Model of random packings of different size balls

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We develop a model to describe the properties of random assemblies of polydisperse hard spheres. We show that the key features to describe the system are (i) the dependence between the free volume of a sphere and the various coordination numbers between the species and (ii) the dependence of the coordination numbers with the concentration of species; quantities that are calculated analytically. The model predicts the density of random close packing and random loose packing of polydisperse systems for a given distribution of ball size and describes packings for any interparticle friction coefficient. The formalism allows to determine the optimal packing over different distributions and may help to treat packing problems of nonspherical particles which are notoriously difficult to solve.

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Understanding the basic properties of sphere packings is a major challenge since this problem may provide valuable knowledge regarding low-temperature phases in condensed-matter physics [1]. The canonical example is perhaps the monodisperse sphere packing problem. It has been mathematically proven that the optimum way to arrange monodisperse spheres is the face-centered cubic lattice; a problem that has been solved recently by Hales, ~400 years after the famous Kepler conjecture on the issue. On the other hand, it is commonly observed that packings arrange in a random fashion at a lower density state called random close packing (RCP) [2]. Furthermore, packings are mechanically stable up to an even lower limit called random loose packing (RLP).

In parallel with the large literature dealing with monodisperse sphere packings, a large body of experimental, theoretical, and numerical works has been devoted to the analysis of polydisperse systems; the interest arising due to the simple fact that polydispersivity is omnipresent in most realistic systems and industrial applications [3,4]. While previous approaches have focused on frictionless packings, an integrated analytical approach that brings together different observations for all packings from RLP to RCP and for any friction or coordination number is still lacking. Based on our previous statistical-mechanics approach [5], here we build such a framework.

We show that the key aspect to solve this problem is the dependence of the various coordination numbers between the different species and the concentration of the species. This is calculated here and shown to agree well with computer simulations. This result is then incorporated into a statistical theory of volume fluctuations as in [5] which calculates the free volume of a particle in terms of the coordination number. The main result is the prediction of the RLP and RCP limiting densities for a given distribution of ball sizes as well as the prediction of densities for any packing in between those limits. The formalism allows for a determination of the best packing fraction in terms of different distribution of ball sizes with specified constraints, as we show with a simple example. We discuss possible generalization of the method to solve more difficult problems such as the phase behavior of systems of nonspherical particles such as rods or spherocylinders in any dimensions; problems of long-standing history in condensed matter [6].

Recent theoretical advances [5] allow for the prediction of the density of RCP and RLP for equal-size ball packings using a relation between the average volume and the geometrical coordination number. Following this approach, we here describe long-range spatial correlations through a mean-field background term. This approximation makes the problem amenable to analytic calculations and is shown to describe well our simulation results. An explicit inclusion of such correlations is possible in our framework, but severely complicates any solution attempts. Thus, we believe that the present approach is accurate enough for many important properties, such as the volume fraction calculation.

The above theoretical framework will guide the present formalism for polydisperse systems. We first treat the case of binary mixtures of hard spheres of radius R_1 and $R_2 > R_1$ in three dimensions (3D) and then generalize the problem to any distribution.

I. CALCULATION OF z_{ij}

The key quantity to calculate is z_{ij}, denoting the mean number of contacts of a ball of radius R_i with a ball of radius R_j, versus the concentration of one of the species. We need a formula for z_{ij} as a function of $(z, x, \frac{R_2}{R_1})$, the latter being the size ratio, x is the fraction of small balls in the packing $x \equiv N_1/(N_1+N_2)$, with N_i the number of i balls, and z is the global geometrical coordination number averaged over all the particles, $z \equiv xz_1 + (1-x)z_2$, where z_i is the average coordination of each species.

