Granular Entropy: Explicit Calculations for Planar Assemblies

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(Received 30 October 2002; published 21 March 2003)

This paper proposes a new volume function for calculation of the entropy of planar granular assemblies. This function is extracted from the antisymmetric part of a new geometric tensor and is rigorously additive when summed over grains. It leads to the identification of a conveniently small phase space. The utility of the volume function is demonstrated on several case studies, for which we calculate explicitly the mean volume and the volume fluctuations.

DOI: 10.1103/PhysRevLett.90.114303 PACS numbers: 45.70.Cc, 64.30.+t

It has been shown experimentally and computationally [1,2] that jammed granular systems can be described by statistical mechanics in appropriate circumstances, validating theoretical concepts [3,4]. The simplest approach [3] involves the introduction of compactivity, $X = \partial V/\partial S$, which plays the role of temperature in thermal systems. To quantify X, one needs to calculate the entropy S as a function of the volume V and therefore the volume as a function of the position and coordination of the N grains, i.e., a function $\mathcal{W}$ to complete the analogy between thermodynamics of equilibrium and these nonequilibrium athermal systems $E \to V$; $H \to \mathcal{W}$; $S(E, V, N) \to S(V, N)$. This Letter follows a recent analysis [5] of planar assemblies in terms of loops and voids. Each grain, g, can be characterized in terms of the Z_g grains which it is in contact with, g', and the position of these contact points, $\vec{r}_{gg'}$ (see Fig. 1). For each grain we define a center, $\vec{r}_g = (1/Z_g)\sum_{g'=1}^{Z_g} \vec{r}_{gg'}$, and vectors $\vec{r}_{lg}$ that connect the contact points. The latter vectors form a loop around grain g that is defined to circulate in the clockwise direction. Each vector along this loop can be uniquely identified in terms of the grain g and a neighboring void l. The vectors r_{lg} also form polygons around the voids, whose edges circulate in the anticlockwise direction (see Fig. 1). To each void, we assign a center $\vec{r}_l = (1/Z_l)\sum_{l=1}^{Z_l} \vec{r}_{lg}$, where the sum runs over the Z_l grains that surround void l. Finally, we define a set of vectors $\vec{R}_{lg} = \vec{r}_l - \vec{r}_g$ that extend from the center of grain g to the center of a neighboring void l. This network is self-dual to the $\vec{r}$ network so that for each $\vec{R}$ vector there is an $\vec{r}$ vector that intersects it.

We can now define a one-grain geometric tensor $\hat{C}_g$,

$$C_g^{ij} = \sum_l r_{lg}^i R_{lg}^j, \tag{1}$$

where the sum runs over all the voids surrounding grain g and $i, j = x, y$ index Cartesian components. Each term in this expression involves only one self-dual pair of vectors and has a straightforward geometrical interpretation: Its antisymmetric part is exactly $A_g\hat{\boldsymbol{\epsilon}}$, where $\hat{\boldsymbol{\epsilon}} = \left(\begin{smallmatrix} 0 & 1 \\ -1 & 0 \end{smallmatrix}\right)$ and $A_{lg} = \frac{1}{2} r_{lg} R_{lg} \cos\alpha_{lg}$ is the volume of the quadrilateral formed by $\vec{r}_{lg}$ and $\vec{R}_{lg}$, shown shaded in Fig. 1. Its symmetric part measures the deviation of this quadrilateral from a perfect rhombus [5]. Note that this holds even when the grain has only two contacts, in which case the quadrilaterals degenerate into triangles. In an isostatic assembly of rough grains $\langle Z_g \rangle = 3$, giving rise to $3N$ quadrilaterals altogether. The volume of the entire system can then be written as a sum over all grains,

$$\mathcal{W}\hat{I} = \frac{1}{2}\sum_g \hat{C}_g \cdot \hat{\boldsymbol{\epsilon}}, \tag{2}$$

where $\hat{I}$ is the identity matrix. It can also be recast more conveniently as a sum over all the quadrilaterals (henceforth indexed by n) that the $\vec{r} - \vec{R}$ pairs make,

$$\mathcal{W} = \frac{1}{2}\sum_n r_n R_n \cos\alpha_n, \tag{3}$$

where $r_n = |\vec{r}_{lg}|$. This volume function (VF) is additive as

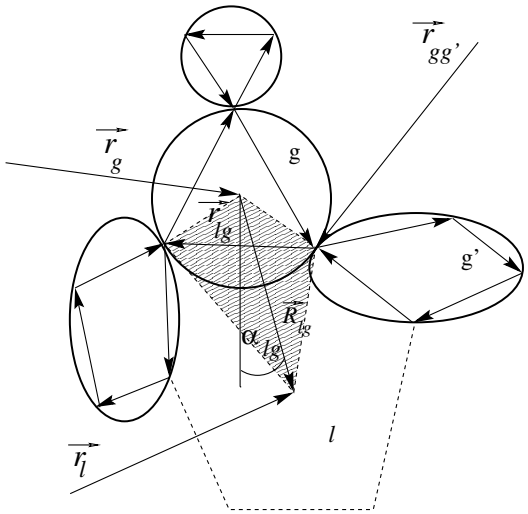

FIG. 1. The geometric construction around grain g. The vectors $\vec{r}_{lg}$ connect contact points clockwise around each grain g and give rise to anticlockwise loops l around each void. The vectors $\vec{R}_{lg}$ connect from grain centers to loop centers. A one-grain geometric tensor is defined as $\hat{C}_g = \sum_l \vec{R}_{lg}\vec{r}_{lg}$.

