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Correlations and aggregate statistics in granular packs

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Abstract. We study how the aggregate statistical properties for density fluctuations in granular aggregates scale with the sample size and how such a scaling is associated with the correlations between grains. Correlations are studied both between grain positions and between Voronoï cell volumes, showing distinct behaviors and properties. A non-linear scaling in the aggregate volume fluctuations as function of the sample size is discovered and the connection between such anomalous scaling and correlations is explained. It emerges that volume fluctuations might be described by means of a single universal equation for all samples at all cluster sizes.

PACS. 45.70.-n Granular systems – 45.70.Cc Static sandpiles; Granular compaction – 81.05.Rm Porous materials; Granular materials

The study of volume fluctuations in large granular aggregates could be the way to validate some statistical mechanics theoretical descriptions of granular matter [1–11], and therefore studies in this domain have recently attracted a large interest from the scientific community [12,9,13]. In a recent paper [13] we have shown that, at *local level*, the fluctuations of the Voronoï cell volumes can be predicted with a remarkable precision by using a distribution function – with no adjustable parameters – which reveals a universal dependence on the packing fraction of the sample. On the other hand, in most experiments the only measurable parameters are the *global* volume fluctuations on the whole sample. The generalization to large aggregates of the local theoretical distribution for the Voronoï cells depends on the presence of correlations. More generally, to understand correlations is essential in any statistical mechanics approach of any given system because the correlation length establishes the level of detail at which the system description must be tuned. In this paper we study correlations and their effects on the scaling laws of the volume fluctuations.

The structure of granular materials is disordered but not random. These systems present a spontaneous organization which can be measured both at local and at global level. Such an organization is the consequence of several different mechanisms which are both physical (e.g. mechanical stability) and geometrical (e.g. close packing configurations). One of the key-questions, which we address in this paper, is to identify the length scale at which structural organization is present. In other words, we want to identify the length-scale above which correlations are smearing out and average quantities become the relevant control parameters.

In this paper we tackle this question by looking at changes in the statistical properties as function of the size of the packing-aggregates. Specifically, we compute two different kinds of correlations: (1) correlations between grain positions; (2) correlations between local Voronoï cells constructed from the grain centers. In particular we calculate the first kind of correlation by dividing the sample in cubic grids of different sizes and comparing the occupation numbers in adjacent grid-units. The second kind of correlation is computed in two different ways: (a) a direct method is used for adjacent Voronoï cells; (b) an indirect method is used at larger scales where the average correlation between couples in clusters is computed from the dependence of the aggregated distribution of the Voronoï volumes on the cluster sizes.

Such analysis concerns six experimental samples made of monosized acrylic beads prepared in a cylindrical container and having packing fractions ranging between 0.58 to 0.64 [14–17]. Such a dataset was acquired by means of X-ray Computed Tomography and it records the positions of more than 385 000 sphere centers. The precision on the coordinates is better than 0.1 % of the sphere-diameters and the sphere polydispersity is within 2 %. In this paper we refer to these samples with labels A, B, C, D, E and F which are the same used to respectively identify the samples in the previous papers [14–17] where other kinds of structural analysis were performed. The present investigations are performed over an internal region (**G**) at 4 sphere-diameters away from the sample boundaries. (Spheres outside **G** are considered when computing the

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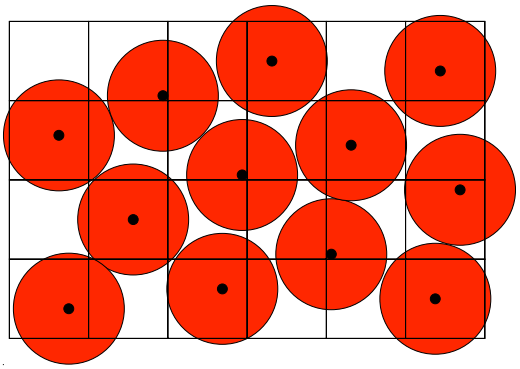

Fig. 1. To calculate correlations, the internal part of each sample is subdivided into cubic grids with different sizes and the occupation numbers of each grid unit $n(x, y, z)$ are compared.