The coordinations are determined by three equations

$$z_i = z_{i1} + z_{i2}, \quad (i = 1,2), \quad xz_{12} = (1-x)z_{21}. \tag{1}$$

We assume that these coordinations are inversely proportional to the average solid angle extended by contacting balls R_1 and R_2. The average solid angles are denoted $\langle S_i^{occ} \rangle$ and are calculated in terms of the solid angle that a ball R_j occupies on a ball R_i according to $\langle S_i^{occ} \rangle = xS_{i1}^{occ} + (1-x)S_{i2}^{occ}$ with [see Fig. 1(a)]

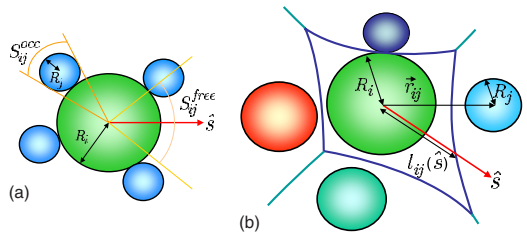

FIG. 1. (Color online) (a) Occupied surface and free surface. (b) Voronoi cell for polydisperse balls. Plots are in two-dimensions (2D) for easier visualization.

$$S_{ij}^{occ}/2\pi \equiv \int_0^{\arcsin(R_j/R_i+R_j)} \sin\,\theta d\theta = 1-\sqrt{1-\left(\frac{R_j}{R_i+R_j}\right)^2}.$$

Thus, $\langle S_i^{occ}\rangle$ represents the mean occupied surface on a i ball weighted by the concentrations x and $(1-x)$. This represents an approximation since the real weights are z_{i1}/z_i and z_{i2}/z_i, respectively. Then, $z_i \propto 1/\langle S_i^{occ}\rangle$ leading to the following normalizations:

$$z_1 = \frac{z}{x+(1-x)\dfrac{\langle S_1^{occ}\rangle}{\langle S_2^{occ}\rangle}}, \quad z_2 = \frac{z}{x\dfrac{\langle S_2^{occ}\rangle}{\langle S_1^{occ}\rangle}+(1-x)}. \tag{2}$$

Thus, the system of Eq. (1) is reduced to a system of three equations for four unknowns z_{ij}. To close the system, we assume proportional laws and deduce z_{ij} from z_i by considering that z_{ij} is proportional to the number of contacts of the i balls times the number of contacts of the j balls

$$z_{11} = A(z_1x)(z_1x), \quad z_{12} = A(z_1x)z_2(1-x),$$

$$z_{21} = Bz_2(1-x)(z_1x), \quad z_{22} = Bz_2(1-x)z_2(1-x). \tag{3}$$

Using the first equation in Eq. (1), we find the constants A and B, leading to the solution

$$z_{11} = \frac{z_1^2x}{z}, \quad z_{12} = \frac{z_1z_2(1-x)}{z},$$

$$z_{21} = \frac{z_1z_2x}{z}, \quad z_{22} = \frac{z_2^2(1-x)}{z}. \tag{4}$$

Figure 2(a) compares this solution to numerical simulations of Hertz packings jammed at RCP [5] for $R_1/R_2=1.4$ and $z=6$. We find that the formulas are very accurate for size ratios below 1.5 and present small deviations up to size ratio 2. The results are also in agreement with [3,4].

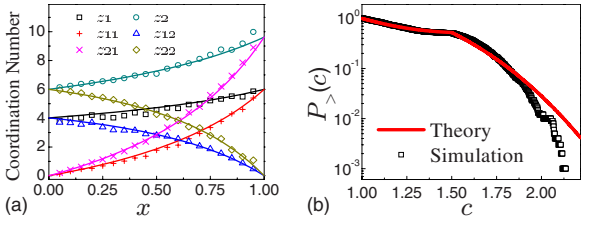

FIG. 2. (Color online) Comparison between theory and numerical simulations for (a) z_{ij}, Eq. (4), and (b) $P_>(c)$, Eq. (11).

