Structural characterization and statistical properties of two-dimensional granular systems

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A recently developed method is used for the analysis of structures of planar disordered granular assemblies. Within this method, the assemblies are partitioned into volume elements associated either with grains or with more basic elements called quadrons. Our first aim is to compare the relative usefulness of description by quadrons or by grains for entropic characterization. The second aim is to use the method to gain better understanding of the different roles of friction and grain shape and size distributions in determining the disordered structure. Our third aim is to quantify the statistics of basic volumes used for the entropic analysis. We report the following results. (1) Quadrons are more useful than grains as basic ''quasiparticles'' for the entropic formalism. (2) Both grain and quadron volume distributions show nontrivial peaks and shoulders. These can be understood only in the context of the quadrons in terms of particular conditional distributions. (3) Increasing friction increases the mean cell size, as expected, but does not affect the conditional distributions, which is explained on a fundamental level. We conclude that grain size and shape distributions determine the conditional distributions, while their relative weights are dominated by friction and by the pack formation process. This separates sharply the different roles that friction and grain shape distributions play. (4) The analysis of the quadron volumes shows that Γ distributions, which are accepted to describe foamlike structures well, are too simplistic for general granular systems. (5) A range of quantitative results is obtained for the ''density of states'' of quadron and grain volumes and calculations of expectation values of structural properties are demonstrated. The structural characteristics of granular systems are compared with numerically generated foamlike Dirichlet-Voronoi constructions.

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I. INTRODUCTION

There has been a growing interest in the application of statistical mechanics methods to granular systems. Unlike conventional thermodynamic systems, loosely compressed granular materials are not in equilibrium unless sufficient initial agitation is applied to the system [1]. Once the agitation stops the system consolidates into a specific state, which can be described by statistical mechanics [2]. In generally porous materials the situation is much less clear since there is no guarantee that the history of the consolidation of the material does not play a role. If it does then equilibrium-based models will not work. However, the entire science of porous media is based on the premise that the statistics of a given block of a particular rock has on average the same structural characteristics as another block of the same material and of the same volume, taken from the same area. Otherwise, it would be pointless to try to derive typical macroscopic properties, such as permeability. This underlying premise allows us to use statistical mechanical tools to describe porous media of similar statistics. It is important, however, to substantiate that this approach is as useful to such systems as it is for shaken cohesionless granular media.

The statistical mechanical formalism of Edwards is based on replacing the energy of thermodynamic systems by volume and the temperature by an analogous quantity—compactivity [2,3]. The partition function of the formalism involves only an entropic contribution, i.e., it is the logarithm of the number of configurations in which the granular material can be arranged.

Using the parallels with conventional statistical mechanics, the partition function is

$$Z = \int e^{-W(\{q\})/X} \Theta(\{q\}) \Pi \, dq, \tag{1}$$

where X is the compactivity and W is a function that gives the volume of the entire system in terms of all the independent variables (q) in the system, such as the sizes of the grains, their orientations, etc. These variables are called the degrees of freedom because their values in any specific realization determine the statistics of the structure. The entropy is defined as $S=\log[\int \delta(V-W)\Theta \, d\{\vec{q}\}]$, where V is the volume of the system, Θ the condition that the configuration is connected and in mechanical equilibrium, and $\vec{q}$ a vector of all the degrees of freedom that the structure depends on. In terms of the entropy, the compactivity is defined as $X = \partial V / \partial S$.

For convenience, (1) is written in terms of a canonical ensemble, namely, the total number of quadrons (and hence the overall number of intergranular contacts), as well as the number of particles, is constrained to be constant in the ensemble. Alternatively, if only the number of particles is constrained, then a grand canonical ensemble must be used since the number of quasiparticles—the quadrons—changes from realization to realization. An interesting question is whether the canonical ensemble can be used to characterize results of

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experiments; that is, to ask whether there are experiments for which the variation of the mean coordination number per realization of the granular system is much smaller than its average over all realizations of the experiment. While there are experiments that fix the packing fraction of grains [1], it is not clear whether there is a close relation between the density of grains and the coordination number for these experiments. At least two relations are needed to clarify this issue: between the means and the standard deviations of the density and the mean coordination number. We are not aware of derivation of such relations. Nevertheless, since the aim of this paper is not an explicit statistical calculation but only an illustration of one with the data that we have, then we consider here only the canonical partition function and leave the grand canonical for future work.

This entropic approach makes it possible to calculate structural properties as expectation values. For example, in porous materials made of consolidated grains, the volume associated with one grain is

$$\langle V \rangle = \frac{1}{Z} \int V_g(\{q\}) e^{-W(\{q\})/X} \Theta(\{q\}) \Pi \, dq, \tag{2}$$

where the angular brackets denotes an average over an ensemble of all possible systems.

The initial doubts about the applicability of equilibrium formalism to these far-from-equilibrium systems has been put to rest by experimental evidence that there is a reproducible distribution of density states [1]. Even so, this approach was slow to catch on because the geometric correlations of the connected structure made it a major challenge to identify the relevant degrees of freedom and therefore the phase space. These problems have been resolved recently, as will be discussed in more detail below, paving the way to a range of systematic calculations both in two and in three dimensions [3,4].

In this paper, we demonstrate the usefulness of the formalism to the study of a range of simulated granular aggregates. In particular, we address two issues. One is a systematic characterization of the structure of granular materials and the other is an attempt to provide a fundamental, rather than phenomenological, understanding of the exact roles that grain size distributions and friction play in the determination of the structure. Starting with the characterization of the granular medium through connectivity and contact networks, space is tessellated and we identify basic volume elements called quadrons [3]. These are used to construct the volume function W in (1) and to investigate the statistics of simulated systems. The quadrons are also used to demonstrate computations of expectation values. The formalism is further used to investigate the structure of the granular systems and to compare the usefulness of either quadron or grain volumes as the basic volume elements.