required and its simple form makes it convenient for analytical calculations.

The partition function can be written as [3]

$$Z = \int e^{-[\mathcal{W}(\{q_n\})]/(\lambda X)} \Theta(\{q_n\}) \Pi dq_n, \tag{4}$$

where $\{q_n\}$ is a set of internal degrees of freedom, $\Theta(\{q_n\})$ is a probability density function (PDF) that is subject to the constraint that appropriate grains are in contact. The coefficient λ is the analogue of Boltzmann's constant with λX ($\equiv 1/\beta$ henceforth) having dimensions of volume (L^2 in two dimensions). The analogue of free energy is the effective volume $Y = -\ln Z/\beta$, the (dimensionless) volumetric entropy is $S = \beta^2 \partial Y/\partial\beta$, and the mean volume is $\langle V \rangle = Y + S/\beta$.

Substituting the VF (3) into Eq. (4), we obtain for the partition function

$$Z = \prod_n \left[\int_0^\infty dr_n \int_0^\infty dR_n \int_{-\pi/2}^{\pi/2} d\alpha_n \right] \times e^{-\beta \sum_n r_n R_n \cos\alpha_n/2} \Theta(\{r_n\}, \{R_n\}, \{\alpha_n\}). \tag{5}$$

The values of the variables R, r, and α are constrained by a minimal volume that the assembly can attain, $v_{\min}$, below which further compactification is impossible without causing grains to overlap, and a maximal volume, $v_{\max}$, above which dilatation is impossible without the loss of mechanical equilibrium and fluidization of the assembly. Although these variables are correlated (see below), it is instructive to first assume independent distributions.

A. Correlation-free systems.—When the degrees of freedom are independent, Θ can be written as a product of PDFs of the individual variables. As a first example, let us consider the following:

$$\Theta = \prod_{n=1}^{\langle Z_g \rangle N} \frac{\delta(R_n - R_0)\delta(\gamma_n - \gamma_0)}{r_{\max} - r_{\min}}; \qquad r_{\min} < r < r_{\max}. \tag{6}$$

Although simplified, this form yields the essential behavior observed in experiments [1,2]. Substituting (6) in (5), the one-grain partition function is found to be

$$z \equiv Z^{1/N} = \left[\frac{e^{-\beta v_{\min}}}{\beta \Delta v} (1 - e^{-\beta \Delta v}) \right]^{Z_g}, \tag{7}$$

where Z_g is the coordination number of the grain and the volumes $v_{\min}$, $v_{\max}$, and Δv are, respectively, $R_0\gamma_0 r_{\min}/2$, $R_0\gamma_0 r_{\max}/2$, and $R_0\gamma_0(r_{\max} - r_{\min})/2$. The volumetric entropy can be directly calculated from this expression and the mean volume is

$$\langle v \rangle = Z_g \left[\frac{v_{\min} + v_{\max}}{2} + \frac{1}{\beta} - \frac{\Delta v}{2} \operatorname{ctgh}\left(\frac{\beta \Delta v}{4} \right) \right], \tag{8}$$

and the volume fluctuations are

$$\langle \delta v^2 \rangle = Z_g \left[\frac{1}{\beta^2} - \left(\frac{\Delta V}{\sinh(\beta \Delta V)} \right)^2 \right]. \tag{9}$$

When $\beta \to \infty (\lambda X \to 0)$, we find that $\langle v \rangle \to Z_g v_{\min}$ and $\langle \delta v^2 \rangle \to 0$, while when $\beta \to 0 (\lambda X \to \infty)$, $\langle v \rangle \to Z_g[(v_{\min} + v_{\max})/2 - \beta \Delta v^2/3]$ and $\langle \delta v^2 \rangle \to \Delta v^2/3$. It is interesting to note that expression (8) has the same form as the one-dimensional result [3,6]. The current formalism extends that result to two dimensions and makes it possible to associate it with a particular distribution of degrees of freedom.