II. VORONOI CELL

The common way to divide a system into volumes associated with each particle is the Voronoi tessellation. The Voronoi cell for monodisperse particles [5] is composed by all the points nearest to the center of the ball than to any other ball. This definition has been extended in [7] to the case of polydisperse systems and nonspherical particles: instead of considering the classical Voronoi polyhedron defined by the center of the particle, one should consider all the points which are closer to the surface of a given particle. Such a construction is called the *Voronoi S region* and tiles a system of nonspherical convex particles and polydisperse systems as can be seen in Fig. 1(b). Following this approach, we calculate the average volume of a polydisperse Voronoi cell, denoted W. The volume fraction is given by $\phi=\frac{V_g}{W}$, where $V_g=\frac{4\pi}{3}[xR_1^3+(1-x)R_2^3]$ is the mean volume of a ball. We first find the analytical formula of the Voronoi S region. The boundary of the Voronoi cell in the direction $\hat{s}$ of a i ball next to a j ball at position r_{ij} is [Fig. 1(b)]

$$l_{ij}(\hat{s}) = \frac{1}{2}\frac{r_{ij}^2-(R_i-R_j)^2}{r_{ij}(\hat{r}_{ij}\hat{s})-(R_i-R_j)}, \tag{5}$$

where $\hat{s}$ and $\hat{r}_{ij}$ are unitary. Thus, the boundary of a Voronoi cell of a ball i in the direction $\hat{s}$ is the minimum of $l_{ij}(\hat{s})$ over all the particles j for any $l_{ij}(\hat{s})>0$. This leads to

$$l_i(\hat{s}) = \frac{1}{2}\min_{\hat{s}\hat{r}_{ij}>\frac{R_i-R_j}{r_{ij}}}\frac{r_{ij}^2-(R_i-R_j)^2}{r_{ij}(\hat{r}_{ij}\hat{s})-(R_i-R_j)}. \tag{6}$$

The volume of the cell of the ball i is then given by $W_i=\frac{1}{3}\oint l_i(\hat{s})^3 ds$. We define the orientational Voronoi volume, W_i^s, along the direction $\hat{s}$ by $W_i \equiv \frac{1}{4\pi}\oint W_i^s ds = \langle W_i^s\rangle_s$. This leads to

$$W_i^s = \frac{\pi}{6}\min_{r_{ij}\hat{s}>\frac{R_i-R_j}{r_{ij}}}\left(\frac{r_{ij}^2-(R_i-R_j)^2}{r_{ij}(r_{ij}\hat{s})-(R_i-R_j)}\right)^3. \tag{7}$$

This definition leads to the results of [5] when $R_1=R_2$. Since the system is isotropic, the mean Voronoi volume can be calculated as

$$W \equiv \langle\langle W_i^s\rangle_s\rangle_i = \langle\langle W_i^s\rangle_i\rangle_s = \langle W_i^s\rangle_i. \tag{8}$$

III. CALCULATION OF THE MEAN VORONOI VOLUME

Having calculated the Voronoi cell exactly in Eq. (7), we now proceed to develop a probability theory of volume fluctuations in the spirit of the quasiparticle approximation used in [5] to obtain the mean Voronoi volume. For a given ball i, the calculation of W_i^s reduces to finding the ball j^* that minimizes $l_{ij}(\hat{s})$. We call j^* the Voronoi ball for the ball i. We consider $r\equiv r_{ij^*}$, $\cos\theta\equiv\hat{s}\hat{r}_{ij^*}$, and $c\equiv 2l_{ij^*}$. We have $c=\frac{r^2-(R_i-R_j)^2}{r\cos\theta-(R_i-R_j)}$. Therefore, we just need to compute the inverse cumulative distribution function, denoted $P_>(c)$, to find all the balls j with $l_{ij}>\frac{c}{2}$ and thus not contributing to the Voronoi volume of the ball i. The average Voronoi volume is then given by the expression

$$W = \frac{\pi}{6}\langle c^3\rangle = -\frac{\pi}{6}\int_0^\infty c^3 \frac{dP_>(c)}{dc} dc = \frac{\pi}{2}\int_0^\infty c^2 P_>(c) dc. \tag{9}$$