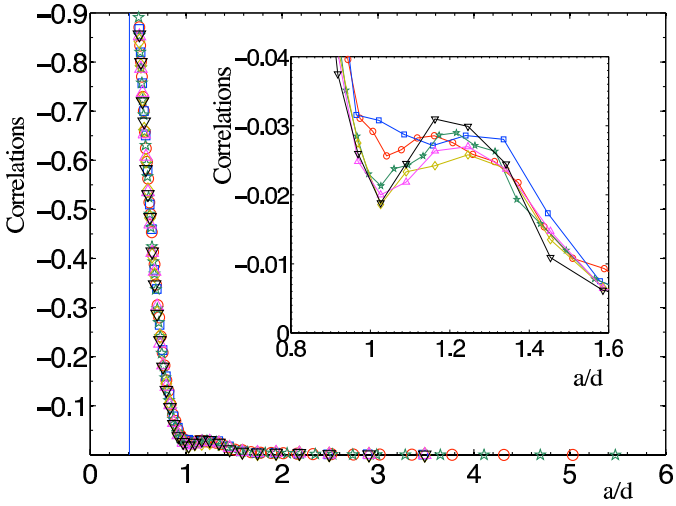

Fig. 2. Correlations between beads centers calculated with the grid method as function of the relative grid size a/d.

neighboring environment of spheres in **G**.) Samples A and C contain about 140 000 beads each (90 000 in the inside). Whereas, samples B, D-F contain about 35 000 beads each (15 000 in the inside). Further details are reported in references [14–17].

The paper is organized as follows. In Section 1, we calculate the correlations between sphere centers by using several grid partitions. In Section 2, we calculate the scaling of the variance of the aggregated Voronoï volumes distribution and we derive the associated correlations among Voronoï cells. In Section 3, we show how the aggregated distribution of the Voronoï volumes is affected by correlations among cells by solving explicitly the one-dimensional case for Poisson points on a line. Section 4 gives concluding remarks and perspectives.

1 Correlations between beads positions from a grid-partition

There are several possible ways to calculate geometrical correlations between objects in space. In this section we calculate such correlations by subdividing the space inside each sample in a cubic grid and comparing the number of sphere centers belonging to each unit. Figure 1 shows a schematic example of such a grid in two dimensions. In three dimensions, each unit is a cube with edge length a and it is labeled by the symbols (x, y, z) with $x = 1...N_x$, $y = 1...N_y$ and $z = 1...N_z$. Samples B, D-F have been subdivided into 30 different grids with unit sizes between $0.5 \leq a/d \leq 3.5$ (where d is the bead diameter), resulting in a total number of units between $34 \times 34 \times 34 \sim 39\,000$ and $5 \times 5 \times 5 = 125$. Whereas samples A and C have been subdivided into 60 different grids with unit sizes between $0.5 \leq a/d \leq 6.5$, with total number of units between $60 \times 60 \times 60 = 216\,000$ and $5 \times 5 \times 5 = 125$. For each grid, the correlations were evaluated by comparing the number of sphere centers in neighboring couples of grid-units that are sharing a face. Specifically, if we call $n(x, y, z)$ the number of sphere centers inside the unit identified by the labels (x, y, z), then the correlation in the x direction is defined as:

$$C_x(a) = \frac{\langle n(x,y,z)n(x+1,y,z)\rangle}{\langle n(x,y,z)\rangle \langle n(x+1,y,z)\rangle} - 1, \qquad (1)$$

where the symbols $\langle(...)\rangle$ represent the following average:

$$\langle g(x,y,z)\rangle = \frac{1}{(N_x-1)N_yN_z} \sum_{x=1}^{N_x-1} \sum_{y=1}^{N_y} \sum_{z=1}^{N_z} g(x,y,z) \quad , \qquad (2)$$

with $g(x, y, z)$ any generic function of x, y, z. The correlations in the other two directions y and z are defined straightforwardly by analogy with equations (1) and (2) and the total correlation can be defined as $C(a) = [C_x(a)+C_y(a)+C_y(a)]/3$. Such quantity measures the covariance of the occupation number in adjacent grid-units normalized by the average number of couples of grains in the two units, making it therefore an average covariance per couple of grains.