The main results reported in this paper are the following. (i) The probability density functions (PDFs) of both quadron and grain volumes are computed. These PDFs are essential to the construction of the Edwards entropic formalism. (ii) We find that these PDFs exhibit nontrivial structure. These cannot be understood by the simple fits practiced in the literature, which miss essential characteristics of dense granular packings. (iii) It is shown that the features of the PDF of quadron volumes stem from the distribution of the number of cell edges, which we call the cell coordination number. (iv) The quadron volume PDFs are decomposed into a superposition of conditional PDFs. We show that the roles of friction and polydispersity can be understood in the context of these conditional PDFs. In particular, we demonstrate the clearly separate roles that grain size distribution and friction have on the structure. We show that this understanding can come only from analysis of the quadron volume PDF; the PDF of the grain volumes mixes the conditional distributions and washes out the above effects. (v) We find distinct differences between grain and quadron volume PDFs. Fundamental arguments are presented that establish the latter as the more useful for entropic characterization.

This paper is constructed as follows. In Sec. II we give a brief review of the construction of the volume function. In Sec. III we describe briefly the simulated systems and the way that we extract the statistics of the quantities that we wish to analyze. In Sec. IV we present the results of the various analyses. We conclude in Sec. V with a discussion and interpretation of the results.

II. CONSTRUCTION OF THE VOLUME FUNCTION

The construction described here has been developed for granular media [3]. The original construction, as indeed the entropic formulation, has been mainly limited to marginally rigid structures [5], but here we extend the description for general structures. The idea is to start from the contact network and construct a set of basic volume elements that cover the system and can be defined in terms of a compact set of variables, called degrees of freedom. To conform with the literature, we use in the following the term "volume" to describe the area of the two-dimensional systems. This is done in several steps, illustrated in Fig. 1.

(1) Starting from a collection of grains in mechanical equilibrium, all the inter-granular contact points are identified.

(2) Around every grain, say g, the contact points are connected by vectors that circulate the grain in the clockwise direction, Fig. 1. The vectors form polygons around the grains as well as around the voids that the grains enclose, circulating in the counterclockwise direction. The polygons of the voids are denoted henceforth as the cells. Note that grains with two contacts produce degenerate polygons of two edges. Grains with fewer than two contact points are mechanically unstable and thus ignored. We also ignore rattlers [6], which are grains resting only on grains underneath.

(3) Every cell polygon is given an index $c = 1, 2, \ldots, N_{\text{cell}}$ where N_{cell} is the total number of cells. Note that every vector can be associated with exactly one grain and one cell and therefore it can be indexed uniquely, $\vec{r}_{gc}$. The r network is the set of all the vectors $\vec{r}_{gc}$.

(4) Centroids are defined for every polygon in the system, around both grains and cells. A centroid is the mean position vector of the corners of the polygon (the contact points of the original grains). A vector, denoted as $\vec{R}_{gc}$, is extended from

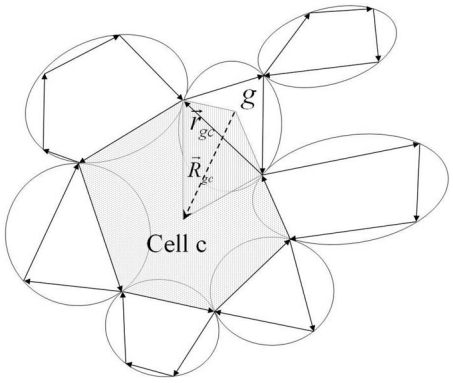

FIG. 1. Tessellation of space in general 2D granular packs. Intergranular contact points are identified. Grains are represented by polygons, generated by connecting contact points with vectors that circulate around the grains in the clockwise direction. Consequently, the vectors circulate around the cells in the counterclockwise direction. This network we call the r network. An R vector network is generated (dashed arrows) which connects the centroids of the grains to the centroids of their neighboring cells. Quadrons are the quadrilaterals whose diagonals are the r-R pairs that correspond to the same grain and cell, e.g., r_{gc}-R_{gc}.

every grain centroid to the centroids of the cells that surround it, e.g., Fig. 1. The network of R vectors is dual to the r network—every $\vec{r}_{gc}$ in the latter corresponds to one vector $\vec{R}_{gc}$ in the former.

(5) Quadrilaterals, named quadrons q_{gc} [3], are constructed, whose diagonals are $\vec{r}_{gc}$ and $\vec{R}_{gc}$, e.g., Fig. 1.

The result of this construction is a perfect tiling of the system—the quadrons cover the entire area seamlessly. This provides a volume function,

$$W = \sum_{g,c} V_{gc} \equiv \sum_{q} V_q,$$

where $V_q \equiv V_{gc} = 1/2 |\vec{r}_{gc} \times \vec{R}_{gc}|$ is the volume of one quadron.

It has been shown in [3] that this volume function makes it possible to (i) identify a compact phase space of degrees of freedom; (ii) take into consideration the geometric correlations explicitly; (iii) calculate explicitly relevant structural expectation values. It is interesting to note that the most basic volume units of the structure are the quadrons, not the grains.

In a system of N grains of mean contact number $\bar{z}_g$, the volume function appears to depend on all the $2\bar{z}_g N$ vectors $\vec{r}_{gc}$ and $\vec{R}_{gc}$, but these are not all independent. First, all the R vectors can be expressed as linear combinations of the r vectors. Second, within the r network, every basic polygon of vectors (irreducible loop) gives one dependent vector. To obtain the number of independent vectors, one therefore has to count these loops. There are N grain polygons and M cell polygons. To find M, we use Euler's expression relating vertices, faces, and edges in a two-dimensional (2D) network. In terms of our variables, this relation reads

$$\frac{\bar{z}_g N}{2} - \bar{z}_g N + (M + N) = 1, \tag{4}$$

where N is the number of grains and M is the number of cells. This gives

$$M = \left(\frac{\bar{z}_g}{2} - 1 \right) N + 1 \tag{5}$$

and, subtracting all the dependent vectors, leaves only $\bar{z}_g N/2$ independent vectors. Since every vector has two components then the phase space consists in total of $\bar{z}_g N$ degrees of freedom. It is important to note that the number of independent variables equals exactly the number of quadrons, regardless of system size. Thus, quadrons are the analogs of quasiparticles in conventional statistical mechanics, a point that we will return to later. The above counting neglects boundary corrections, which are small for large systems.