We calculate the mean Voronoi volume for the balls of radii R_1 and R_2 separately and then average them. We denote $P_>^1(c)$ and $P_>^2(c)$ the inverse cumulative distributions, respectively, and $W = x\frac{\pi}{2}\int_0^\infty c^2 P_>^1(c)dc + (1-x)\frac{\pi}{2}\int_0^\infty c^2 P_>^2(c)dc$ and therefore

$$P_>(c) = xP_>^1(c) + (1-x)P_>^2(c). \tag{10}$$

IV. CALCULATION OF $P_>(c)$

There are three salient steps in the calculation of $P_>(c)$. (i) The separation of $P_>(c)$ following Eq. (10). (ii) The separation of each term $P_>^i(c)$, $i=1,2$, into two contributions: a term taking into account the contact spheres, $P_>^{iC}(c)$, and a bulk term, $P_>^{iB}(c)$. The contact term clearly depends on z_{ij}. The bulk term averages over all spatial correlations of noncontact particles and, without significant loss of accuracy as shown below, we assume that it only depends on the average value of W. In principle, it is possible to use a more realistic form for this term, but this would render the problem practically unsolvable. (iii) The separation of $P_>^{iC}(c)$ and $P_>^{iB}(c)$ into two terms $P_>^{ijC}(c)$ and $P_>^{ijB}(c)$, $j=1,2$, for each species.

An assumption of the theory (to be tested *a posteriori* with computer simulations) is that all of these terms are independent. Thus,

$$P_>(c) = xP_>^{11C}(c)P_>^{12C}(c)P_>^{11B}(c)P_>^{12B}(c) + (1-x)P_>^{21C}(c)P_>^{22C}(c)P_>^{21B}(c)P_>^{22B}(c). \tag{11}$$

Also, we work in the limit of large number of particles leading to Boltzmann-like exponential distributions for each $P_>^{ijC}$ and $P_>^{ijB}$ [5]. Four important quantities are then defined. (i) $V_{ij}^*(c)$ and (ii) $S_{ij}^*(c)$: the excluded volume and surface on the ball, respectively, where no center of a ball j can be located for a given ball i and for a given c. (iii) ρ_j: the mean number of balls j by the left free volume. (iv) ρ_{ij}^s: the mean number of balls j by the left free surface on a ball i. These considerations lead to

$$P_>^{ijB}(c) = \exp[-\rho_j V_{ij}^*(c)],$$

$$P_>^{ijC}(c) = \exp[-\rho_{ij}^s S_{ij}^*(c)]. \tag{12}$$

Next, we calculate these four quantities. To simplify, we denote $l \equiv R_i + R_j$, $k \equiv R_i - R_j$, and Θ the step function. We obtain

$$S_{ij}^*(c) = \int \Theta\left(c - \frac{l^2-k^2}{l\cos\theta - k}\right) ds = 2\pi\left(1 - \frac{l^2-k^2-kc}{lc}\right),$$

$$V_{ij}^*(c) = \int \Theta\left(c - \frac{r^2-k^2}{r\cos\theta - k}\right) dr^3 = 2\pi\left\{-\frac{1}{4c}[(c+k)^4 - l^4] + \frac{1}{3}[(c+k)^3 - l^3] + \left(\frac{k^2}{2c} + \frac{k}{2}\right)[(c+k)^2 - l^2]\right\},$$

$$\rho_j(W) = \frac{x_j}{W - V_g}, \tag{13}$$

where $x_1 = x$ and $x_2 = (1-x)$. The fourth quantity, ρ_{ij}^s, is the most difficult to calculate. In terms of the occupied areas Eq. (1), we have $\rho_{ij}^s = \frac{z_{ij}}{4\pi - z_{i1}S_{i1}^{occ} - z_{i2}S_{i2}^{occ}}$. However, for the contact terms, the analogy with the Boltzmann-like exponential distribution of volumes is far from being exact. This is because this form is based on the large number limit which in the case of contacting balls is no more than around 6. Therefore, the exponential distribution is used as an ansatz with ρ_{ij}^s a variational parameter as in [5]. We denote $\langle S_{ij}^{\text{free}}\rangle$ the mean solid angle of the gaps left between the j contacting neighbors of a i ball [Fig. 1(a)]. We obtain

$$\langle S_{ij}^{\text{free}}\rangle = \int_0^\infty S_{ij}^*(c)\frac{d[1-P_>(c)]}{dc} dc \approx \rho_{ij}^s \int_0^{2\pi} S_{ij}^* \exp(-\rho_{ij}^s S_{ij}^*) dS_{ij}^* = \frac{1}{\rho_{ij}^s}. \tag{14}$$