Figure 2 reports the values of $C(a)$ measured in the six experimental samples A-F. Not surprisingly, we find only negative correlations. Indeed, in these systems there are no attractive forces between beads and therefore there are no mechanisms that spontaneously tend to gather beads together. The large negative correlations below $a/d = 1$ are the effect of excluded volume: two spheres cannot stay too close to each other. It is easy to calculate that the presence of a sphere in a given unit will completely exclude the possibility to find another sphere in a neighboring unit when $a < 1/\sqrt{6}$. The vertical line in Figure 2 indicates such a threshold value below which $C(a)$ must become equal to -1. The small resurgence of negative correlations above $a/d = 1$ (see inset in Fig. 2) might be due to mechanical equilibrium which induces some spheres to stay in contact creating in this way a larger region of excluded volume around a given sphere and its neighbors in contact. It is interesting to note that the behavior of $C(a)$ is

very similar in all the samples and therefore it is little related to the packing fraction and sample preparation. The detailed plot in the inset of Figure 2 shows that the main differences are, eventually, in the region around $a = 1$ where the packings with lowest densities are slightly more anti-correlated. A fit of $C(a)$ with the exponential law $\exp(-a/\Lambda)$ in the region $a > d$ might indicate a characteristic correlation length $\Lambda \sim 0.6d$. However, the range of sizes a is too small to produce a proper fit. Indeed, a power law kind of decreasing trend cannot be excluded from this set of data. For instance, we observed that the correlation coefficient between the occupation numbers $n(x, y, z)$ in adjacent cells is oscillating around the value -0.5 for all the sizes a measured. Such a constant correlation coefficient would imply that $C(a)$ must decrease as a power law with exponent -6. Larger samples must be investigated to address properly this matter.

Furthermore, we must stress that, despite the fact that $C(a)$ is fast decreasing, other measures and other effects of correlations remain sizable even at relatively large distances. One of such effects can be observed from the variance of the occupation numbers $\langle n(x,y,z)^2\rangle - \langle n(x,y,z)\rangle^2$, which reveals a scaling as $\langle n(x,y,z)\rangle^\gamma$ with exponent $\gamma \sim 0.71$ which is smaller than the exponent ($\gamma^* = 1$) expected for the scaling of independent quantities. The covariance between occupation numbers in adjacent cells also reveals a non-linear scaling with exponent ~ 0.73. All these experimental observations show that geometrical correlations in these systems produce important sizable effects even at large scales.

In the next section we will see that similar effects are also emerging from the study of the aggregated distribution of Voronoï volume fluctuations.

2 Scaling of the aggregate distributions and correlations

We have recently discovered [13] that in three dimensional packings of equi-sized beads the empirical distribution for the Voronoï cells volumes is remarkably well described by the following distribution

$$f(V,k) = \frac{k^k}{(k-1)!}\frac{(V-V_{min})^{(k-1)}}{(\langle V\rangle - V_{min})^k}\exp\left(-k\frac{V-V_{min}}{\langle V\rangle - V_{min}}\right), \quad (3)$$

with V_{min} the minimum attainable Voronoï volume in a packing of spheres with equal diameters d (which is, $V_{min} = 5^{(5/4)}/\sqrt{2(29+13\sqrt{5})}d^3 \simeq 0.694d^3$) and $\langle V\rangle$ the average Voronoï volume (that is $\langle V\rangle = \pi d^3/(6\rho)$, with ρ the packing fraction). The theoretical framework that yields to equation (3) assumes that each Voronoï cell is made from the contribution of k 'elementary cells'. The analysis of a very large dataset containing more than 2 millions beads revealed a very good agreement between $f(V,k)$ and the empirical distributions for $k \simeq 12$ [13], indicating that about 12 elementary cells are contributing to building each Voronoï cell. The exact geometrical nature of such elementary cells is not known, the only requirement is that they must be space-filling and any given combinations of such cells must yield to a mechanically-stable packing. From a statistical mechanics perspective one can see that such 'elementary cells' are the independent degrees of freedoms which are contributing to the aggregate volume.