III. SYSTEMS

For our analysis we chose a number of systems. One family of systems consists of granular packs, constructed numerically by depositing disks (or dispherical grains) onto a rectangular container of width L. In these systems, the disk radii were chosen either from a monodisperse or from a uniform probability density. In all samples the average diameter was chosen to be less than $L/30$. The discrete element method [7] was used to simulate particle deposition. Groups of 100 randomly placed disks were generated above the packing, and the grains of each group were allowed to settle down before another group of disks was added. In order to save computational time, the positions of disks from all but the three last added groups were fixed. Because these "frozen" disks are several disk diameters below the actual top of the packing, we assume that their position will not be affected by newly deposited disks. The algorithm for deposition of dispherical grains is analogous and is described in detail in [8]. In the simulations, we have varied the intergranular friction coefficients within the range [0,57]. The finite friction coefficient limits the amount of crystallization in monodisperse systems.

For each choice of friction and size distribution, we have varied the total number of disks, to check the dependence of the resultant distributions on system size. We have found that the statistics of all the quantities that we have measured stabilized when the number of disks exceeded 3000. In particular, we have established that 40 8000-disk simulations with the same parameters, but starting from different initial conditions, gave rise to practically the same distribution of quadron volumes. When the number of disks was reduced below 3000 per sample, noticeable differences started to appear between samples. We therefore set 4000 as the minimum number of disks per sample.

Altogether, we have analyzed four families of systems.

(1) Monodisperse disks of four different friction coefficients $\mu = 0$, 0.36, 0.84, and 57. These values correspond to angles of friction of 0°, 20°, 40°, and 89°.

(2) Polydisperse disks, whose radii were distributed uniformly between $r_{\min}$ and $3r_{\min}$, and whose friction coefficients were the same as above.

(3) Dispherical grains, with friction coefficient $\mu=0.47$ and 57, corresponding to angles of friction of 25° and 89°. A grain was made of two rigidly attached disks of equal radii R. The centers of the disks were separated by variable distances, ranging within $(0.42 \pm 0.25)R$.

(4) Trivalent foamlike structures generated by a Dirichlet-Voronoi tessellation as follows. First, Poisson seeding points were distributed within a rectangular container of width L. Once all the points have been distributed, a Voronoi diagram was generated, where a unique cell was constructed around each point. The vertices generated by the edges of the Voronoi cells are all trivalent, namely, they connect exactly three edges. The resulting cellular structure was mapped onto a granular system using the procedure described in [9]. Briefly, the mapping generates pseudograins by connecting the middle of the edges connecting to each of the foam's vertices. This allows us to compare directly the statistical properties of the structures in our granular systems and the Dirichlet-Voronoi structures—a class of structures for which there is an extensive literature and which are therefore very useful for comparison.

IV. ANALYSIS

The analysis of the above structures was carried out, following the procedure outlined in Sec. II. Once the contact network had been generated, we studied the statistics of quantities that relate to two types of data: the connectivity of the structure and the partition of the volume into quadrons or grains. Below, we report our results, and we discuss them and their interpretations in the concluding section.

A. Connectivity

The connectivity of a granular pack is arguably the one most significant factor in determining macroscopic structure-dependent properties. In the first instance we studied the distributions of the number of grain contacts and the number of grains around cells. These quantities are strongly sensitive to the consolidation dynamics and therefore are key to the understanding of the effects of the formation of the structure on macroscopic properties. In Fig. 2(a) we show the PDF of the number of grain contacts, z_g, for all the systems described above. In Fig. 2(b) we plot the PDF of the number of grains around a cell, z_c, for these systems.

The figures show that, as a general rule, the wider the distribution of the contact numbers, the narrower the distribution of the number of grains around cells, and vice versa. This could be best seen in the polydispersed systems. For the polydispersed disk samples with $\mu=0,0.36,0.84,57$, the standard deviation of grain coordination number is $\sigma_g=0.979,0.933,0.926,0.913$ and the standard deviation of cell coordination number is $\sigma_c=0.54,0.79,1.02,1.21$, respectively. For monodispersed packing of disks, crystallization effects change the above conclusion for very low friction coefficients. This is in accord with the idea that the number

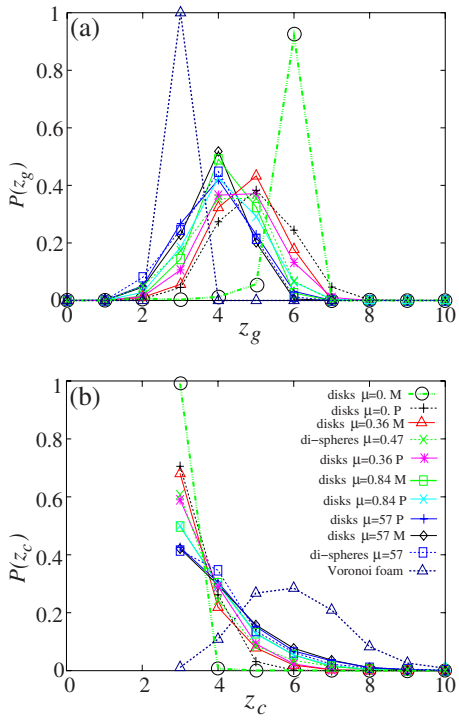

FIG. 2. (Color online) Probability density functions of the grain coordination number (a) and cell coordination number (b) for all the systems studied in this paper. Note that $\bar{z}_g$ decreases with increasing friction, while $\bar{z}_c$ increases, in agreement with Eq. (7). The most probable value of $\bar{z}_c$ is 3 in all the granular systems. This is in contrast with the Voronoi cellular system, where the most probable value of $\bar{z}_c$ is 6.

of grains around a cell is the contact number of the reciprocal (or dual) network. An extreme manifestation of this relation can be observed for trivalent foams, where $z_g=3$ for all grains. The zero width of this PDF corresponds to the broadest PDF of z_c.

