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An invariant distribution in static granular media

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Abstract – We have discovered an invariant distribution for local packing configurations in static granular media. This distribution holds in experiments for packing fractions covering most of the range from random loose packed to random close packed, for bead packs prepared both in air and in water. Assuming only that there exist elementary cells in which the system volume is subdivided, we derive from statistical mechanics a distribution that is in accord with the observations. This universal distribution function for granular media is analogous to the Maxwell-Boltzmann distribution for molecular gasses.

Granular materials are complex systems characterized by unusual static and dynamic properties. These systems are comprised of large numbers of dissipative macroscopic particles assembled into disordered structures. There is a very large number of microscopic configurations corresponding to the same macroscopic properties. Edwards and coauthors [1,2] have proposed that the complexity of static granular systems could be disentangled by means of a statistical-mechanics approach reducing the description of the system state to a few parameters only [3–17]. An essential part of Edwards' idea is that in static granular media volume plays the role held by energy in usual thermodynamics. Therefore, an understanding of the volume distribution function is the key to connect micro scopic details of the system with macroscopic state variables.

Since granular materials are dissipative, they can change their static configurations only when energy is injected into the system. For instance, landmark experiments by the Chicago group [18–20] obtained different average packing fractions by tapping the container with different intensities and different numbers of times. Similarly, an experiment by Schröter *et al.* [21] obtained reproducible average packing fractions by driving the system with periodic trains of flow pulses in a fluidized bed. A general idea underlying a statistical-mechanics description is that the properties of the system do not depend on how the energy is injected but depend only on the portion of the configurational space that the system explores under such disturbances. This is not self-evident; *e.g.*, [22] showed that aging and irreversible behavior occur in tapped granular samples that have not reached a stationary state. The present paper demonstrates, using the largest sets of experimental data on particle positions presently available, that a statistical mechanics framework can predict local volume distributions. The system volume (V_T) is shown to be the relevant state variable, while the preparation history has no discernible influence on the questions studied here.

Experiments. – We analyze the structural properties of static granular packings produced in 18 different experiments, 6 with acrylic spheres in air and 12 with glass beads in water. The packing fractions (ρ) range from 0.56 to 0.64. Three-dimensional density maps have been obtained for these systems using X-ray computed tomography [23]. Coordinates of the bead centers have been calculated for more than two million spheres with a precision better than 1% of their diameters, which is better than the uncertainty arising from polydispersity (5% for the glass particles and 2% for the acrylic particles).

The 12 samples of glass beads were prepared using the fluidized bed technique described in [21]; each sample consisted of about 145000 spherical grains with

diameter $250 \pm 13\,\mu$m placed in a cylindrical glass container with inner diameter 12.7 mm. The beads were fluidized with pulses of deionized water produced by a computer-controlled syringe pump. During each flow pulse the bed expanded until its height reached a stable value. After each flow pulse the bed was allowed to settle into a mechanically stable configuration. Packing fractions in the range between 0.56 and 0.60 were obtained using pulses with different flow rates. The particle configurations have been studied for sub-sets of grains that were at a distance larger than four sphere diameters from the sample boundaries; these internal regions contained about 90000 grains per sample.

The 6 samples with acrylic spheres were described in [24]. Two of these samples each contained about 140000 particles (90000 in the internal volume) with diameter 1.59 ± 0.02 mm in a cylindrical container with an inner diameter of 55 mm, filled to a height of 75 mm. The other four samples each contained about 35000 acrylic spheres (15000 spheres in the internal volume) with diameter 1.00 ± 0.02 mm in the same cylindrical container. In these samples (labeled with symbols A-F) different packing fractions were achieved by different preparation methods: (A, $\rho = 0.586$) and (B, $\rho = 0.596$) were obtained by placing a stick in the middle of the container before pouring the beads into it and then slowly removing the stick; (C, $\rho = 0.619$) was obtained by gently and slowly pouring the spheres into the container; (D, $\rho = 0.626$) was achieved by a faster pouring; (E, $\rho = 0.630$) was realized by gently tapping the container walls; (F, $\rho = 0.640$) was reached by a combined action of gentle tapping and compression from above (leaving the upper surface unconfined at the end of the preparation).

The 12 samples of glass beads in water are examined for the first time in this paper, while the 6 samples of previously investigated acrylic beads [23–26] serve as reference data. The different types of beads, prepared by different methods, help differentiate between general properties of granular media and those properties specific to a given material or preparation procedure.

