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Dense Packings of Random Binary Assemblies of Disks

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Abstract. — The most dense random packings of binary assemblies of hard disks in the plane are considered for the case of disks with a small difference in their radii. An expression for the dependence of the packing density on the relative difference in disk sizes, disk concentration, and mean value of the gaps is derived. The computer simulation of the random dense packing is carried out using various algorithms and theoretical results are confirmed. The gap distribution is shown to be of the exponential type for very dense random packings.

1. Introduction

Binary assemblies of disks in the plane have recently drawn the growing attention of researchers [1–4]. This can partially be explained by the fact that binary mixtures represent a transitional case between mono- and polydisperse systems, the latter being a more general case. At the same time the traditional interest for the structure of dense packings and mechanical properties of such systems, usually considered as models of granulated materials, is retained [5]. Moreover, it should be noted that at certain ratios of disk radii the crystallization of random binary assemblies in the plane does not take place; hence these systems can be convenient models to study the order-disorder [2] transition.

First the theoretical model that allows to establish an analytical relation between the parameters of dense packing of binary systems of hard disks was proposed by Dodds [1] for relative difference in disk radii $\Delta$ from 0 to $\approx 0.845$ ($\Delta = 1 - r_1/r_2$, $r_1$ and $r_2$ are the radii of small and large disk, respectively). At $\Delta > 0.845$ the difference in particle sizes becomes so large that it allows one to fit a small disk in the triangle formed by densely packed large disks. However this model can be considered only as a first approximation, since it does not take into account the presence of interparticle gaps.

Further the Dodds model was developed in the study by Troadec et al. [4], where the segregation in binary mixtures of disks at any packing fraction has been observed. In this study some statistical regularities in the neighbor distribution of the binary assembly of disks were obtained that allowed to analyze the local arrangements of particles. As shown, the assemblies built grain after grain by numerical simulations, without reorganization, behave like random packings. On the contrary, the assemblies formed on the air table [4] show a little segregation

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due to the building procedure that allows local reorganization. It is not evident whether the segregation still takes place when other numerical techniques are applied.

The method of system generation or realization of the most dense packing is a problem of importance. The simple compression of the rarefied gas of particles and the different modifications of the generation of random sequential packing without subsequent special relaxation procedure result in less dense packings, which are stable at hydrostatic compression. The reason is that 2D disk assembly show mechanical stability when the mean number of contacts with neighbors for each particle is equal to 4 [3,6]. In the paper of Polukhin [7] various methods of generation of dense systems are considered in detail. The relaxation procedures are shown to result in denser packing.

Here we present a detailed discussion of the regularities of the most dense random packing for binary systems of hard disks of similar sizes in the plane, as well as the results of computer simulation. In Section 2 a generalization of Dodds statistical model (taking into account small gaps) is proposed. In Section 3 the methods of generation of dense random packing are described, and the gap distributions in the systems obtained by these methods are given. In this section we also verify the extent of randomness in the generation of the packing. The ratios derived in Section 2 are verified in Section 4.

2. Statistical Model

2.1. Relation Between Packing Fraction and System Parameters. — Let us consider the random dense packing of \( N \) disks with similar radii, \( r_1 \) and \( r_2 \) (\( r_1 < r_2 \)), in the plane. Let us assume that \( r_2 = r \), \( r_1 = r(1 - \Delta) \), and numbers of disks of each type are \( N_2 = N\alpha \) and \( N_1 = N(1 - \alpha) \), respectively. Suppose that the condition of static equilibrium (that the average number of particle contacts is equal to 4) is fulfilled. At \( \Delta \ll 1 \) the mean gap value, \( G = \beta r \), will be low: \( \beta \ll 1 \). In this case, the number of neighbors for both types of disks is close to 6, i.e. the presence of two contacts and one gap between three neighbor particles can be assumed.

Let us consider four possible combinations of three neighbor particles forming Delone triangles: I - two large and one small disks, II - one large and two small disks, III - three large disks, and IV - three small disks. If the disk distribution in the plane is random and the mean gap value is independent of the combination of neighboring particles, the mean area, \( S_r \), of the corresponding Delone triangle and their number in the system, \( n_r \), can be calculated. The results obtained with linear approximation by \( \beta \) and \( \Delta \) for each case above are listed.