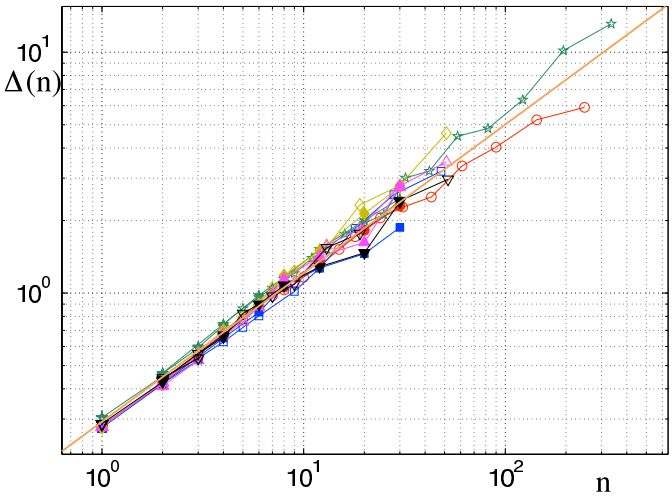

Fig. 3. Plot of $\Delta(n) = \sigma_v^2(n)/(\frac{\pi}{6\rho}d^3 - 0.694d^3)^2$ as function of the cluster size n. The variance of the aggregate distribution of clusters of n Voronoï cells scales as $\sigma_v^2(n) \propto n^\beta$ with $\beta \simeq 1.23$. Empty symbols are associated to statistics on clusters gathered with method (a), whereas full symbols correspond to the gathering method (b) – see text. The data from all the samples A-F collapse onto a common behavior when $\sigma_v^2(n)$ is divided by the factor $(\frac{\pi}{6\rho}d^3 - 0.694d^3)^2$.

There are two interesting features that arise from the reasoning that leads to equation (3): (i) all the observable statistical properties must collapse onto a universal behavior, independent on the packing fraction, when plotted as function of $(V - V_{min})/(\langle V\rangle - V_{min})$; (ii) equation (3) is valid for *any* aggregate of k elementary cells and therefore it must also describe how the statistical properties scale with the size of the cluster investigated. In this respect, these two properties lead to the following expression for the variance of the aggregate distribution of n Voronoï cells:

$$\sigma_v^2(n) = \frac{n^2 k(1)}{k(n)}\sigma_v^2(1) = \frac{n^2}{k(n)}\left(\frac{\pi d^3}{6\rho} - 0.694d^3\right)^2, \quad (4)$$

with $k(n)$ the number of elementary cells contributing to the aggregate of n Voronoï cells.

Let us first verify whether this theoretical prediction is backed by the empirical observations. In particular, we already observed in [13] that – indeed – the distribution for the Voronoï cells volumes of a large set of different empirical samples (including the six samples A-F discussed in this paper) collapses onto a single universal curve (well described by $f(V, k = 12)$) when plotted vs. $(V - V_{min})/(\langle V\rangle - V_{min})$. We now want to verify if such a collapse holds also for the aggregated distributions of

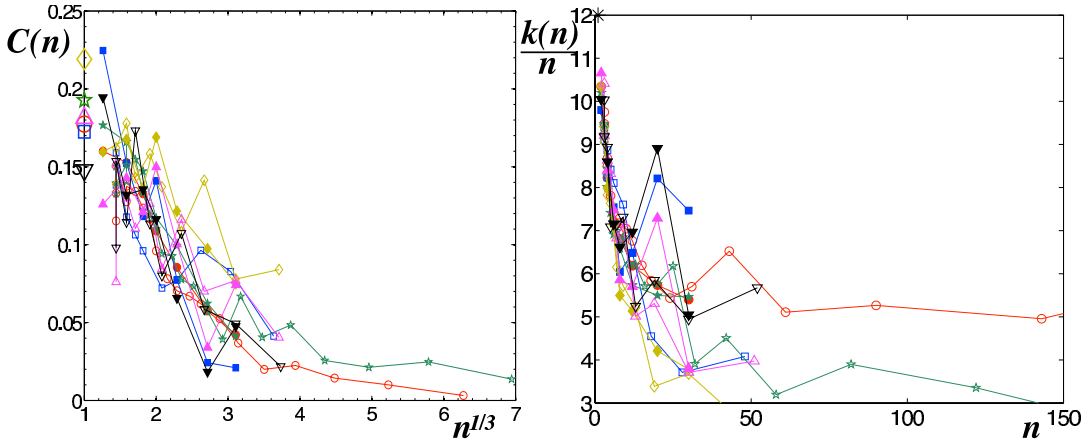

Fig. 4. *(Left)* Average correlation between Voronoï cells in a cluster of size n, calculated from equation (7). The empty and full symbols are associated to the two different gatherings (a) and (b) — see text. The large empty symbols at $n = 1$ are the correlation coefficients computed between couples of neighboring Voronoï cells. *(Right)* The behavior of $k(n)/n$ vs. n shows that the average number of elementary cells per Voronoï cell is decreasing with the cluster size n and it seems to settle at a value around $k(n)/n \sim 3$ for $n \to \infty$. The large fluctuations are mostly due to the bad statistics at large n.