Theory. – Several statistical mechanics approaches have been proposed to describe granular materials [1,2, 4–6,9,11,14,27]. We make here the minimal number of assumptions in considering a granular system at rest that occupies a given volume V_T. We consider that the system is subdivided into C elementary cells which can have any volume v_i $(i = 1 \dots C)$ larger than or equal to a minimum volume v_{min}, under the condition $\sum_{i=1}^{C} v_i = V_T$. Under the sole assumption that any assembly of such elementary volumes will produce proper mechanically stable packings, we can obtain the probability distribution for the cell-elementary volume ($p(v)$) by computing the ratio between the number of configurations containing a cell of volume v ($Z(v) = (V_T - Cv_{min} - v + v_{min})^{C-1}/(C-1)!$) and the total number of configurations $Z = (V_T - Cv_{min})^C/C!$). This yields $p(v) = Z(V)/Z = \frac{C}{V_T - Cv_{min}}(1 - \frac{v - v_{min}}{V_T - Cv_{min}})$, which in the limit $C \to \infty$ and V_T/C finite, becomes

$$p(v) = \frac{1}{\chi} e^{-(v - v_{min})/\chi}, \tag{1}$$

with

$$\chi = \langle v \rangle - v_{min}, \tag{2}$$

where $\langle v \rangle = V_T/C$ is the average volume per elementary cell and χ is an intensive thermodynamic parameter accounting for the exchange of volume between the elementary cell and the surrounding volume "reservoir". There is no need to specify the nature and kind of such "elementary cells"; the only assumptions are that they exist, that they can have any volume larger than v_{min}, and that their assembly fills a volume V_T. Space can be divided arbitrarily into pieces. Common examples of such partitions into space-filling blocks are the Delaunay and the Voronoï decompositions [26,28,29]. Such cells do not coincide with the elementary ones, but they might be assemblies of such elementary cells. For instance, in a Delaunay partition of a three-dimensional packing there are about six times more cells than in the Voronoï decomposition. The present theory applies to any degree of space partition made by any agglomerate local structure made of a given number k of elementary cells, since the probability distribution function for each elementary cell follows an exponential distribution (eq. (1)). The aggregate probability distribution function $f(V, k)$ of a sum ($V = \sum_{i=1}^{k} v_i$) of k volumes must be a Gamma distribution [30],

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

with $\langle V \rangle = kV_T/C$ and $V_{min} = kv_{min}$ being, respectively, the average and the minimum volumes for a given packing. It follows from eqs. (1) and (3) that

$$\chi = \frac{\langle V \rangle - V_{min}}{k}, \tag{4}$$

which is the average free-volume per elementary cell. Therefore, the intensive variable χ is a measure of the kind and the degree of space-partition into elementary cells. The distribution $f(V, k)$ is a Gamma distribution in the variable $V - V_{min}$; it is characterized by a "shape" parameter k and a "scale" parameter χ [30]. Gamma distributions have been observed in two-dimensional Voronoï networks [31], and empirical fits with a Gamma distribution were proposed for Voronoï partitions from random Poisson points in three dimensions [32].

We have obtained $f(V, k)$ assuming that the cells are uniquely characterized by their volumes, and that any combination of C cells with arbitrary volumes $v_i \geqslant v_{min}$ will produce a structurally stable, space-filling system of cells. This is possible in one dimension only where the packing is an arbitrary arrangement of grain centers at distances larger or equal than v_{min}. In this case the elementary cells are the Delaunay cells [29], which are

the segments in between two successive grain centers; the distribution of their sizes is exactly described by $p(v)$. In three dimensions, the Delaunay cells are tetrahedra with vertices on the centers of neighborhood grains chosen in a way that no other grains in the packing have centers within the circumsphere of each Delaunay tetrahedron. Clearly, in this case the Delaunay cells are not uniquely described by their volumes, and an arbitrary collection of cells is neither space-filling nor mechanically stable.

