right)^{1/2}\frac{l}{2}, \tag{5b}$$

one gets $a_0=H_i/L_i=0.7946$. The corresponding point ($c_0$, $a_0^{-1}$) is shown by a filled circle in Fig. 2. It is remarkably located on the straight-line fit of the data for $a^{-1}$.

## IV. VORONOI CELLS

We have recently written a code able to perform a Voronoi tessellation in three dimensions given a set of points in a cubic box with periodic boundary conditions at the box edges [31]. The algorithm first determines the Delaunay tetrahedral simplicial cells, which are, among all the tetrahedra involving four arbitrary sphere centers, the ones where the four centers are chosen such that no other sphere center lies inside their circumscribed sphere (i.e., the sphere whose surface is defined by the four centers). Then all the elements, vertices, edges, and faces of a Voronoi cell of a given sphere are determined, knowing that the vertices are the centers of the circumscribed spheres of all the tetrahedra sharing the given sphere. For each cell, not only have the number of vertices $V$, the number of faces $F$, and the number of edges $E$ been determined, but also the total volume of the cell $v$, the area $s$ for each face, the length $l$ for each edge, as well as the total surface area $S$ for the cell (sum of the face areas).

In Fig. 3(a) we report the distribution function $h_v$ for the cell volumes defined such that $h_v(v)dv$ is the fraction of cells with a volume lying between $v$ and $v+dv$. In Fig. 3(b) we report the distribution function $h_S$ for the cell surfaces defined similarly. Both histograms become more and more peaked as the concentration increases. As a measure of their width we have calculated the standard deviations $\sigma_v$ and $\sigma_S$ of $h_v(v)$ and $h_S(S)$, which have been reported as a function of $c$ in Fig. 4(a). The filled circle on the horizontal axis represents the concentration $c_0$ defined in Sec. III. This point lies nicely on the extrapolations of both curves as if both the surface and the volume of the cell would be well defined at $c_0$. As a measure of the positions of the peaks we have calculated the averages $v_m$ and $S_m$. We have verified that $v_m=b^3/N=\pi/6c$. In Fig. 4(b) we have reported the dimensionless quantity $v_m^{1/3}/S_m^{1/2}$ as a function of $c$. The filled circle corresponds to the value of that ratio for a perfect dodecahedron at $c_0$. Here again the point is well located on the extrapolated curve suggesting that the packing develops towards a pure icosahedral order, with dodecahedra as Voronoi cells, as the concentration increases. The same analysis (not reported here) has been done for the distribution functions $h_s$ and $h_l$ for the face areas and the edge lengths and leads to the same conclusion.

Concerning the quantities $F$, $V$, and $E$, it is not necessary to study all of them since they are related. They not only verify the well-known Euler relationship [22,25]