The mean number of quadrons per cell, which is the same as the number of grains per cell, can be readily found from relation (5). In the dual network the values of M and N are interchanged and therefore from their ratio we find that

$$\frac{\bar{z}_g}{2} - 1 = \left(\frac{\bar{z}_c}{2} - 1\right)^{-1}, \tag{6}$$

which gives

$$\bar{z}_c = \frac{2\bar{z}_g}{\bar{z}_g - 2}. \tag{7}$$

The overbars in Eqs. (6) and (7) denote averages over the volume of the system. In granular systems, the smallest coordination number of grains is constrained by the fact that the system needs to be mechanically stable. This is the isostatic limit for which all forces can be deduced from the force and torque balance equations. In the isostatic limit $\bar{z}_g=3$ for infinitely rough grains, $\bar{z}_g=6$ for smooth noncircular grains, and $\bar{z}_g=4$ for smooth circular grains [10]. The corresponding mean numbers of quadrons per cell are 6, 3, and 4. Figure 2

is in good agreement with these considerations.

The large-value tail of the PDF of z_g is sensitive to the grain size distribution. In the monodisperse samples, the maximal value of z_g can be 6 for obvious reasons. This number increases as the ratio between the largest and smallest radii gets further from unity, and it can be readily shown that

$$(z_g)_{\max} = \alpha\left(\frac{r_{\max}}{r_{\min}} + 1\right), \tag{8}$$

where $3 \leq \alpha \leq \pi$, depends weakly on $r_{\max}/r_{\min}$. Note that very broad size distributions give rise to extremely large values of z_g. In our polydisperse samples, the distribution of radii is between $r_{\min}$ and $3r_{\min}$, giving $(z_g)_{\max} \approx 12$.

Because of the duality relation between the grain and cell structures, similar considerations apply to the PDF of z_c but with some differences. One is that expression (8) does not apply and the corresponding relation for cells is much more difficult to formulate. Another is that the smallest value of z_c cannot go below 3. This is in contrast to grains, which can be in equilibrium in contact with as few as two neighbors.

B. Volume partition

The partition of the volume is essential to the characterization of porous structures. In particular, the partition into elemental volumes is a key to the entropy-based statistical formalism described above. As can be seen from expression (1), the elemental units in the volume function can be either the quadrons or the grains. For completeness and to assess the advantages and disadvantages of either description, we have analyzed the volume distributions of both. First, we calculated the volume of every quadron, defined above. Then we obtained the volume associated with every grain by summing the volumes of the quadrons around it, $V_g = \sum_{c=1}^{z_g} V_{gc}$. In Fig. 3 we plot both the quadron and grain PDFs for foam and for the polydispersed disk systems.

In the samples of Figs. 3(a)–3(d), the granular systems are identically polydisperse with a uniform disk size distribution, but with different friction coefficients. Commonly, partitions of space by traditional tessellations, such as the Dirichlet-Voronoi tessellation, have been shown to give rise to Γ distributions of cell volumes [11–14]. Therefore, we have tried to fit Γ distributions to the quadron volume PDF,

$$P(V_q) = \frac{b^a}{\Gamma(a)} V_q^{a-1} \exp(-bV_q), \tag{9}$$

under the assumption that V_q can be extrapolated to $V_q = 0$. For this fit we approximated the upper bound on V_q as infinite, $V_q \to \infty$. To compare the quality of the fit, Fig. 3(e) shows the PDF of a Dirichlet-Voronoi sample that was produced using the aforementioned Poisson seeding process. The PDF is fitted excellently with the Γ distribution, in agreement with previous works [15,16].

The PDFs of the grain-associated volumes are shown on the right-hand side of Figs. 3(a)–3(e). The differences between the grain and quadron PDFs are clearly visible in all these systems. To quantify these differences, we tried to fit the grain PDF with the Γ distributions as well. The fits of the PDFs of V_g are consistently poorer than the fits of V_q: the uncertainties in the parameters a and b for the former are two to three times larger than for the latter and the tails of the grain distributions are especially poorly fitted. In addition, the value of the parameter a is markedly different between the fits to the grain and quadron PDFs. The importance of this observation is discussed in the next section.

Expectation values of desired properties can be calculated analytically, from the partition function Eq. (1), once a density of states is given. Assuming that the quadrons are not correlated, the density of states is a product of the single-quadron PDFs $P(\{V_{q_i}\}) = \Pi_i P(V_{q_i})$, and the partition function can be rewritten as

$$Z = Z_q^{N_q} = \left(\int e^{-V_q/X} P(V_q) dV_q\right)^{N_q}, \tag{10}$$

where N_q is the total number of quadrons in the system. Using a Γ distribution for V_q as an illustration, the expectation value of the nth moment of the quadron volume is

$$\langle V_q^n \rangle = \frac{\int e^{-V_q/X} V_q^n P(V_q) dV_q}{Z_q} = \frac{\Gamma(n+a)}{\Gamma(a)(b+1/X)^n}. \tag{11}$$

It is interesting to note that the above functional form of (11) has been used in the literature for the cell size distribution of Dirichlet-Voronoi systems [15].