Results. – The distributions of the Delaunay volumes obtained from the 18 sets of data for different kinds of beads in different media and different conditions are all described well by the *same* function, as shown fig. 1(a). The collapse of the data onto a single curve was obtained using $V_{min} = \sqrt{2}/12d^3$ (with d the sphere diameter), which is the volume of a regular Delaunay tetrahedron for four spheres in contact (the smallest compact tetrahedron [26]). In a recent paper, Aste [26] showed that the tails of the distribution of the Delaunay volumes in the 6 experiments with dry acrylic beads (A-F) were exponential with coefficients associated with the packing fraction; this is confirmed by the present investigation, which shows that the same kind of exponential decay applies both to dry acrylic bead packs prepared with different methodologies (A-F) and to glass beads in water prepared with fluid pulses. All the data fit well to the inverse cumulate distribution, $F(V) = 1 - \int_{v_{min}}^{V} f(v', 1)\mathrm{d}v'$, as fig. 1(b) shows; the agreement extends over four orders of magnitude and uses no adjustable parameters. The observation of the same behavior for all 18 samples demonstrates that such a result is robust and likely to be universal for a broad class of granular systems. Further, fig. 1(a) shows that the Delaunay volume distributions for all the samples collapse on the same master curve when plotted *vs.* $(V - V_{min})/(\langle V\rangle - V_{min})$. This new finding reinforces the underlying idea that volume fluctuations in granular packs are described by a universal probability distribution with parameters depending only on the packing fraction. The agreement between the experiments and the predictions in eq. (3) for $k = 1$ suggests that Delaunay cells might be considered candidates for the "elementary cells"; however, it was noted in [26] that, at small $V - V_{min}$, in the region where the spheres make contact ($V < \sqrt{3}d^3/12$), the empirical distributions deviate from the simple exponential form predicted by $p(v)$, which indicates that other constraints such as mechanical equilibrium should be also taken into account to correctly describe such a region.

An alternative way for dividing space into space-filling cells is the Voronoï partition. Let us first consider the one-dimensional case where the present theory is exact. In one dimension, the Voronoï cell around grain 'i' is constructed by taking the segment between the two mid-points between grains $i-1$ and i, and between grains i and $i+1$. The size of such a segment is equal to $(v_i + v_{i+1})/2$ where v_i and v_{i+1} are the distances

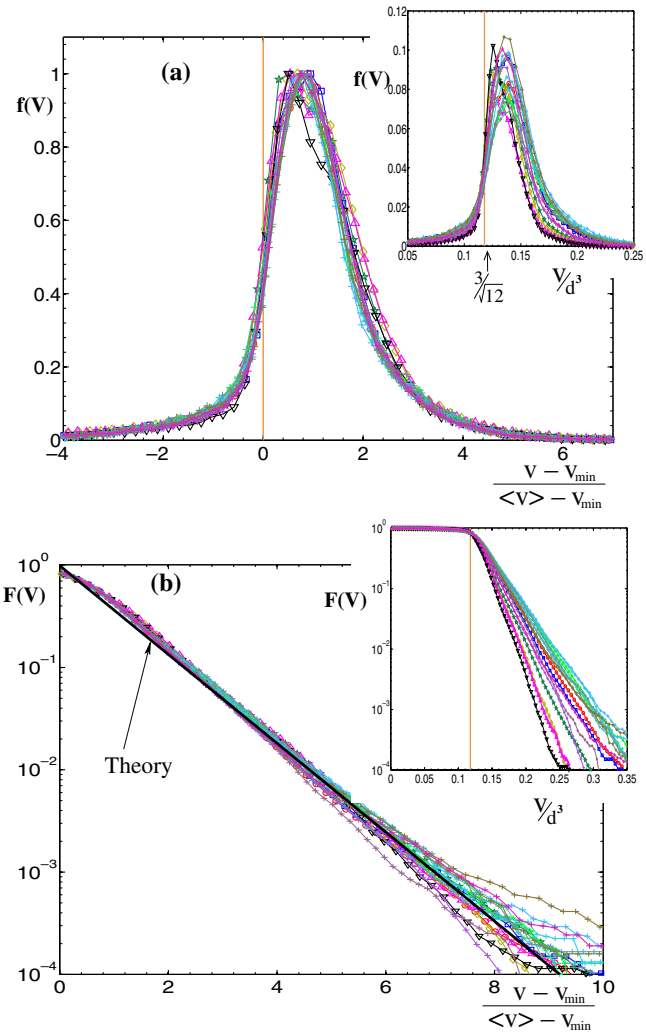

Fig. 1: (a) Distributions of the Delaunay cell volumes from 18 experiments collapse onto a universal curve when plotted *vs.* $(V - V_{min})/(\langle V\rangle - V_{min})$. The inset shows that the same distributions plotted *vs.* V/d^3 do not collapse. (b) The inverse cumulate distributions, $F(V) = 1 - \int_{V_{min}}^{V} f(V)\mathrm{d}V$, also show evidence of a collapse onto a universal curve when plotted *vs.* $(V - V_{min})/(\langle V\rangle - V_{min})$. The line is the inverse cumulate distribution for $f(V, k = 1)$ (eq. (3)). The inset shows the cumulate distributions plotted *vs.* V/d^3. The distributions are obtained from a statistical analysis of more than six million cells in 18 different experiments with three different kinds of beads in air and in water. The open symbols correspond to the 6 experiments with dry acrylic beads [24]: (A) circles; (B) squares; (C) stars; (D) diamonds; (E) triangles; (F) triangles down. The "+" corresponds to the 12 experiments with packing fractions $0.56 \leqslant \rho \leqslant 0.60$ made with glass spheres in fluidized bed.