$$V-E+F=2, \tag{6}$$

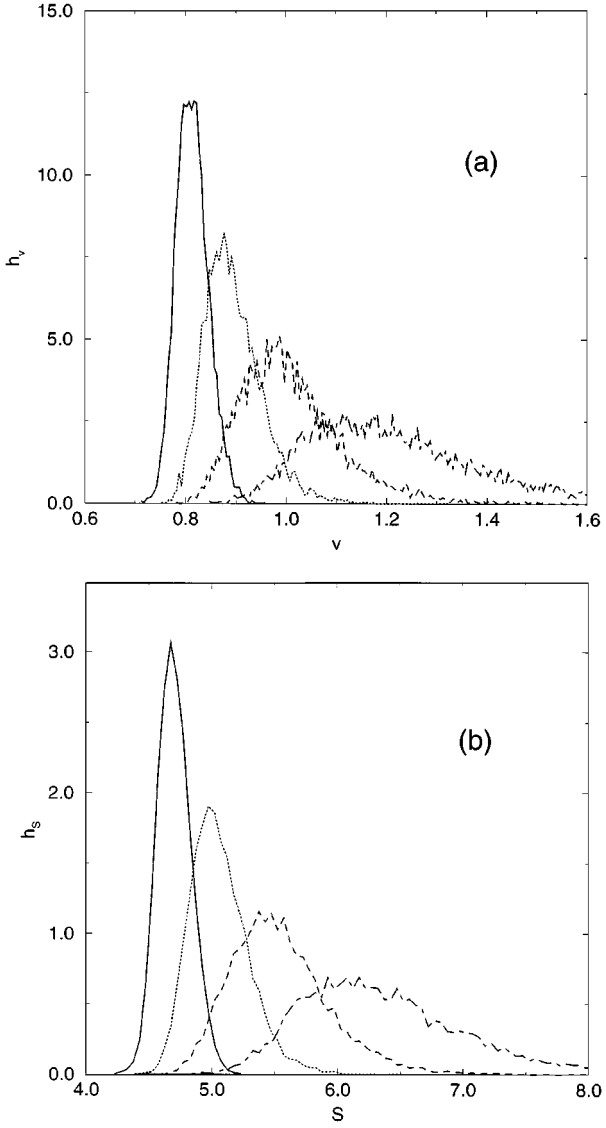


FIG. 3. (a) Cell volume $h_v(v)$ and (b) cell surface $h_S(S)$ distribution functions. Dot-dashed, dashed, dotted, and solid lines correspond to $c=0.424, 0.518, 0.585$, and 0.634, respectively.

but also, since the system is disordered, there are always three edges meeting at a given cell vertex and therefore $2E=3V$. Other situations, which occur in some regular packings, correspond to the existence of degenerated zero-area faces and have no chance to occur in the presence of randomness. Hence both $E$ and $V$ are related to the number of faces, or coordination number (number of nearest neighbors), $F$,

$$E=3(F-2), \tag{7a}$$

$$V=2(F-2). \tag{7b}$$

These relations have been verified on our numerical results. Another interesting quantity characterizing a given face is its number of edges $e$. A relationship exists between its mean value $\langle e\rangle$ and the mean coordinance $z=\langle F\rangle$ [22]:

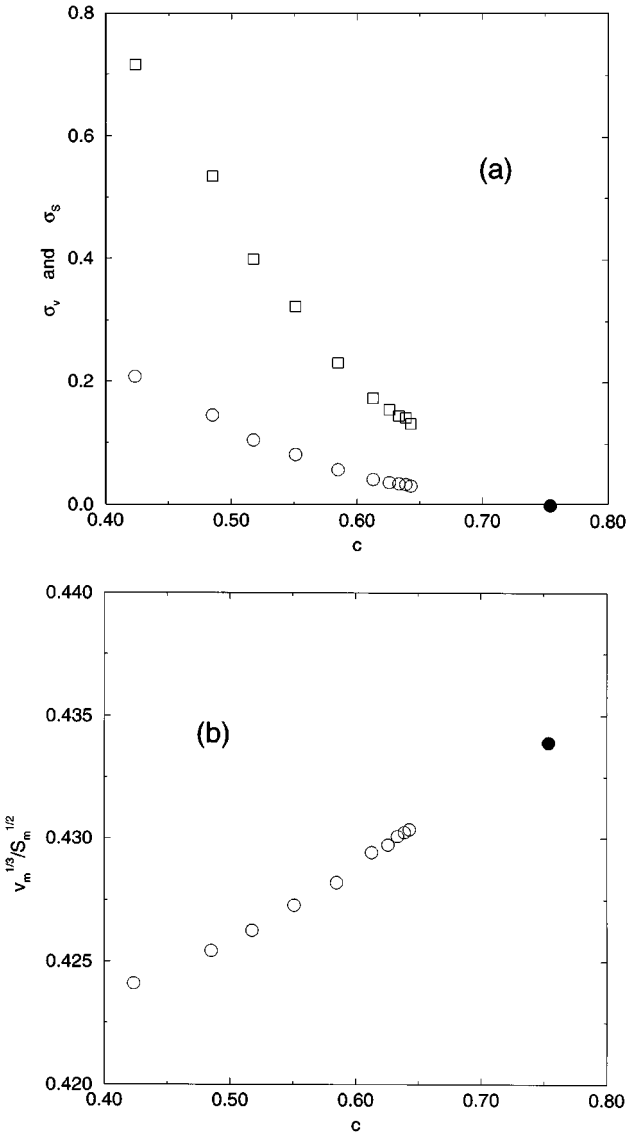


FIG. 4. (a) Standard deviations $\sigma_v$ and $\sigma_S$ and (b) dimensionless ratio $v_m^{1/3}/S_m^{1/2}$ (b) as a function of $c$. The filled circle indicates their expected extrapolations for $c=c_0$.

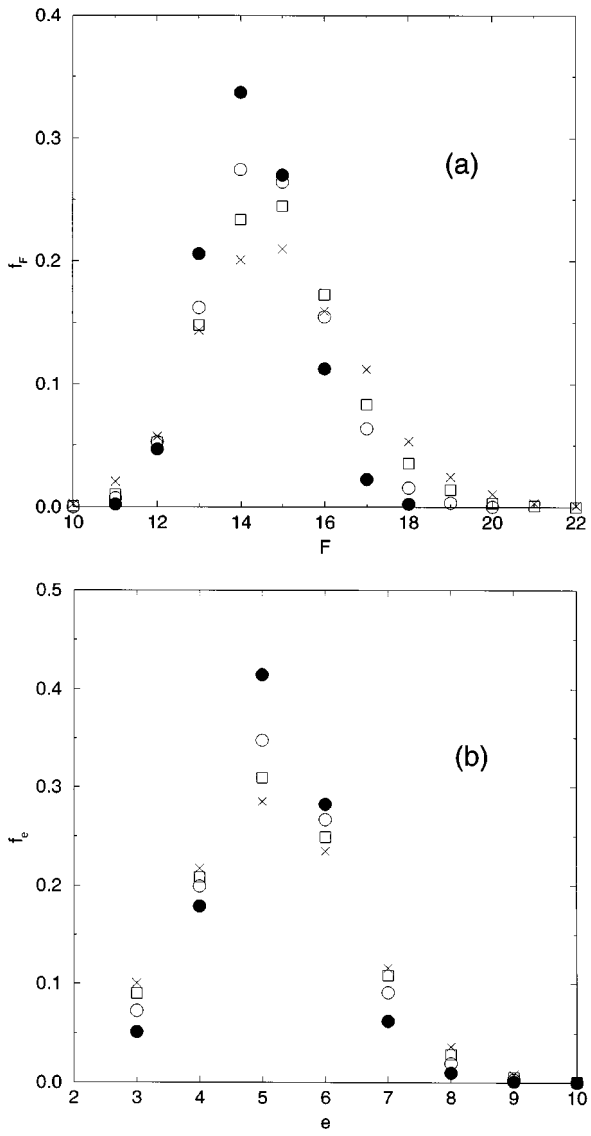


FIG. 5. (a) Fraction $f_F$ of cells with $F$ faces and (b) fraction $f_e$ of faces with $e$ edges. Crosses, open squares, open circles, and filled circles correspond to $c=0.424$, 0.518, 0.585, and 0.634, respectively.

