Volume Fluctuations and Geometrical Constraints in Granular Packs

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(Received 15 July 2005; revised manuscript received 3 October 2005; published 12 January 2006)

I study the structural organization and correlations in very large packings of equally sized spheres, reconstructed in three dimensions with x-ray computed tomography. I show that the geometrical structure can be conveniently studied as a packing of irregular tetrahedra with volume distribution that must decay exponentially with parameters controlled by the conditions of mechanical stability, nonoverlapping, and space filling. I argue that the system's structure can be described as constituted of two phases: (1) an ''unconstrained'' phase which freely shares the volume and (2) a ''constrained'' phase which assumes configurations accordingly with the geometrical constraints. It results that the granular system exploits heterogeneity maximizing freedom and entropy while constraining mechanical stability.

DOI: 10.1103/PhysRevLett.96.018002 PACS numbers: 45.70.−n

In this Letter I investigate how space is shared among the particles in a granular pack and I discuss how the study of such a space partition is essential for understanding both the static properties of these structures and the dynamical mechanisms which generate them. When equal spheres are packed in a container they can arrange in a way to minimize potential (gravitational) energy by maximizing the packing fraction. The pursuit of maximum compaction is common to several physical systems and, at atomic level, it is a feature associated with metallic bounding. From a purely geometrical perspective, it is known that the largest attainable packing fraction in a system of equal spheres is $\rho = \pi/\sqrt{18} \sim 0.74$ [1,2], which corresponds to a stack of parallel hexagonal layers of spheres (forming the so-called Barlow packings). Conversely, it is observed empirically that when balls are poured in a container they spontaneously arrange in a disorderly fashion occupying a fraction of the total volume between 0.555 and 0.64. The study of these disordered structures is very challenging and the available investigation tools appear to be inadequate to capture their essential features. Indeed, a complete description of the structure of a disordered system requires a very large amount of information about coordinates, orientations, shapes, and connectivities of all the elements. It is, however, clear that not all this information is necessary to determine the properties of these systems. On the contrary, there exist several states with different microscopic realizations which share the same macroscopic properties.

One of the challenges of the research in this field is to find a simple measure which characterizes the state of the system giving information about the packing structure and its properties [3–8]. In this Letter, I show that a disordered sphere pack can be described in terms of a simple parameter which depends on the packing fraction and on a simple topological property. This is demonstrated by *first* searching for the local structural motifs which make the ''building blocks'' of such systems. *Second*, by exploring the allowed local fluctuation of the volumes of such building blocks, and predicting their volume distribution. *Third*, by comparing the theoretical predictions with the experimental results.

The experimental data reported in this Letter are based on the analysis of the largest empirical data set presently available in the literature [9]. Such a data set is constructed from the study, by means of x-ray computed tomography, of large samples of disorderly packed monosized spheres. This database records the positions of more than 385 000 sphere centers from 6 samples of acrylic beads prepared in a cylindrical container. The precision on the coordinates is better than 0.1% of the sphere diameters and the sphere polydispersity is within 2%. In this Letter I refer to these samples with labels A, B, C, D, E, and F; their packing fractions and sample sizes are reported in Table I. The investigations reported in this Letter are performed over an internal region (G) 4 sphere diameters away from the sample boundaries. (Spheres outside G are considered when computing the neighboring environment of spheres in G.)

The search for the elementary building blocks is performed by introducing a new technique to investigate the structural correlations among the packed spheres. This analysis is based on two important definitions: *bounded* spheres and *common neighbor* [10]. In particular, two spheres are defined ''bounded'' if they stay within a given

TABLE I. Sample density and their intervals of variations (±) within each sample, total number of spheres (N), number of spheres in the central region (N_G), and average number of incident Delaunay neighbors $\langle f \rangle$.