Note that both V_q and V_g in Fig. 3 have been normalized by the mean volume associated with the sample's grains, $\bar{V}_g$. This has been done for comparison between the quadron and grain PDFs, but it has the disadvantage that it washes out some very interesting features. The point is that the mean volume associated with a grain is different for each sample, since the volume associated with a grain, V_g, is the sum over its quadrons, and the porosity is different for different samples. Thus, to compare the peak position and width of curves of different samples having the same grain distribution but different friction coefficients, we need to use a common normalization. Indeed, two interesting features can be observed when comparing systems having different friction, but that are otherwise identical, Figs. 3(a)–3(d). To highlight these features, we replot in Fig. 4 the quadron PDFs of Figs. 3(a)–3(d), normalized by their common mean disk size (unlike $\bar{V}_g$, the mean disk size is just the mean of the volume of each disk Πr^2, which depends only on the size distribution of the disks). The figure shows a trend of widening of the quadron PDF as friction is increased. This is because, on formation, higher friction coefficients lead to less dense granular packs, which have higher values of the compactivity X. This feature is especially clear in disk systems as $\mu \to 0$, which tend to form polycrystals. In the limit $\mu = 0$ the distribution should approach a δ function. To demonstrate this we give here the values of the mean and the standard deviation for

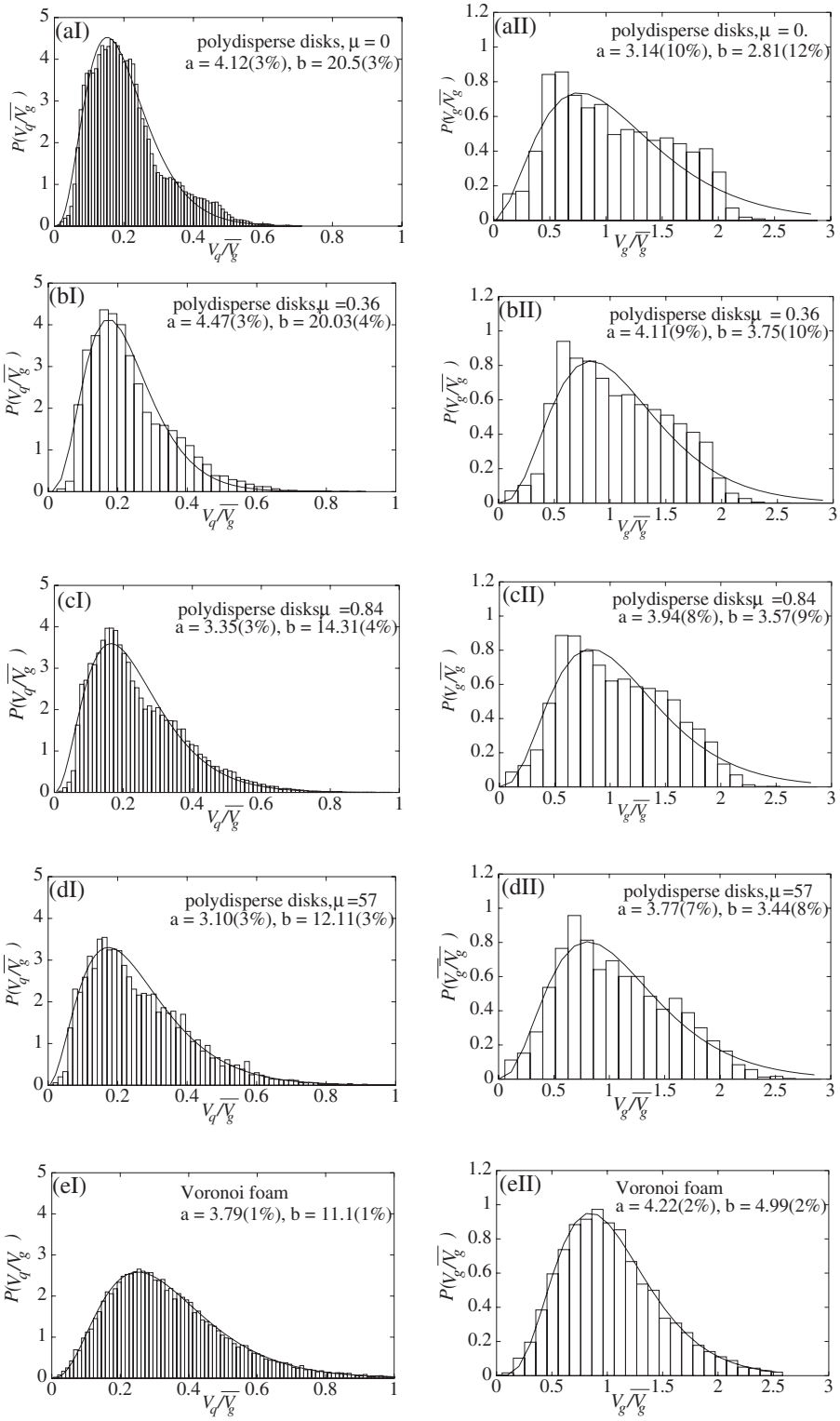

FIG. 3. Probability density functions of quadron volume (left) and grain volume (right) for representative samples: (a)–(d) polydispersed systems of disks with increasing friction coefficients μ =0, 0.36, 0.84, and 57, respectively; (e) Voronoi foam. Unlike the Dirichlet-Voronoi foam the quadron volume PDF of the dense disk samples show nontrivial structure in the form of shoulders. The figures are fitted with a Γ distribution. As seen, the Γ distribution fails to capture the fine details of (a)–(d) and especially the evident shoulders. Clear differences are also seen between the corresponding quadron and grain PDFs. These are quantified by the differences in the fitting parameter a and the uncertainty.

each curve. The four curves in Fig. 4 have friction coefficients of $\mu=0,0.36,0.84,57$, mean quadron volumes of $\bar{V}_q=0.223,0.260,0.275,0.290$, and standard deviations of $\sigma=0.11,0.13,0.15,0.16$, respectively. Another interesting feature is that the peak of the PDFs hardly changes position over the entire friction range, regardless of the aforementioned widening. Interpretation of this feature will become clear in view of the discussion in the next paragraph.