$$z=\frac{12}{6-\langle e\rangle}\,. \tag{8}$$

In Figs. 5(a) and 5(b) we report the quantities $f_F$ and $f_e$, which are the fraction of cells with $F$ faces and the fraction of faces with $e$ edges, respectively. On these figures the tendency to build mostly dodecahedra as $c$ increases is less clear than with the other quantities previously reported. The fraction of cells with 14 faces increases with $c$; also, but less quickly, the fraction of cells with 13 and 15 faces increases, while the fraction of cells with $F>15$ decreases. Surprisingly, the fraction of cells with 12 faces stays almost constant. Note that in the present work the faces are not weighted by their areas, which is probably the main reason for the misleading occurrence of large values of $F$ or $e$. In Fig. 5(b) the situation is slightly better since the fraction of pentagons is always in the majority and increases with $c$. But also the fraction of hexagons increases. In Fig. 6 we report the mean coordination number $z=\langle F\rangle$ as a function of $c$. Unless one accepts a very slow convergence, there is no clear evidence for an extrapolation to $z=12$ (dodecahedron) for $c\rightarrow c_0$. In fact, all the results reported in Figs. 5 and 6 are not incompatible with recovering a dodecahedron for $c=c_0$ because extra faces might persist whose areas vanish only at $c=c_0$. In that case $z$ might be discontinuous at $c=c_0$. The same kind of behavior occurs when disturbing any degenerate regular structure, for example, when including an infinitesimal temperature in a fcc crystal.

## V. DISCUSSION AND CONCLUSION

We have built large random packings of hard spheres with volume fractions very close to the upper limit and we have systematically studied some of their structural characteristics as a function of the volume fraction. From a quantitative analysis of both the pair-correlation function and some Voronoi cell characteristics, we conclude that the system ap-

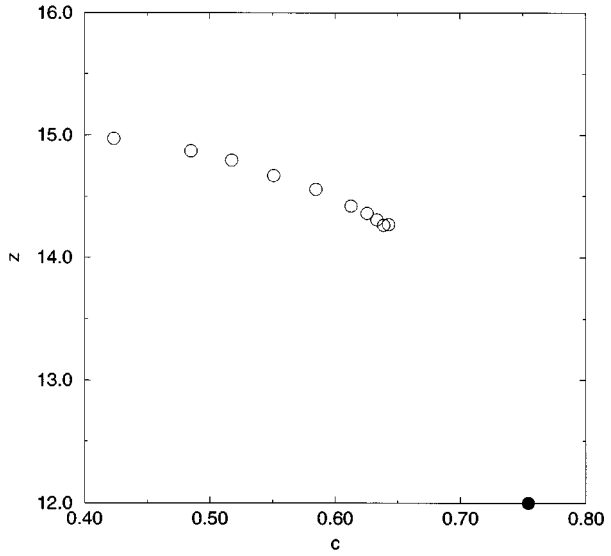


FIG. 6. Coordination number $z$ as a function of $c$. The filled circle indicates $z=12$ for $c=c_0$.

proaches icosahedral order when increasing the density towards $c_0$, but it stops at the upper limit $c=c_b$ due to geometrical frustration. We plan to fill the gap by building sphere packings in curved space, but the extension of the JT algorithm to curved space is a difficult task.

Although the motivation is different, our work on the Voronoi cells is complementary to the recent one by Oger *et al.* [29]. These authors have considered packings built by random sequential addition and therefore are bounded to the so-called jamming limit $c_j=0.385$ [33], to which they have added a Bennet-like packing of volume fraction $c=0.58$ [34]. Their aim was to contrast their numerical results with some analytical conjectures on the statistics of the Voronoi cells. When comparing both studies, it appears that the numerical results are remarkably close. For example, we get $z=14.56$ for $c=0.585$, while they get $z\simeq 14.53$ for $c=0.58$. Such agreement might be surprising if one knows that any curve giving a structural characteristic as a function of the volume fraction is not universal. The detailed structure of a random packing depends not only on its volume fraction, but also on the procedure used to build it. In particular, the Bennet packing is anisotropic [34] while the JT packings are isotropic. Hence the nice agreement between both studies suggests that a quasiuniversality exists for random packings, at least for very large packing fractions (larger than about 0.55).

Extensions of this study to the thermodynamics of soft spheres are in progress and some preliminary results have been obtained in connection with glass transitions [31]. When the temperature decreases from the liquid phase, the system behaves the same way as it does here when increasing its density, i.e., it tries to reach a local icosahedral order. All these results appear to be in agreement with a recent theory of glass transitions that also invokes an unreachable critical point [30].

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