between the two consecutive couples of points. Therefore, the probability of finding a one-dimensional Voronoï cell of size V is associated with the probability of finding two successive Delaunay cells with sizes $v_1 + v_2 = 2V$. This probability is $2\int_{v_{min}}^{2V - v_{min}} p(v_1)p(2V - v_1)\mathrm{d}v_1 \propto (V - v_{min})\exp[(-2V + 2v_{min})/(\langle V\rangle - v_{min})] \propto f(V, k = 2)$,

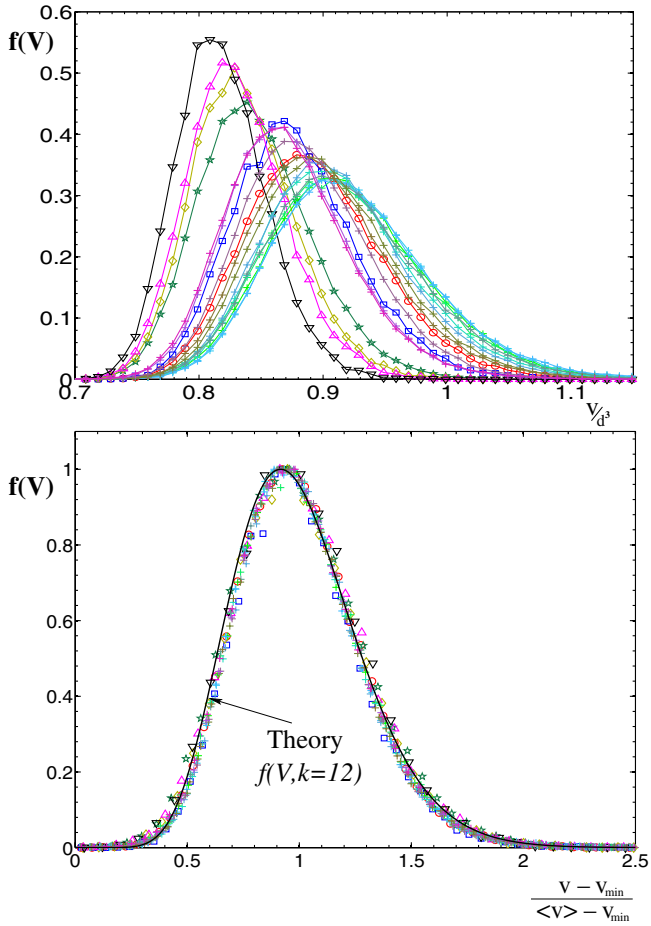

Fig. 2: Top: distributions of the Voronoï cell volumes plotted *vs.* V/d^3. The data refer to the 18 experiments, and the symbols are the same as in fig. 1. Bottom: all the distributions collapse onto a universal curve when plotted *vs.* $(V - V_{min})/(\langle V\rangle - V_{min})$. The theoretical line is $f(V, k = 12)$ (eq. (3)).

with $\langle V\rangle$ the average size of the Voronoï cell. In three dimensions, the Voronoï cell can be also seen as the combination of several elementary cells with distribution $f(V, k)$; however, in this case the number of sub-cells involved is not fixed at $k = 2$ but depends on the kind of packing.

In [26] it was noted that the Voronoï volume distributions for the 6 samples A-F do not exhibit an exponential decay. Indeed, in this paper we have demonstrated that such distribution must follow eq. (3), which is not a simple exponential. In fig. 2 it is shown that data for over a million of Voronoï cells from all 18 experiments for dry and wet packings of glass and acrylic spheres collapse to the the same distribution function. In this collapse of the data there are no adjustable parameters, just $\langle V\rangle = V_T/(number\ of\ grains)$ and $V_{min} = 5^{(5/4)}/\sqrt{2(29 + 13\sqrt{5})}d^3 \simeq 0.694d^3$, which is the smallest Voronoï cell that can be built in a equal-spheres packing [29]. Figure 2 shows that such a universal distribution function is well described by eq. (3) with