A close scrutiny of the quadron PDF, found in Figs. 3(a)–3(d), shows that, although the Γ distribution appears to give a reasonable description of the statistics, it misses some of the fine features of the distributions. To understand this discrepancy and the origin of these features, it is convenient to focus on the system of very rough monodisperse disks—arguably the simplest of the granular systems. We have decomposed the PDF of the quadron volumes, $P(V_q)$, into the

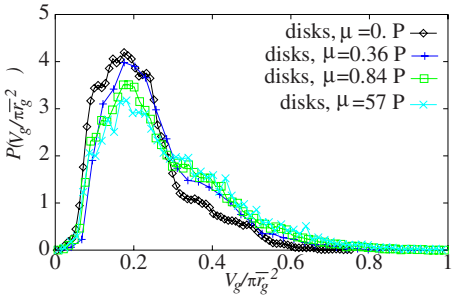

FIG. 4. (Color online) Comparison of the quadron volume PDFs of samples with identical disk size distribution but different friction coefficients. The PDF widens noticeably with friction, but the most probable volume appears to be independent of friction. This feature is washed out in the grain volume PDF and further supports the idea that the structure is best understood in terms of the quadrons as basic elements.

conditional PDFs $P(V_q|z_c)$, where z_c is the number of grains that surround the cell where the quadron q resides. In Fig. 5(a) we plot these conditional PDFs alongside $P(V_q)$. It shows clearly that the features of $P(V_q)$ originate directly from the conditional PDFs. The significance of this observation will be discussed in the concluding section.

In Fig. 5(b), the disphere sample shows peaks and shoulders that preclude the simplified Γ distribution fit and support the decomposition of the PDF into conditional probabilities of Z_c. Note also the broadening of the peaks in Fig. 5(b)(I) in comparison with Fig. 5(a)(I). This is due to the distribution of orientations of the dispheres. Both figures exhibit large differences between the PDFs of the quadron and grain volumes.

The apparent robustness of the peak structure in Fig. 5 prompts us to explore this issue further, and we focus attention on the conditional PDFs $P(V_q|z_c)$. These are the conditional probability densities that quadrons residing in cells of coordination number z_c have volumes that lie between V_q and V_q+dV_q. We observe a significant result: the conditional PDFs are *independent of friction*. Figure 6 shows the first three peaks for two disk systems with friction coefficients $\mu=0.18$ and 0.84. Note that the PDFs fall on top of one another although the distributions of z_c are different between them. In particular, for the former the maximal coordination number is $z_c=6$ and for the latter it is $z_c=9$. On reflection, it is plausible that the independence of the conditional probabilities of friction is a *general property of granular packs* and that our observation may hold for any distribution of grain shapes. This is because, given a grain shape and size distribution, a conditional PDF depends only on the number (or density) of ways that exist to arrange exactly z_c grains to close one loop. If all these configurations occur with equal probability during the formation of the structure then this arrangement is independent of μ.

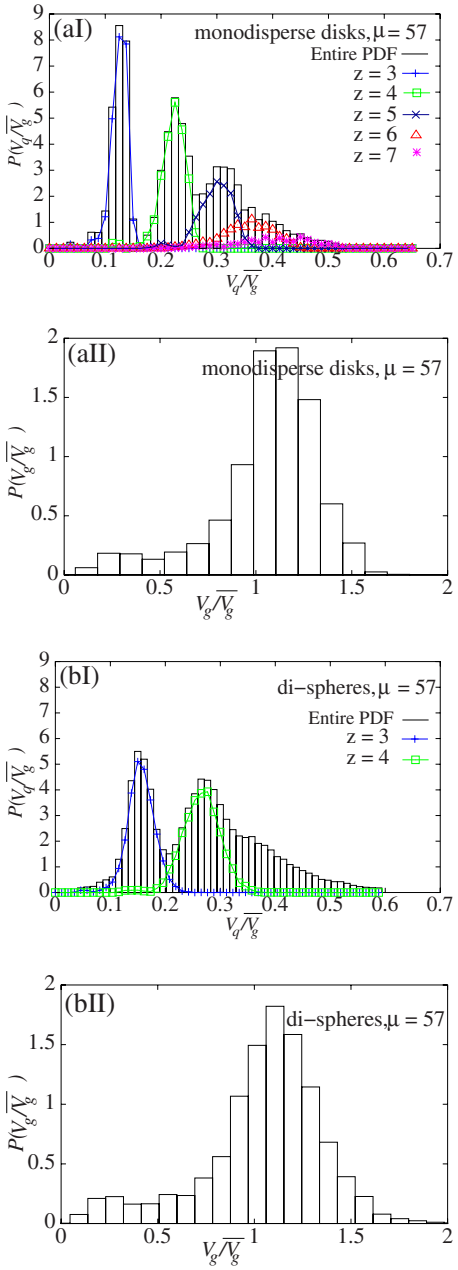

FIG. 5. (Color online) (a) Total PDF of quadron volume in a typical high-friction ($\mu=57$) system of equal-sized disks $P(V_q)$. Also shown are the conditional PDFs $P(V_q|z_c)$ for $z_c=3-7$. It can be clearly seen that the features of the total PDF originate from these. (b) Dispheres with $\mu=57$. Note the apparent differences between the grain and quadron volume PDFs.