Then, we perform numerical simulations to find $\langle S_{ij}^{\text{free}}\rangle$. We randomly generate balls of radius R_i and R_j with the proportion z_{i1}/z_i and z_{i2}/z_i, respectively, around a ball of radius R_i and then evaluate the mean-free surface $\langle S_{ij}^{free}\rangle$ and its inverse to obtain ρ_{ij}^s. We find

$$\rho_{ij}^s(z_{ij}) = \frac{z_{ij}}{2\pi}\left(1 - \frac{z_{i1}}{z_i}\frac{S_{i1}^{occ}}{2\pi} - \frac{z_{i2}}{z_i}\frac{S_{i2}^{occ}}{2\pi}\right) = \frac{z_{ij}}{2\pi}\left(\frac{z_{i1}}{z_i}\sqrt{1 - \left(\frac{R_1}{R_i+R_1}\right)^2} + \frac{z_{i2}}{z_i}\sqrt{1 - \left(\frac{R_2}{R_i+R_2}\right)^2}\right), \tag{15}$$

which is a generalization of the results of [5] to polydisperse systems.

Using Eqs. (12), (13), and (15) into Eq. (11), $P_>(c)$ can be calculated by solving the equations numerically. Figure 2(b) depicts the comparison of the theoretical results of the probability of Voronoi volumes $P_>(c)$ to computer generated Hertzian packings with $z=6$ for $x=0.5$ at RCP. The calculated distribution is quite accurate for most of the range except for small deviations at high values of c which, however, do not affect much the value of the average $\langle c^3\rangle$ in Eq. (9). This shows that our mean-field approximation already captures the main contribution of the probability distribution function $P_>(c)$.

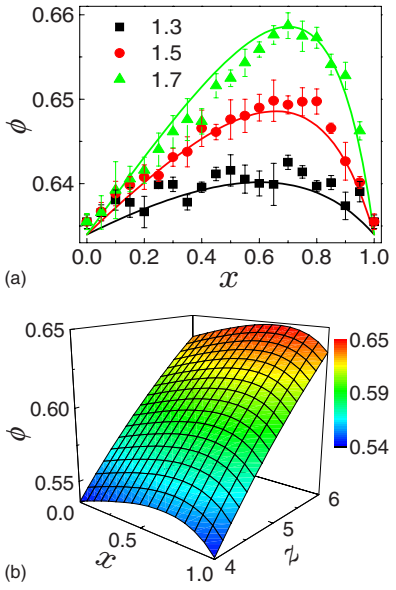

FIG. 3. (Color online) (a) Comparison between theory and numerical simulations of Hertzian packings at RCP, i.e., $z=6$ vs x for different values of R_1/R_2 as indicated. Error bars are std over ten realizations of the packings with 10 000 balls. (b) Three dimensional surface plot of ϕ as a function of z and x for $R_1/R_2=1.5$. The numerical results at RCP and RLP are provided in supplementary information A.

V. CALCULATION OF W

The above considerations lead to a final form to calculate W using Eq. (11) into Eq. (9)

$$W = \frac{\pi}{2}\sum_i x_i \int_0^\infty c^2 \exp\left\{\sum_j \left[-\rho_{ij}^s S_{ij}^*(c) - \rho_j(W) V_{ij}^*(c)\right]\right\} dc. \tag{16}$$