The total area of the system is equal to the sum of Delone triangle areas:

\[
S = \sum n_r S_r = 2\sqrt{3}N\pi r^2 \left[ 1 - 2(1 - \alpha)\Delta + \frac{\beta}{3} \right].
\]

For the area occupied by disks, \( S_d \), we have:

\[
S_d = \pi r^2 N \left[ (1 - \Delta)^2 (1 - \alpha) + \alpha \right] \approx \pi r^2 N [1 - 2(1 - \alpha)\Delta].
\]

Thus, the packing density, \( \rho \) (correct to second order by \( \Delta \)), is expressed as:

\[
\rho = S_d / S = (\pi / 2\sqrt{3}) \left( 1 - \frac{1}{3} \beta \right).
\]

If the determination of the mean gap value, \( \beta_c \), is carried out by averaging over all neighbors, then \( \beta = 3\beta_c \), and we have:

\[
\rho = \rho_0 (1 - \beta_c),
\]

where \( \rho_0 = (\pi / 2\sqrt{3}) \) is the density of hexagonal packing.
Thus, at low values of $\Delta$ and $\beta$ the packing density depends directly only on the mean gap value. It should be noted that in the absence of gaps the density corresponds always to the value for the hexagonal crystal and is independent of radii difference, as can be seen from (2).

2.2. Evaluation of Mean Gap Value. — Let us estimate the mean interparticle gap value, $\beta$. Assume that one large particle is surrounded by $6\alpha$ of large and $6(1 - \alpha)$ of small particles. Since each particle has six neighbors and four contacts with them, on the average four contacts and two gaps should be present between the disks of the first coordination shell. Then the summary length of two gaps is approximately equal to $12(1 - \alpha)\Delta r$, and taking into account that the concentration of large particles in the system is equal to $\alpha$, we have for $\beta_c$:

$$\beta_c \approx 2\alpha(1 - \alpha)\Delta.$$  \hspace{1cm} (3)

The substitution of the latter expression in (2) gives:

$$\rho = \rho_0[1 - 2\alpha(1 - \alpha)\Delta].$$  \hspace{1cm} (4)

At equal concentrations of disks of different sizes ($\alpha = 0.5$) we have for the system density:

$$\rho/\rho_0 = (1 - \Delta/2),$$  \hspace{1cm} (5)

where $\rho_0 \approx 0.907$, and $\rho_0$ becomes equal to 0.82 (a value characteristic of random disk packing) at $\Delta \approx 0.1$. In other words, the linear dependence of $\rho$ on $\Delta$ should be observed at $\Delta < 0.1$.

3. Generation of Dense Random Packing

Since the density and structure of random dense packings are strongly dependent on the procedure of their generation, we considered three methods differing in the degrees of relaxation. In the first case the relaxation is absent, in the second one a partial relaxation takes place, and in the third case the molecular dynamics (MD) method used results in different degrees of the “shaking down”. In all cases, binary systems of disks with radii $r_1$ and $r_2$ were considered.

3.1. Sequential Packing (SP). — The method of sequential packing was repeatedly used to generate dense packing [3]. In our case the disks were placed one by one on the plane $XY$ in the half-band $0 \leq X \leq X_{\text{max}}$ ($X_{\text{max}} \approx 100r$) and $0 \leq Y$ according to the following algorithm:

- the radius of a new particle was randomly defined corresponding to the given concentration of large and small disks;

- the $X$-th coordinate of the disk center was chosen in the segment $[0, X_{\text{max}}]$ using the generator of random numbers;

- the minimum value of the $Y$-th coordinate was found so that a new disk contacted either with the ordinate axis (then its position was fixed) or with another disk not overlapping with other particles;

- if the disk was in contact with a disk placed earlier, it was displaced along the latter toward the lower values of $Y$-th coordinate until it contacted with another disk or ordinate axis.

Using this algorithm the dense packings of 2500 disks for the systems with a dispersion $\Delta$ from 0 to 0.5 and concentration of large disks $\alpha$ from 0 to 1 were generated. In all cases the average number of contacts was equal to 4 with a high degree of accuracy; the density of such packings was always 0.82, that is lower, as compared to the systems generated using other algorithms. It is of interest that the gap size distribution was nearly uniform for these packings.
3.2. Dense Packing with Limited Relaxation (LR). — An ideal hexagonal crystal consisting of 2500 disks with the radius \( r_1 \) was chosen as an initial system; in this system a fraction of disks was randomly replaced by the larger ones, with the radius \( r_2 \). The disk overlap arising from this replacement was eliminated using the iteration procedure, all overlapping disk pairs being successively considered. To eliminate the overlap in the pair, a disk with larger \( X \)-th coordinate was displaced toward the higher \( X \) values, whereas a disk with larger \( Y \)-th coordinate was displaced toward the higher \( Y \) values. The displacement value during one cycle was no more then 0.2\( r \). As a result, the dense, but sufficiently “loose” (as for mechanical stability) disk packing was generated: the average number of contacts per particle was no greater than three.