V. CONCLUSIONS AND DISCUSSION

In this paper we have presented statistical analyses of several families of granular systems and of Dirichlet-Voronoi foams in 2D. Our main interests were (i) to demonstrate the use of a recent structural characterization method and to determine the relative merits of the use of either quadrons or grains as basic volume elements for the Edwards entropic formalism; (ii) to apply these alternative descriptions to the analysis of the structure; (iii) to use the method to gain a fundamental understanding of the effects of grain shapes and intergranular friction on the statistics of the structure. Both connectivity, via grain and cell coordination numbers, and

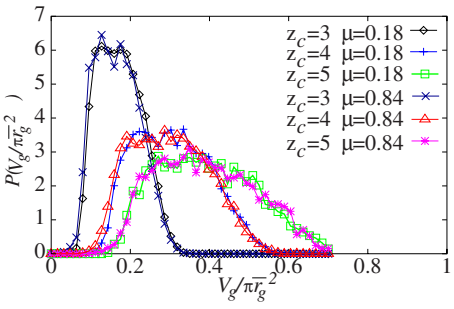

FIG. 6. (Color online) Comparison of the conditional probabilities $P(V_q|z_c)$ for $z_c=3$, 4, and 5 between two monodisperse disk systems of friction coefficients $\mu=0.18$ and 0.84.

the partition of the system volume, by quadrons and grains, have been studied.

Our main results are the following. (i) There are distinct differences between the volume PDFs of quadrons and of grains, favoring the use of quadrons as the basic volume elements for the entropic formalism; (ii) we find that the PDF's of quadron volumes have nontrivial peaks and shoulders, which we have traced directly to the distribution of cell coordination numbers z_c; (iii) we make a clear separation between the effects of the grain size and shape distribution and of friction on the system structure.

To analyze the structure we initially studied the distributions of the grain and cell coordination numbers. This produced several expected results—the mean cell coordination number and size increase with friction while the mean grain coordination number decreases. This is in agreement with our proposed relation (7) between the mean coordination numbers of grains, $\bar{z}_g$, and of cells, $\bar{z}_c$. We further observe that, in all the granular systems that we have studied, the most probable coordination number of cells for any friction coefficient was $z_c=3$. Although not surprising, these results are useful for the discussion of our main result (ii).

Next we studied the distributions of grain and quadron volumes. It has been shown that, in two dimensions, the number of quadrons coincides with the number of degrees of freedom [3] and consequently that these variables can be chosen to span the phase space. As such, quadrons are the quasiparticles in the entropic formalism—the equivalent of particles in traditional statistical mechanics. In other words, these are the variables to integrate on in the partition function. Yet, throughout the literature, it is customary to use the distribution of grain volumes for statistical analyses [12,13], and in some cases also for entropic analyses [2,17,18]. Not much attention has been given to the question of whether the two distributions can be used interchangeably. Our results here suggest that they cannot. On a fundamental level, the number of degrees of freedom is equal to the number of quadrons and not to the number of grains, which suggests that these are indeed the natural quasiparticles to use. A possible justification for using the grain volumes instead for the entropic analysis would be if the two PDFs were similar. One of the main results of this paper was a test of this issue. Following conventional wisdom from the literature [12–14], we tried to fit the quadron and grain volume PDFs of polydispersed disks with Γ distributions. While both fits are less than satisfactory, the fit parameters of the grain volume PDFs have errors three times larger. This supports direct visual observations that the two distributions are markedly different and therefore that the common use of the grain volume distribution is misconceived.

This is not to say that the grain volumes cannot be used as the integration variables in the partition function. It is possible in principle if their probability density is known. However, for that, one has to integrate over the $N(\bar{z}_g-1)$ variables that are the difference between the true dimensionality of the phase space and the number of grains. The problem with this is that, even if the integration were possible to carry out practically, it introduces correlations between grain volumes—correlations that need to be taken into account explicitly. Such correlations preclude, *a priori*, low-level approximations such as independence of variables. In contrast, such approximations are not fundamentally wrong when quadrons are used as the basic variables. Thus, the use of quadrons is preferable also for the computation of the partition function and expectation values of structural properties.

Concerning the integration over quadrons, note that, while the quadron volumes are independent, they can nevertheless be correlated. This is no different in thermal systems, where the degrees of freedom are, for example, particle positions and momenta, and where correlations can exist between particles due to interactions. It is because of their independence of one another that quadrons can be used as degrees of freedom. To analyze a system of correlated degrees of freedom, a model is required for the correlations, and this is when approximations come in. Since there are no models so far for interacting quadrons—an interesting and challenging problem in itself—we have used the simplest possible approximation, the ideal quadron gas, wherein quadron volumes are uncorrelated, and which is the analog of the conventional ideal gas model.

Beyond the fundamental question of the suitability for entropic description, we have looked into the features of the quadron volume PDFs and their dependence on system parameters. It had been expected that pack structures would depend on geometrical properties of the grains, such as shape and size distributions, on physical properties, such as friction, and on the formation process of the structure. However, the exact relations between these and the structure have never been understood on a fundamental level. Rather, there are many empirical observations in the literature that could not be modeled theoretically in any systematic manner. Our results here make a significant step in this direction, as we now proceed to discuss.

Our investigation of the PDFs of the quadron and grain volumes has both uncovered nontrivial structures in the form of shoulders and peaks and shown that the quadron and grain volume PDFs are quite different. As mentioned, neither the quadron nor grain volume PDFs of polydispersed disks are described well by Γ distributions. Following the idea that the quadrons may be the fundamental structural elements, rather than the grains, we focued on a detailed study of the quadron volume PDFs, $P(V_q)$. We have discovered that these features can be traced to conditional probabilities of cell volumes given coordination numbers, $P(V_q|z_c)$. A striking observation

is that the position and the form of each conditional probability $P(V_q|z_c)$ are independent of friction and depend only on the distribution of grain sizes. Friction has been found to control only the distribution of z_c and hence the weights of the conditional probabilities within the total PDFs $P(V_q)$. Combining this observation with the fact that $\bar{z}_g$ depends directly on friction shows that expression (7) between the mean coordination numbers of grains $\bar{z}_g$ and of cells $\bar{z}_c$ relates friction directly to structure.