	Packing fraction	N	N_G	$\langle f \rangle$
A	0.586 ± 0.005	102 897	54719	14.6
B	0.596 ± 0.006	34016	15013	14.6
C	0.619 ± 0.005	142 919	91984	14.4
D	0.626 ± 0.008	35511	15725	14.4
E	0.630 ± 0.01	35881	15852	14.4
F	0.640 ± 0.005	36461	16247	14.3

threshold radial distance $\tilde{r}$, whereas a "common neighbor" of two bounded sphere is a third sphere which is also bounded to both the two spheres. It can be calculated that the maximum number of common neighbors which can be placed around two bounded spheres is equal to 5 for any threshold distance smaller than $\tilde{r} \leq \sqrt{5/4}d \simeq 1.118d$, where d is the spheres' diameter. Fixed a threshold distance, the numbers of configurations with n common neighbors is a very sensitive measure of the local organization. For instance, when the threshold distance is $1.1d$, the fraction of configurations with 4 common neighbors increases sensibly during compaction varying from 17% at $\rho = 0.586$ (A) to 31% at $\rho = 0.640$ (F). Similarly, the configurations with 5 common neighbors increase from less than 3% to above 8% when packing fraction varies between 0.586 to 0.640 (A to F).

The fact that the number of common neighbors is so sensitive to the packing properties suggests that the study of the local organization around couples of bounded spheres could be the key to understand the structure of these systems. To this end, in this Letter, I introduce a technique to reveal how common neighbors are distributed. This analysis consists of the study of the dihedral angles between common neighbors around couples of bounded spheres. These angles are calculated by first constructing a triangular face with two vertices occupied by a couple of bounded spheres and the third vertex occupied by the common neighbor. Then the dihedral angles are measured between such a triangle and all the other triangles formed with the other neighbors common to the original couple of spheres. The resulting distribution of angles is shown in Fig. 1 (for threshold distance $\tilde{r} = 1.1d$). Such distribution is symmetric in θ and $360 - \theta$ and has two large peaks at $\theta = \arccos(1/3) = 70.5\ldots$ and $360 - \arccos(1/3) = 289.4\ldots$. These values coincide with the dihedral angles in a regular tetrahedron. Other two (smaller) peaks are also visible at $\theta = 2\arccos(1/3) = 141.0\ldots$ and $\theta = 218.9\ldots$. They also correspond to configurations made of two touching regular tetrahera. These peaks clearly indicate that the common neighbors tend to gather together forming tetrahedral packings. It is worth noting that the

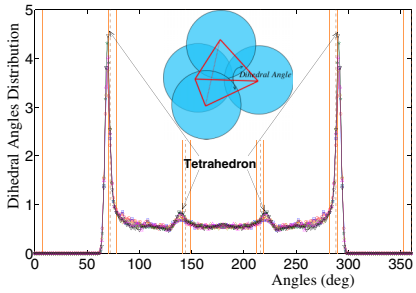

FIG. 1 (color online). Dihedral angle distribution (x axis: angular degrees; y axis: renormalized frequencies). The vertical lines indicate the angles $\theta = n\arccos(1/3)$ (and $360 - \theta$) with $n = 1, 2, 3, 4, 5$ (tetrahedral packings). The dashed lines are at the angles $\theta = n360/5$ ($n = 1, 2, 3, 4$).

essential features of this distribution, and, in particular, the position of the peaks, are a little sensitive to the choice of the threshold. Indeed, the same kind of distributions are obtained for different values of the threshold distance in a range between $1.0d$ and $1.11d$. A detailed analysis of the subset of configurations with dihedral angles in the interval within $\arccos(1/3) \pm 1$ degree [and within $360 - \arccos(1/3) \pm 1$] confirms that they are originated by tetrahedral configurations. In particular, these configurations are very regular tetrahedra with edge lengths between $0.99d$ and $1.01d$ and volumes which take values within the limits $0.11d^3$ and $0.13d^3$ in 99% of the configurations (a regular tetrahedron with edge lengths equal to d has volume $v^* = \sqrt{2}/12d^3 \simeq 0.118d^3$).