clusters of Voronoï cells. To this purpose we use the grid partition introduced in the previous section and we collect clusters of n Voronoï cells by using two distinct methods: a) for all grid-sizes a we take all the units containing n sphere centers; b) for each grid-size a we take all the units containing a number of sphere centers equal to the smallest integer which is larger or equal to the average unit occupation number. For each one of these two methods and for each sample A to F, we collect all the clusters containing n Voronoï cells, we calculate the sum of the Voronoï volumes in each cluster and we compute their variance $\sigma_v^2(n)$.

In Figure 3, we show that, accordingly with equation (3), also the aggregated distributions and in particular their variances $\sigma_v(n)$ collapse on a common behavior when divided by the factor $\pi d^3/(6\rho) - 0.694d^3$. Controversially, we observe that $\sigma_v^2(n)$ does not scale linearly with the cluster size, showing instead a power law behavior $\sigma_v^2(n) \propto n^\beta$ with exponent $\beta \simeq 1.23$. A similar scaling was found by Lechenault et al. [18] for two dimensional disk packings. However, this non-linear scaling is not necessarily in contradiction with equation (3) which applies to clusters of k *elemetary* cells; it simply tells that $k(n)$ has a non-linear dependence on n. Indeed, deviations from the linear scaling can arise from correlations among cells in the aggregate. Elementary cells are uncorrelated local volumes but this does not exclude correlations between aggregates of elementary cells, such as Voronoï cells, if they are partially overlapping. This point will be clarified in Section 3 by means of an example in one dimension. Let us here, formally show such a link between the scaling of $\sigma_v^2(n)$ and correlations by explicitly writing such variance: $\sigma_v^2(n) = \left\langle (\sum_{i=1}^n v_i)^2 \right\rangle - \sum_{i=1}^n \langle v_i \rangle^2$, which can be expanded to

$$\sigma_v^2(n) = n\sigma_v(1)^2[1 + (n-1)\mathcal{C}(n)] \qquad (5)$$

where $\mathcal{C}(n)$ is the average correlation per cell in the cluster of n Voronoï cells

$$\mathcal{C}(n) = \frac{1}{n(n-1)} \sum_{i,j \neq i} \frac{\langle v_i v_j \rangle - \langle v_i \rangle \langle v_j \rangle}{\langle v^2 \rangle - \langle v \rangle^2} \quad . \qquad (6)$$

Therefore equation (6) shows that any deviation from the linear scaling is directly associated to correlations. It follows that correlations can be directly calculated from such deviations:

$$\mathcal{C}(n) = \frac{\sigma_v^2(n) - n\sigma_v^2(1)}{n(n-1)\sigma_v^2(1)} = \frac{1}{n-1}\left(\frac{nk(1)}{k(n)} - 1\right) \quad . \qquad (7)$$

Let us therefore make use of this equation and calculate, from the scaling law of $\sigma_v^2(n)$, the average correlations between couples of Voronoï cells in a cluster.

Correlations from scaling

Figure 4 shows the behavior of $\mathcal{C}(n)$ calculated from the statistics on the clusters made of n Voronoï cells gathered with the two methods (a) and (b) described previously. The data are plotted as function of $n^{1/3}$ which gives an information about the linear cluster size. We first note that, despite the fact that the correlations among points are negative (Fig. 2), here the Voronoï cells are positively correlated indicating that – for instance – near to a large Voronoï cell it is more likely to find another large Voronoï cell than a small one. Second, we note that small clusters ($n \leq 4$) have correlations between 0.1 and 0.2. These numbers are consistent with the correlation coefficients between neighboring Voronoï cells calculated by using a direct independent method. The large empty symbols at $n = 1$ in Figure 4 report such correlation coefficients. We also note that correlations decrease fast becoming negligible for distances above three sphere diameters. However,

we must stress that the geometrical correlations for the positions $C(a)$ had a much faster decay and we should also note that despite such small values, these correlations lead to sizable effects on $\sigma_v^2(n)$ for rather large n.