Notice that $\rho_j(W)$ depends on W, Eq. (13), and $\rho_{ij}^s(z_{ij})$ depends on the z_{ij}, Eq. (15), which in turn depends on the concentration x and z through Eq. (4). Therefore, Eq. (16) is a self-consistent equation to obtain $W(z,x)$ for a given R_1/R_2. A numerical integration of Eq. (16) is performed versus x for a given value of z. By considering the isostatic limits of $z=6$ and $z=4$, we predict the limits of RCP and RLP at zero friction and infinite friction between the spheres, respectively [5]. The results for the volume fraction at RCP versus x are depicted in Fig. 3(a) which also compares the results to numerically generated packings of Hertz spheres [5]. We see a very good agreement indicating that the theory captures well the behavior of polydisperse packings and that the approximations used are reasonable. For size ratios larger than 2, deviations are found indicating the limit of validity of the approach. For any other value of interparticle friction between 0 and ∞, the density is obtained by setting z between the limiting isostatic values of 6 and 4, respectively. The resulting volume fraction is shown in Fig. 3(b). Our results promote new experiments to test the RLP limit of polydisperse systems shown in Fig. 3(b).

The formalism can be extended to consider any distribution of sphere radius. The main modification is that all the sums over the radius are replaced by integrations over the desired distribution of radius $P(R)$ (the binary case corresponds to two delta functions at R_1 and R_2).

And all the quantities are calculated for balls of internal radius r and external R and x and $(1-x)$ are replaced by $P(r)dr$ and $P(R)dR$, respectively, including the coordinations (see supplementary information B). This result allows to explore the space of radius distributions in search of the optimal packings for a given polydispersity. This analysis could be of industrial interest if one wishes to optimize the packing fraction by introducing different species in the mixture.

We calculate the volume fraction for several distributions $P(R)$ constraint by ball radius in the range [1,2] in search of the optimal packing. We calculate the volume fraction for various $P(R)$ ranging from uniform to two-peaked Gaussian distributions of varied widths. We find that the more small balls we add, the better the packing until a certain point where the volume fraction starts to decrease. This maximum can be rationalized assuming that the small balls always fill the gaps between the large ones as long as there are enough large balls. Further extensions of the theory to any dimension can be performed by replacing 3 by d in Eq. (7) and developing a theory of volume fluctuation in d dimensions. We notice that many of the approximations employed in 3D may become exact for large d, thus we expect improved results in the mean-field limit of infinite dimensions.

The method allows to treat more difficult problems. For instance, the prediction of the volume fraction of a jammed system of nonspherical particles is a long-standing problem. Theoretical predictions of Onsager [6] are valid for large aspect ratios, such as elongated rods. Experiments, however, find interesting new physics for small aspect ratios. In this respect, the present polydisperse theory could be mapped to the problem of ellipsoids, spherocylinders, or rods. A Voronoi cell needs to be calculated as a function of the angles defining the orientation of the nonspherical particles in analogy of the calculation between two particles of different radii. The integration over $P(r)dr$ in Eq. (16) is then replaced by integration over weighted orientational angles. The above analysis can also be extended to dimensions beyond three [8]. Although many of the approximations should work better in higher dimensions, some of the hypotheses (for example, the contact term ansatz) need to be reassessed. Thus, higher-dimension studies cannot be addressed as trivial extensions and need to be handled with care.

In summary, a theoretical framework is presented that predicts the RLP and RCP limits of a system of polydisperse spheres and brings together distinct results into a common theoretical framework. The formalism has the potential to solve other problems in condensed-matter physics such as the mixing and phase behavior of systems of hard particles of different shapes and size.

APPENDIX A: VOLUME FRACTION VALUES PREDICTED BY THE THEORY

We show in Tables I and II the volume fraction values predicted by the theory.

TABLE I. Volume fraction for $z=4$, RLP.

R_2/R_1 \ x	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9
1.3	0.5359	0.5376	0.5392	0.5405	0.5416	0.5423	0.5426	0.5423	0.5413	0.5393
1.5	0.5359	0.5393	0.5426	0.5456	0.5482	0.5503	0.5516	0.5517	0.5499	0.5453
1.7	0.5359	0.5409	0.5457	0.5506	0.5550	0.5588	0.5616	0.5628	0.5611	0.5359

TABLE II. Volume fraction for $z=6$, RCP.