The analysis of the dihedral angles distribution has revealed that these amorphous structures can be conveniently viewed as the result of a packing of tetrahedra. There is a natural way to subdivide a structure into a system of tetrahedra. This is the *Delaunay decomposition* which constructs a system of minimal tetrahedra with vertices on the centers of neighboring spheres chosen in such a way that no other spheres in the pack have centers within the circumsphere of each Delaunay tetrahedron. One of the advantages of such decomposition is that it does not require the introduction of any threshold. The Delaunay decomposition uniquely associates the packing of N particles with space-filling systems of T tetrahedra, with

$$T = \left(\frac{\langle f \rangle}{2} - 1\right)N, \tag{1}$$

where $\langle f \rangle$ is the average number of tetrahedra incident on each particle. In general, $\langle f \rangle$ takes values in the narrow range between $14 \leq \langle f \rangle \leq 2 + 48\pi^2/35 \simeq 15.53$, with the lower limit corresponding to close packed configurations and the upper limit associated with a "granular gas" of randomly positioned particles [11]. In mechanically stable equal-spheres packings, under gravity, this interval of variation reduces further with typical values around $\langle f \rangle \sim 14.5$. This is—for instance—the case across the 6 samples A–F (see Table I).

Once established that the elementary building blocks are tetrahedra, the further step is to explore how these tetrahedra are arranged in space. Indeed, some local configurations are closer and others are looser and the whole packing is made by gluing together these tetrahedra in a disordered way which is compatible with the following three conditions: (1) mechanical stability, (2) geometrical constraints, and (3) space filling. Let me consider each of these conditions separately.

Mechanical stability is ensured by the network of contact between spheres. Indeed, in order to equilibrate forces and torques, a mechanically stable packing must satisfy topological conditions. In terms of Delaunay decomposition, it is empirically established [2,9,12] that such topological conditions constrain the average number of Delaunay neighbors to stay in a narrow range around $\langle f \rangle \simeq 14.5$. This is in agreement with what is observed in the

samples A–F where $\langle f \rangle$ varies within the two extremes 14.3 (sample F) and 14.6 (sample A).

The *geometrical constraints* are enforced by the condition of nonoverlapping. Equal spheres pack locally in the closest way when disposed all in touch with each other with centers on the vertices of a regular tetrahedron. Such a tetrahedron has volume $v^* = \sqrt{2}/12 d^3$. Therefore, the geometrical constraints fix a lower value for the volume attainable by a close packed Delaunay tetrahedron to v^* [Roger's bound [2]].

The constraint of *space filling* implies that the sum over all Delaunay volumes is equal to the total volume ($\sum_i v_i = V$).

If one considers the Delaunay decomposition as an ensemble of T independent cells with volumes v_i that freely exchange volume among each other under the three constraints described previously, then the partition function of such a system can be calculated exactly: $Z = (V - Tv^*)^T/T!$, and the probability to find a tetrahedron with a volume v reads

$$P(v) = \frac{1}{V/T - v^*}\left(1 - \frac{v - v^*}{V - Tv^*}\right)^{T-1}. \qquad (2)$$

In the (thermodynamic) limit $T \to \infty$, this expression simplifies to

$$P(v) = \frac{1}{\langle v \rangle - v^*} \exp\left(-\frac{v - v^*}{\langle v \rangle - v^*}\right), \qquad (3)$$

with $\langle v \rangle = V/T$ the average volume.

The empirical analysis of the six samples A–F confirm such theoretical prediction. In Fig. 2 it is shown that the inverse normalized cumulative distributions $P_>(v) = 1 - \int_0^v P(v)dv$ are well described, at large volumes, by the exponential behavior: $P_>(v) \propto \exp(-\beta v)$ [linear behavior in log-linear scale, Fig. 2(c)]. The best-fits values for the coefficients are: $\beta d^3 = 43.9$, 45.4, 55.2, 64.6, 66.8, 72.9 (samples A to F, respectively). Remarkably, the present theory is able to recover *quantitatively* such coefficients. Indeed, Eq. (3) predicts: $\beta^{-1} = \langle v \rangle - v^*$. The expected theoretical values for β can be obtained by imposing the three criteria on mechanical stability, geometrical constraints, and space filling. In particular, the geometrical constraint gives: $v^* = \sqrt{2}d^3/12$. The space-filling condition implies $\rho = \pi d^3 N/(6V)$, which, by using Eq. (1), gives: $\langle v \rangle = \pi d^3/[3(\langle f \rangle - 2)]\rho^{-1}$. By substituting these values, the coefficient β can be written as:

$$\beta^{-1} \simeq \frac{\pi d^3}{3(\langle f \rangle - 2)}\rho^{-1} - \frac{\sqrt{2}d^3}{12}, \qquad (4)$$

where the value of $\langle f \rangle$ can be conveniently fixed to 14.5 by observing that the criterium for mechanical stability requires $\langle f \rangle$ to stay in a narrow range around such a value. A comparison between this theoretical prediction and the empirical results is shown in Fig. 3. The agreement between the theory and the experimental data is remarkable. In this figure it is also shown that variations of $\langle f \rangle$ within the extreme values 14.3 to 14.6 experimentally observed

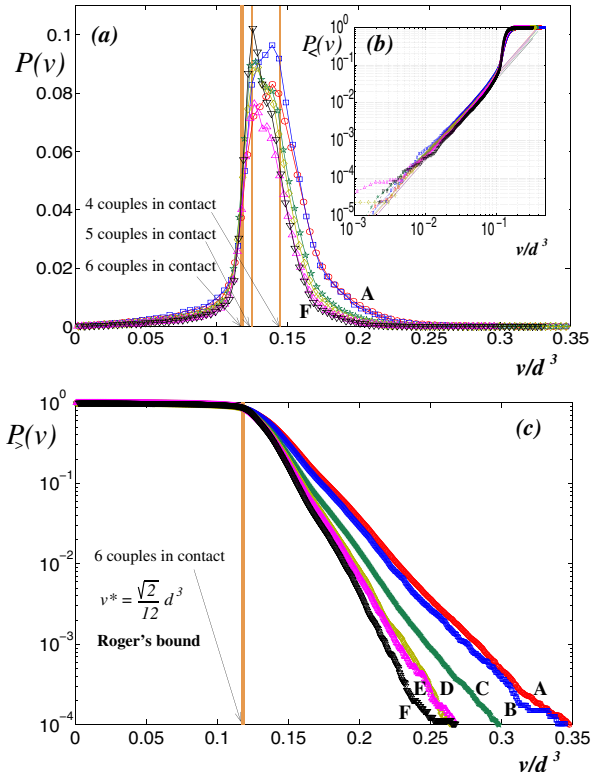

FIG. 2 (color online). (a) Normalized frequencies of the distribution of Delaunay volumes in G. The vertical lines indicate the maximum volumes attainable by tetrahedra with 4, 5, or 6 couples of spheres in contact (respectively: $\sqrt{3}d^3/12$, $d^3/8$, and $\sqrt{2}d^3/12$). (b) Log-log plots of the cumulative distributions $P_<(v)$ (the probability of finding a volume smaller than v). (c) Log-linear plots of the inverse normalized cumulative distribution $P_>(v)$ (the probability of finding a volume larger than v).

(Table I) do not affect significantly the results. Note that other choices for the elementary volumes, such as the Voronoï decomposition, do not yield to such neat exponential decay in the volume distributions [13]. An empirical study for the *void* volume distribution was proposed recently by Richard *et al.* [14]. Although they analyzed rather different systems (polydisperse packing of glass beads prepared at different densities by vertical shaking with different amplitudes), they also observe an exponential behavior for the probability distribution of the void volumes. The present theory can be similarly applied to the void-volume repartition and it predicts an exponential decay at large volumes with coefficient: $\beta^{-1} = \langle v_{\text{void}} \rangle - v^*_{\text{void}}$, with $\langle v_{\text{void}} \rangle$ the average void volume and v^*_{void} the minimum void volume. Remarkably, also in this case, this theoretical prediction results in good quantitative agreement with the empirical data reported in [14].