In the following stage the hard disks were changed for elastic ones and a compressing force directed toward the cluster center was applied. To decrease the nonuniformity arising from hydrostatic effects the force value was made proportional to the distance from the cluster center. Using the displacement along the gradient of forces the equilibrium state was determined. The compressing force was gradually lowered during the relaxation process. As a result, a rather dense disk system was obtained, the disks being practically identical to absolutely hard ones, since the average disk overlap (the difference between the sum of disk radii and intercenter distance) was no more then 0.001\( r \) with the maximum overlap not exceeding 0.005\( r \). The average number of contacts per particle was close to 4 (the theoretical value) and the maximum error was no greater than 0.5.

The above algorithm was used to generate the dense packings for systems with a dispersion of up to 0.1 over a wide range of concentrations of large disks, the mean value of the contacts being always equal to 4 with a good accuracy. The density of the packings generated in such a way was dependent on \( \Delta \) and \( \alpha \) values, this is discussed in Section 4. The gap size distribution for these packings was close to the exponential type, unlike in the preceding algorithm.

However, in the first stage of the above procedure at \( \alpha > 0.5 \) a too rarefied system is obtained which does not relax with the formation of a high density state in the second stage. This can be clearly seen in the simulation of the dense packing from the identical large disks (\( \alpha = 1 \)) using the same procedure. In this case instead of a hexagonal crystal, a polycrystalline system with relatively low density is obtained.

3.3. Packing by the Method of Molecular Dynamics. — In this case the method of molecular dynamics of elastic disks was used. The elastic interaction potential, \( U_{\alpha \beta}(r) \), was introduced which was different from zero only for the disks with the intercenter distance, \( r \), less than the sum of their radii \( r_\alpha \) and \( r_\beta \):

\[ U_{\alpha \beta}(r) = k(r_\alpha + r_\beta - r)^2, \]

where \( \alpha \) and \( \beta \) are equal to 1 or 2 depending on disk type. The coefficient \( k \) defines the disk “hardness” and is equal to 100, the smaller disk diameter being always equal to 1. The numerical integration was made using the known algorithm [8], which seems to be very convenient for obtaining the equilibrium state of the system at given values of pressure and temperature.

The three-stage generation of the dense packings of hard disks was realized. At first, the particles were placed into the rectangular simulation cell with periodic boundary conditions, then the system was relaxed to the equilibrium state at the temperature \( T = 1 \) and the pressure \( p = 2 \) followed by gradual cooling to \( T = 0.3 \). Since the glass point of the above system is close to 0.2, at this stage we have the equilibrium state of the two-dimensional fluid of elastic disks.

In the second stage the system was cooled to zero temperature. Two cases were considered, i.e. the slow stepwise cooling with relaxation at each step, and the fast cooling. The type of
cooling had an important effect on the final packing density, the critical parameter being the cooling rate within the glass transition region, where the system structure was frozen.

In the third stage the pressure was lowered stepwise, with relaxation at each step. The linear dependence of density versus pressure was observed to excellent accuracy, allowing the reliable extrapolation of the pressure values to zero. An analogous extrapolation was accomplished for the mean values of gap and disk overlap, the maximum overlap during extrapolation to zero pressure being no more than 0.001\(r\).

3.4. Segregation at Local Scale. — The relationship (2) was derived assuming a random disk distribution in the system, more exactly, the random disk distribution by Delone triangles. This criterion of random packing is certainly rough, but some estimates can be made.

Let us consider the simplest case with 6 neighbors of each disk. As it has been noted in 2.1, there are four types of triangles with different compositions. We shall consider as a random one the packing in which the triangle fraction of each type corresponds to the assumption of equal probability of location of a disk with given diameter at any vertex of the triangle (taking into account the fraction of this type of disks in the system). The total number of triangles is \(2N\); the error in the determination of the number of triangles is equal to \(\sqrt{2N}\). If the number of simplices of a given type deviates from the random value within the error arising from the system finiteness, this packing shall be taken to be a random one. The random packing can be conveniently characterized by a generalized numerical parameter:

\[
\xi = \sqrt{\sum (N_m - N_c)^2 / \sqrt{N}},
\]

where \(N_m\) and \(N_c\) are the measured and calculated numbers of different types of triangles, respectively. For the random packing \(\xi \approx 1\) and when the system is separated into two phases \(\xi \approx \sqrt{N}\).