Equation (7) gives us another important insight to understand the non-linear scaling behavior of $\sigma_v^2(n)$. Indeed, we observe that $k(n)/n = n\sigma_v^2(1)/\sigma_v^2(n)$ indicating that the non-linear scaling law of $\sigma_v^2(n)$ can be associated to the scaling behavior of the average number of elementary cells per Voronoï cell in the cluster. Indeed, the scaling of this quantity is not trivial because it depends on the way that elementary cells and Voronoï cells combine. Figure 4 shows that for clusters containing more than 50 cells $k(n)/n$ appears to settle down to a constant value. This might indicate that the effects of correlations will smear out for sizes above 2-3 sphere diameters (a cluster containing 50 cells has linear size $\sim 3d$). However, the sample-sizes do not allow us to get a reliable statistics above $n \sim 30$ making very difficult to derive any definite conclusion.

It is important to stress that, even in the case in which the elementary cells are assumed to be independent and uncorrelated, the resulting Voronoï cells and their aggregated clusters will be *correlated* if they are sharing elementary cells. In the next section we will clarify some of these points by studying a one-dimensional example which has the great advantage to be analytically solvable.

3 Correlations and aggregate Voronoï distributions from random points in one dimension

Let us consider a one-dimensional system where points are placed at random on a line with average distance equal to λ (Poissonian points). In this case, it is known that the probability to find a gap of length l between two points is $p_{del}(l) = 1/\lambda \exp(-l/\lambda)$ and, by construction, such intervals are uncorrelated. In this case, these intervals are the Delaunay cells and they are also the 'elementary cells' in one-dimension [13]. The Voronoï cell 'i' can be built by taking the segments between the two mid-points between three successive points $i-1$ and i, and between i and $i+1$. The probability to find a Voronoï cell of size v can be computed from the probability of finding two successive Delaunay segments respectively with lengths l_1 and l_2 such that $l_1 + l_2 = 2v$, obtaining

$$p_{vor}(v) = \int_0^\infty \int_0^\infty p_{del}(l_1)p_{del}(l_2)\ \delta(\frac{l_1+l_2}{2} - v)dl_1dl_2 = \frac{4v}{\lambda^2}e^{-2\frac{v}{\lambda}} \quad , \quad (8)$$

which coincides with equation (3) for $k = 2$.

Correlations among neighboring Voronoï cells can be calculated from the combined probability to find one Voronoï cell of size v_1 next to a Voronoï cell of size v_2, which is

$$p_{com}(v_1,v_2) = \int_0^\infty \int_0^\infty \int_0^\infty p_{del}(l_1)p_{del}(l_2)p_{del}(l_3) \times\delta(\frac{l_1+l_2}{2} - v_1)\delta(\frac{l_2+l_3}{2} - v_2)dl_1dl_2dl_3 = \frac{4}{\lambda^2}e^{-2\frac{v_1+v_2}{\lambda}}(e^{-2\frac{\min(v_1,v_2)}{\lambda}} - 1) \quad . \quad (9)$$

From the above expression one can calculate that the correlation coefficient between neighboring Voronoï cells for random points in one dimension is:

$$\frac{\langle v_1v_2\rangle - \langle v_1\rangle\langle v_2\rangle}{\langle v^2\rangle - \langle v\rangle^2} = \frac{1}{2} \quad . \quad (10)$$

Interestingly, as we stressed before, it formally results that the Voronoï cells are *positively correlated* even if their construction starts from *uncorrelated* random points.