Probably the most striking property of granular materials is their eclectic behavior which is nether classifiable as a solid nor as a fluid [15]. I have shown previously that the structure of such systems can be conveniently described in terms of a packing of tetrahedra with exponential probability distribution at large volumes. From Fig. 2 one iden-

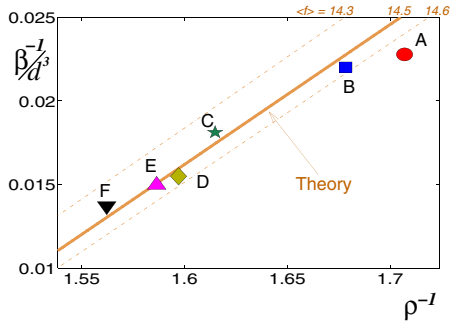

FIG. 3 (color online). Coefficients β^{-1}/d^3 vs the inverse packing fraction ρ^{-1}. The symbols correspond to the six samples A–F. The full line is the theoretical predictions from Eq. (4) with $\langle f \rangle = 14.5$. The two dot-dashed lines are the theoretical results for the two extreme cases $\langle f \rangle = 14.3$ and $\langle f \rangle = 14.6$.

tifies that the distribution is indeed exponential for volumes larger than $\sim 0.14d^3$. Meaningfully, the (small) volume region at which such distribution ceases to be exponential corresponds to Delaunay tetrahedra where most of the couples of spheres are in contact. In particular, when all the 6 couples spheres are in touch, the volume is $v^* \simeq 0.118d^3$, whereas when 5 couples are in touch, a tetrahedron can reach a maximum volume of $d^3/8 = 0.125d^3$; conversely, when only 4 spheres are in touch, the maximum reachable volume is $\sqrt{3}d^3/12 \simeq 0.144d^3$ [see Fig. 2(a)]. This fact indicates that below a given volume the tetrahedra are made of spheres in contact and geometrical constraints become unavoidable and relevant. One can therefore argue that these systems can be conveniently viewed as comprised of two phases: (1) a phase made by compact tetrahedra ($v < 0.144d^3$) which are geometrically constrained and are responsible for the mechanical stability and (2) a phase made by loose tetrahedra ($v > 0.144d^3$) which are geometrically unconstrained and take volumes accordingly with the distribution in Eq. (3).

Let me note that some tetrahedra can assume very small volumes (see Fig. 2). A Delaunay tetrahedron with zero volume corresponds to a configuration of 4 in-plane spheres. Therefore, configurations with volumes smaller than v^* are, in general, loose packings. Remarkably, Fig. 2(b) reveals that the probability distribution for such small volumes follows a power law behavior with typical exponents between 1.09 and 1.17. Such power laws might be related with the power laws observed in the distributions of the radial distance between couples of spheres [9].

In conclusion, by means of two independent methods (namely the analysis of the dihedral angles and the study of the volume distribution), I have shown that sphere packs can be conveniently studied as space-filling assemblies of elementary tetrahedra. I have demonstrated that the volumes of such tetrahedra follow an exponential distribution (at large volumes) which is controlled by the three conditions of mechanical stability, geometrical constraints, and space filling. It has been discussed that the system's state can be described in terms of the coefficient at the exponent β which is analogous to Edwards' compactivity $(\lambda X)^{-1}$ [16,17]. The theoretical predictions for β are in very good agreement with the empirical observations. Such an agreement is particularly remarkable considering that there are no adjustable parameters. The analysis of the probability distribution at small volumes reveals that, below $v \simeq \sqrt{3}/12d^3 \simeq 0.144d^3$, geometrical constraints, associated to the nonoverlapping condition, lead to a more complex distribution which is shaped by the accessible configurations in systems of touching spheres. Such differences in the kind of distributions at large and small volumes is a signature of structural heterogeneity. The granular system exploits such heterogeneity maximizing entropy and freedom while constraining mechanical stability.

Many thanks to T. J. Senden and M. Saadatfar for the tomographic data and several discussions. My thanks to P. Richard for the data of the pore distribution. This work was partially supported by the ARC discovery Project No. DP0450292.

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