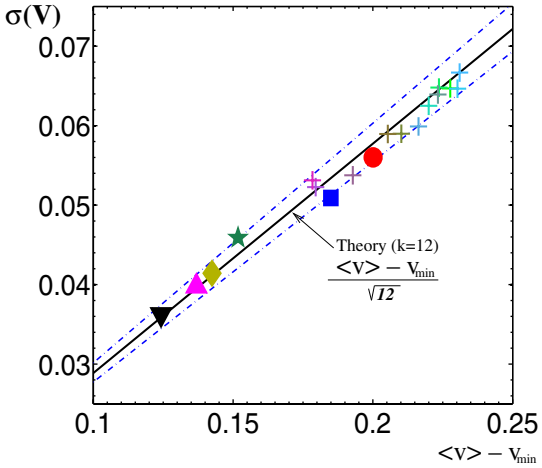

Fig. 3: The standard deviations of the Voronoï volume distributions in the 18 experiments fit the linear relation given by eq. (6) with $k = 12$. The dot-dashed line above and below the solid line corresponds to $k = 11$ and $k = 13$, respectively.

$k = 12$, which indicates that about 12 elementary cells contribute in building each Voronoï cell. Such a number is meaningful, since about 12 spheres are expected to be found in the close neighborhood of any given sphere in the packing. We find also that the distribution in eq. (3) with $k = 12$ holds for the Voronoï volumes from the simulations of granular packings reported in [27]. An equivalent collapse of the distributions can be obtained by plotting the volume distributions *vs.* $(v - \langle v\rangle)/\sigma = \sqrt{k}((v - v_{min})/(\langle v\rangle - v_{min}) - 1)$, as proposed by Starr *et al.* [33] for simulations of a polymer melt, water, and silica, but in this case the parameter k and the minimum volume v_{min} must be changed.

A further demonstration that such statistical distributions are independent of the details of a sample and the method of sample preparation is provided by the behavior of the volume fluctuations, which can be calculated directly from the relation (see eq. (3)),

$$\sigma^2(V) = \left\langle (V - \langle V\rangle)^2 \right\rangle = \chi^2 \frac{\partial \langle V\rangle}{\partial \chi}, \tag{5}$$

which becomes, using $\partial \langle V\rangle / \partial \chi = k$ (from eq. (4)),

$$\sigma(V) = \chi\sqrt{k} = \frac{\langle V\rangle - V_{min}}{\sqrt{k}}. \tag{6}$$

The good correspondence of the data from the Voronoï volume distributions with eq. (6) (fig. 3) provides further evidence that $\langle V\rangle$ and k are the relevant control parameters.

Conclusions. – We have shown that the local volume distributions of granular packings of monodisperse spherical grains are described by a universal distribution function (eq. (3)). This distribution function was derived using a statistical-mechanics approach and the assumption that the volumes are composed of a set of elementary

cells. Granular samples have been prepared by water fluidization pulses, tapping, and pouring. Fluidization pulses have been shown to produce stationary, history-independent states for the range of volume fractions studied here [21], and tapping fulfills those criteria only in a range different than that of our samples [22]. (For pouring the independence of preparation history has not been established.) Therefore, the agreement between theory and experiment that we have found indicates that the local volume distribution is not sensitive to different ways of sampling the granular phase space. This suggests some generic ergodicity, which gives hope for a statistical mechanics of static granular media.

In the present theory the only tunable parameter is k, which has been found to be $k \sim 12$ for Voronoï decompositions throughout the accessible density ranges of the different static granular packings studied. Interestingly, for granular gasses the same empirical distribution (eq. (3)) applies but with $k = 5.586$ [32,34]. A similar kind of distribution (Gamma distribution) with different values of the parameter k has also been observed in a two-dimensional Voronoï tessellation generated from disk packings [35]. Therefore, the parameter k could be the 'structure parameter' which depends on the system phase. From fig. 3 one can see that the deduced k values (eq. (6)) lie in a narrow range between $k \simeq 11$ and $k \simeq 13$. Further investigations are needed to understand if such a range of values is associated with statistical uncertainty or is associated with changes occurring in the system structure. Since the intensive quantity χ is inversely proportional to k (eq. (4)), the parameter k (together with the packing fraction) is a control parameter for these systems. Within the framework of a statistical mechanics description, eq. (3) can be regarded as analogous to the Maxwell-Boltzmann distribution for granular media.

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