The corresponding calculations were carried out for the packings generated using the methods mentioned above. At low values of the relative difference in disk sizes (\(\Delta < 0.1\)) the concentrations of different triangles correspond to the random packing definition given here. The mean gap size and the number of gaps per one simplex were determined for each combination. Those values were found to be practically independent (to an accuracy of 0.1\%\) of the disk combination type, that correlates with the assumptions made during the derivation of (2) and (3).

4. Computer Simulation of Packing Fraction

LR and MD algorithms were used to generate the random dense packings for different concentrations of large disks and relative radii differences (\(\Delta\) values were in the range from 0.002 to 0.1). The density, \(\rho\), and mean gap, \(\beta_c\), values were calculated for each packing. The results obtained are given in Figures 1-4.

As can be seen in Figure 1, the packing fraction is lowered with a growing relative difference in particle sizes (in \(\Delta\) range from 0 to 0.1) and then remains unchanged. The dependence obtained by the LR algorithm was steeper and reaches the limit value of about 0.82 at \(\Delta \approx 0.1\). MD algorithm results in a dependence with linear initial part which is less sensitive to the \(\Delta\) and attains the limit value 0.84 at \(\Delta \approx 0.2\). As can be seen, the MD method allows to obtain more dense packings (\textit{i.e.} more relaxed), that is supported by a good correlation of MD simulation results with the theoretical predictions (5) for the maximum dense packing. The structures obtained by LR method proved to be less dense due to a relatively high concentration of "frozen" structure defects.
Fig. 1. — Density as a function of relative difference in disk radii. Solid line: theoretical results for \( \alpha = 0.5 \).

Fig. 2. — Density as a function of concentration of large disks. Solid lines - theoretical result.

As can be seen from Figure 2 the expression (4) is very suitable in \( \alpha \) range from 0.2 to 1. At a low concentration of large disks (\( \alpha < 0.2 \)) the distortions of the crystal structure look like gap bands of finite length. The presence of such a spatial distribution of gaps different from the random one results in the deviations from the theoretical dependence. The above bends begin to overlap with increasing concentration of large disk and at \( \alpha = 0.2 \) the spatial distribution of gaps becomes close to random.

In spite of a rather rough evaluation of the effect of relative difference in particle size on the mean gap value for the derivation of (3), the linear dependence of \( \beta_c \) on \( \Delta \) values was confirmed to a good precision for MD data (see Fig. 3). Hence the mean gap value in the most dense packings is defined by the first coordination shell. The mean gap value for the systems
obtained by the LR method appeared more sensitive to \( \Delta \) values. During the derivation of (3) the first coordination shell alone was considered and the coordination number in the system was always equal to 6. As has been shown in the LR numerical experiment, at the relative difference in disk sizes exceeding 0.002 a significant number of particles with neighbors differing from 6 appeared in the system; at \( \Delta = 0.08 \) and \( \alpha = 0.5 \) the concentration of such particles reached 15\%. In the system obtained by MD algorithm with the same \( \alpha \) and \( \Delta \) values the concentration of such particles was no more than 5\%.

In Figure 4 the dependence of packing density, \( \rho \), on the mean gap value, \( \beta_c \), is shown, among them for \( \alpha = 0.9 \) and 0.1. At low values of \( \beta_c \) (\( \beta_c < 0.05 \)) the results of numerical simulation are in good accordance with theoretical predictions. This relationship is more general since the derivation of (1) requires no assumptions on the dependence of \( \beta \) on \( \Delta \) and \( \alpha \) values. The pronounced deviations from linearity are only observed in the case of mean gap values as high
as 0.05; these deviations are due to the fact that the linear approximation by $\beta$ and $\Delta$ becomes incorrect.

The qualitative analysis of the structure suggests that the changes in the density and ordering degree at the crystal-glass transition correlate with each other in the $\Delta$ range from 0 to 0.1. The quantitative analysis of this problem will be done in the following paper.

5. Conclusion

The above results suggest that the binary system of disks in the plane can represent a convenient model to describe the transition region between the crystal and amorphous state. In this region mechanically stable packings are observed with intermediate values of packing fraction defined by the mixture composition (concentration of particles and relative difference in their diameters). The transition from the system state with high packing fraction corresponding to the hexagonal packing to the state related to the amorphous structure proved to take place in a narrow range of relative differences in the disk radii ($0 < \Delta < 0.2$). In this case the mean gap value seems to be one of the most convenient parameters to analyze the random structure of dense packings.

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