Furthermore, the dependence of $P(V_q|z_c)$ only on grain size distribution can be extended readily to the distribution of grain shapes—the number (or density) of ways to partition a cell of z_c sides into quadrons is determined predominantly by the shapes of the grains around it. In contrast, friction governs the relative frequency of occurrence of cells of z_c sides through the formation process of the granular pack. This argument further suggests that, in addition to friction, any factor that affects the formation of the pack, such as gravity, density of deposition, and inertial effects, determines the relative weights of the conditional probabilities. Thus, there is a clear separation between the effects of friction and the formation process and of grain shape characteristics on the structure. This observation is quite significant in view of the extensive work in the literature devoted to understanding these issues.

The insight from our results implies that it may be possible to make quantitative predictions about the exact shape, position, and width of given the shape characteristics, but such calculations are not straightforward theoretically, except for very simple cases (see below). For example, the orientation of the dispheres in Fig. 5(b) broadens the peaks, in comparison to the peaks of equal-sized disks found in Fig. 5(a), and blends them together. The size distribution of grains also broadens the peaks and mixes them up [Figs. 3(a)–3(d)]. We expect that wide polydispersity of grain sizes would wash out some of the peak structure and the fine details. Comparing the cell coordination number of cells for the monodispersed and polydispersed disks and the dispheres reveals that there may still be minor effects of the shape characteristics on the weights as well, but this issue requires more detailed investigation.

The above insight suggests several possible experiments. (i) It would be interesting to test the hypothesis that the process of pack formation affects the structure in a similar way to friction, i.e., by modifying the relative weights of the $P(V_q|z_c)$'s. This could be tested experimentally on a range of structures where the grain size and shape distributions are kept fixed while friction and shaking are varied, and vice versa. (ii) The difference between the effects of shape characteristics and other factors, such as friction and different formation processes, suggests guidelines for the study of granular systems. For example, with a fixed distribution of grain shapes, we expect unchanged peak forms as discussed above. Changing then the nonshape parameters, e.g., friction or shaking protocols, would make it possible to isolate the effects of these parameters on packing fractions within the entropic formalism. Conversely, it is possible to fix physical properties and vary grain shape distributions to investigate the effects on peak shapes, a problem that is of essential importance in mining technology [20]. We believe that such experiments, combined with our analysis here, will provide better understanding of the direct effects of friction on the structure, and therefore on the porosity, of granular systems. Further combination of the analysis presented here with the entropic formalism should provide a powerful way to classify and characterize structures of granular systems.

The absence of data on the PDF of quadron volumes led to the use of simplistic forms in the literature of the density of states [2,3,18,19]. Therefore, the quantitative results obtained here form a much-needed link between real granular materials and the analytical studies based on compactivity and the entropic formalism. Our results for the rich structure of the PDFs of quadron and grain volumes and for the relations between shape characteristics, formation protocols, and pack structural statistics suggest that the practice of a heuristic fit of the total PDF by one Γ function is too naive. A more appropriate fit would be of the various peaks in the PDFs using the relations to cell statistics as outlined in this paper. This can be done in a number of ways, using a Γ [14] or Gaussian fit to the peaks. Unfortunately, this requires a large number of fitting parameters. For example, if we assume that the peaks are Gaussian, then for each peak we have to fit three parameters: the position (mean), the width, and the height. Even in our systems, which have a relatively limited range of cell coordination numbers $3 \leq z_c \leq 9$; this results in about 20 parameters. In contrast, the Voronoi cells produced by a Poisson process of unit density are a special case because the PDFs of cell volumes with a specific coordination number z_c are Γ distrributed with $a=z_c$ and $b=1$ [14]. This leaves only one parameter to fit per peak, its height, which is related to the occurrence probability of a z_c-sided cell. However, it is doubtful that this simplification can be extended to more general grain packings.

In view of these difficulties, it is useful to try to understand the shape of the basic peaks from first principles. This is feasible in disk systems for $z_c=3$, for example. In these systems $z_c=3$ corresponds to cells enclosed within three circles and, if we know the polydispersity of the radii, it is possible to calculate the distribution of the volumes of the three quadrons that such a cell gives rise to. In particular, in monodisperse systems this would be exactly a δ function. A similar analytical calculation for the peak corresponding to $z_c=4$ in disk systems is probably also possible. But even for the disk systems these calculation become increasingly involved as z_c increases. For nondisk systems such calculations are prohibitively difficult and it is best to resort to numerical evaluations. Nevertheless, it is probably possible to obtain estimates for the positions of the peaks, using simplifying assumptions. We shall not continue along this line here because our aim has been only to attract attention to this phenomenon and to the understanding of the PDFs.

The weights of the peaks cannot be addressed using the above analysis because it involves the relative occurrence probabilities of different values of z_c and these weights must be strongly sensitive to the physical properties of the system, especially friction. For example, in perfectly smooth systems, we expect all cells to have essentially three edges, $z_c=3$, which obviates the question of the relative weights. With increasing friction, cells of more edges occur. Indeed, one of our main results is that the relation between μ and the oc-

currence probability of z_c is a key to understanding the effect of friction on the structure of granular systems.

Finally, it is important to emphasize that the fine details of the PDFs are easier to detect and understand by choosing the quadrons as the basic volume elements, rather than grains. This is because the volume associated with any grain consists of several quadrons, each belonging in principle to a different cell. Therefore, grain volume distributions mix different peaks and make them difficult to detect. This is another advantage of analyzing the quadron, rather than grain, volumes.

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