In this one-dimensional case we can also compute exactly the form of the aggregate Voronoï distribution as function of the cluster-size. In particular, we know already that the aggregate of k Delaunay cells has the distribution given by equation (3) (with $V_{min} = 0$ and $\langle V\rangle = k\lambda$). We can note that an aggregate of n Voronoï cells with size v can be constructed by the combination of a central cluster made of $n-1$ Delaunay cells with total size x plus two Delaunay cells of sizes l_1 and l_2 (at the right and left ends) such that they sum to $l_1/2 + l_2/2 + x = v$. This implies that the aggregate distribution of n Voronoï cells can be written as

$$p_{vor}(v,n) = \int_0^\infty \int_0^\infty \int_0^\infty p_{del}(l_1)f(x,n-1)p_{del}(l_2) \times \delta(\frac{l_1+l_2}{2} + x - v)dl_1dl_2dx \quad . \quad (11)$$

This equation reveals that the probability distribution of the aggregated Voronoï volumes is more complex than the simple generalization of equation (3) that one could obtain by substituting k with $k(n)$. However, such substitution is exact in the two limits $n = 1$ and $n \to \infty$ and it turns out to match quite well $p_{vor}(v,n)$ by using $k(n) = 2n^2/(2n-1)$ which is the scaling law predicted by equation (4). Indeed, from equation (11) one can calculate explicitly the variance of the aggregate distribution, obtaining

$$\sigma_v^2(n) = \lambda^2(n - \frac{1}{2}) \quad , \quad (12)$$

which coincides with $\sigma_v^2(n) = n^2k(1)/k(n)\sigma_v^2(1)$ when $k(n) = 2n^2/(2n-1)$. In this case the resulting scaling law for $\sigma_v^2(n)$ is linear but the value of the variance is almost twice the value expected for uncorrelated cells: $n\sigma_v^2(1) = n\lambda^2/2$. We can note that this expression is consistent with equations (5) and (6). Indeed, $\sigma_v^2(1) = \lambda^2/2$ and therefore from equation (7), $\mathcal{C} = 1/n$ which is the result one obtains by imposing a correlation coefficient equal to 1/2 (Eq. (10)) between the $n-1$ adjacent couples of cells in the cluster. The scaling law for $k(n)$ is instead not

linear (and it is similar to the one observed in Figure 4) revealing that the effective number of degrees of freedom per Voronoï cell is decreasing from 2 at $n = 1$ to 1 when $n \to \infty$.

4 Conclusions and perspectives

We have studied correlations in packings made with equi-sized beads. We observed the presence of correlations both between the beads-centers positions and between the Voronoï volumes. It results that correlations between the beads positions are negative and short-ranged but we observed effects that rest persistent over relatively large sizes. However, from the available experimental data it was not possible to obtain a precise estimation of the correlation length. In particular, we observed that correlations become very small already at distances above three sphere diameters. On the other hand, we also observed that the law of decay could be a power law, implying therefore infinite correlation length. Similarly, we observed sizable effects on the scaling of the aggregate Voronoï volume fluctuations with the cluster size. In particular, we measured that the variance of the aggregated Voronoï volumes in clusters of n cells scales as $\sigma_v^2(n) \propto n^{1.23}$. We have shown that such effect is the consequence of a different kind of correlation that is present among Voronoï cells. How these two kinds of correlations are related is unclear and an example in one-dimension reveals that Voronoï cells can be correlated even when generated from random points. We have discussed that the non-linear scaling law for the aggregated volume fluctuations in clusters on n grains can be consistently described with equation (3) by assuming that the number of elementary cells in the cluster ($k(n)$) might not scale linearly with n. From a statistical mechanics perspective, the quantity $k(n)$ must be interpreted as the number of degrees of freedom contributing to the volume fluctuations in the aggregate cluster. When a single Voronoï cell is concerned the positions of all its neighbors are contributing to its volume and in this case the value of the parameter $k(1) \sim 12$ is a sensible empirical finding. On the other hand for large clusters the degrees of freedoms cannot be larger than $3n$. Indeed, only the grain positions are determining the Voronoï volumes. An estimate of $k(n)/n$ as function of n reveals that the number of degrees of freedom per grain decreases from $k(1) \sim 12$ to some plateau value $k(n)/n \sim 3$ when the cluster size becomes larger than 50 grains. However, the present experiments cannot give reliable statistics for clusters larger than 30-50 grains and therefore it is difficult to estimate if $k(n)/n$ is settling down to a constant plateau. To answer this point, the study of fluctuations in large aggregates must be undertaken. This must be done by looking at the global volume fluctuations of the whole sample in different repeated trials.

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