R_2/R_1 \ x	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9
1.3	0.6340	0.6356	0.6370	0.6382	0.6392	0.6399	0.6402	0.6399	0.6389	0.6340
1.5	0.6340	0.6372	0.6401	0.6423	0.6453	0.6472	0.6484	0.6484	0.6468	0.6426
1.7	0.6340	0.6386	0.6431	0.6475	0.6515	0.6549	0.6575	0.6586	0.6571	0.6506

APPENDIX B: DISTRIBUTION OF SPHERE RADIUS

According to the theory, the average Voronoi volume for a packing with a distribution of radius $P(r)$ is given by the following self-consistent equation:

$$W = \frac{\pi}{2}\int dr P(r)\int_0^\infty c^2 \times \exp\left\{\int dR[-\rho_{rR}^s S_{rR}^*(c) - \rho_R(W)V_{rR}^*(c)]\right\}dc, \tag{B1}$$

where the different quantities are calculated as follows:

$$S_{rR}^*(c) = \int \Theta\left(c - \frac{l^2-k^2}{l\cos\theta - k}\right)ds = 2\pi\left(1 - \frac{l^2-k^2+kc}{lc}\right),$$

$$V_{rR}^*(c) = \int \Theta\left(c - \frac{r^2-k^2}{r\cos\theta - k}\right)dr^3 = 2\pi\left\{-\frac{1}{4c}[(c-k)^4 - l^4] + \frac{1}{3}[(c-k)^3 - l^3] + \left(\frac{k^2}{2c} - \frac{k}{2}\right)[(c-k)^2 - l^2]\right\}.$$

To simplify, we denoted $l=r+R$, $k=r-R$, and Θ the step function

$$\rho_R(W) = \frac{P(R)}{W - V_g},$$

$$\rho_{rR}^s = \frac{z_{rR}}{2\pi}\int_0^\infty \frac{z_{rr'}}{z_r}\sqrt{1-\left(\frac{r'}{r+r'}\right)^2}dr',$$

$$z_r = z\frac{A}{\langle S_r^{occ}\rangle},$$

$$\langle S_r^{occ}\rangle = \int_0^\infty 2\pi\left[1 - \sqrt{1-\left(\frac{r'}{r+r'}\right)^2}\right]P(r')dr',$$

$$A^{-1} = \int_0^\infty \frac{P(r')}{\langle S_{r'}^{occ}\rangle}dr',$$

$$z_{rr'} = \frac{z_r z_{r'} P(r')}{z}.$$

[1] *Unifying Concepts in Granular Media and Glasses*, edited by A. Conigliothe, A. Fierro, H. J. Herrmann, and M. Nicodemi (Elsevier, Amsterdam, 2004).

[2] J. D. Bernal and J. Mason, Nature (London) **188**, 910 (1960).

[3] J. Dodds, Nature (London) **256**, 187 (1975); K. de Lange Kristiansen, A. Wouterse, and A. Philipse, Physica A **358**, 249 (2005); M. Clusel, E. I. Corwin, A. O. N. Siemens, and J. Brujić, Nature (London) **460**, 611 (2009).

[4] I. Biazzo, F. Caltagirone, G. Parisi, and F. Zamponi, Phys. Rev. Lett. **102**, 195701 (2009).

[5] C. Song, P. Wang, and H. A. Makse, Nature (London) **453**, 629 (2008); C. Briscoe, C. Song, P. Wang, and H. A. Makse, Phys. Rev. Lett. **101**, 188001 (2008).

[6] L. Onsager, Ann. N.Y. Acad. Sci. **51**, 627 (1949).

[7] V. A. Luchnikov, N. N. Medvedev, L. Oger, and J.-P. Troadec, Phys. Rev. E **59**, 7205 (1999).

[8] J. A. van Meel, B. Charbonneau, A. Fortini, and P. Charbonneau, Phys. Rev. E **80**, 061110 (2009).