Let us consider a more realistic case,

$$\Theta = \prod_{n=1}^{\langle Z_g \rangle N} P(r_n)\delta(R_n - R_0) C_\gamma e^{-[(\gamma_n - \gamma_0)^2]/(2\sigma_\gamma^2)}, \tag{10}$$

where $\sigma_\gamma \ll 1$, C_γ is the normalization constant of the PDF of γ, and $P(r_n)$ is kept arbitrary for the moment. The Gaussian PDF for γ around γ_0, whose value is close to 1, represents a narrow distribution of the angle α_n around zero and therefore small deviations from a rhombus [7]. As we argue below, $P(R_n)$ is Gaussian-like and is narrower than $P(r_n)$, which justifies its approximation as a delta function. Integration over all γ_n and R_n gives

$$Z = \left[\int dr P(r) \psi(r) e^{[(\beta \sigma_\gamma v)^2/(2\gamma_0^2)] - \beta v} \right]^{\langle Z_g \rangle N}, \tag{11}$$

where

$$\psi(r) = \left[\phi\left(\frac{u}{\sqrt{2}} + \frac{1 - \gamma_0}{\sqrt{2}\sigma_\gamma} \right) - \phi\left(\frac{u}{\sqrt{2}} + \frac{\gamma_{\min} - \gamma_0}{\sqrt{2}\sigma_\gamma} \right) \right] \times \frac{\sigma_\gamma C_\gamma}{\sqrt{2/\pi}},$$

ϕ is the error function, and $u = (\sigma_\gamma/\gamma_0)\beta v$. For $u\sigma_\gamma \ll 1$ (high compactivity)

$$Z \approx \left[\int_{r_{\min}}^{r_{\max}} P(r) e^{-\beta \chi v} dr \right]^{Z_g}, \tag{12}$$

where

$$\chi = 1 - [e^{-(1-\gamma_0)^2/(2\sigma_\gamma^2)} - e^{-(\gamma_{\min} - \gamma_0)^2/(2\sigma_\gamma^2)}] \frac{C_\gamma \sigma_\gamma^2}{\gamma_0}.$$

For example, assuming that $P(r)$ is uniform gives that the mean volume per grain is

$$\langle V \rangle \approx Z_g \chi \left[\frac{v_{\max} + v_{\min}}{2} + \frac{1}{\beta\chi} - \Delta v \operatorname{ctgh}(\beta \chi \Delta v) \right], \tag{13}$$

and the volume fluctuations are

$$\langle \delta v^2 \rangle = Z_g \left[\frac{1}{\beta^2} - \left(\frac{\chi \Delta V}{\sinh(\chi \beta \Delta V)} \right)^2 \right]. \tag{14}$$

The analysis of these expressions as $\beta \to 0, \infty$ is straightforward and gives the same results as for the previous case with Δv replaced by $\chi \Delta v$. Similarly, it is straightforward to evaluate these quantities for low compactivity.

The expressions for the volume fluctuations can be now related to response functions and diffusion processes in

granular systems. It is unclear at this stage how large the error is that is introduced by the assumption of independence of the variables. One advantage of the VF proposed here is that it makes it possible to elucidate this issue.

B. Correlated systems.—To go beyond the assumption of independence, the key observation is that correlations arise from two sources: (i) relations between the vectors $\vec{r}$, and (ii) the self-duality of the r and R networks. The former can be traced to loops. Whenever M $\vec{r}_{lg}$ vectors close a loop, their sum vanishes and one of them (two degrees of freedom) can be expressed in terms of the other $M-1$ vectors. This introduces two δ functions into the Jacobian, reducing the phase space dimensionality by two. The smallest such loops occur around the grains, $\sum_{l=1}^{Z_g} \vec{r}_{lg} = 0$. When these are independent, each such loop contributed to the Jacobian a term of the form $\prod_{i=1}^{Z_g} P(\vec{r}_i)$. The correlations force a modification of this term to $[\prod_{i=1}^{Z_g-1} P(\vec{r}_i)]\delta(\vec{r}_n + \sum_{i=1}^{Z_g-1} \vec{r}_i)$. Note that the value of Z_g is distributed throughout the system, giving rise to a corresponding distribution of such terms in the partition function. Voids are also surrounded by loops of $\vec{r}$ vectors (e.g., void l in Fig. 1), each further reducing the phase space dimensionality by two. Grain and void loops are the only irreducible loops in the system (namely, all other loops can be decomposed into these) and therefore only they give rise to correlations of the first type. This observation has an interesting implication: An isostatic system of N grains has $3N$ $\vec{r}$ vectors. The N granular loops and the $N/2$ void loops yield $3N/2$ of these dependent on the rest. Namely, only *half* the $\vec{r}$ vectors are independent, giving $3N$ degrees of freedom.

Turning to the second type of correlations, recall that $\vec{R}_{lg}$ extends from the centroid of grain g, $\frac{1}{Z_g}\sum_{l'=1}^{Z_g-1}(Z_g - l')\vec{r}_{l'g}$, to that of loop l, $\frac{1}{Z_l}\sum_{g'=1}^{Z_l-1}(Z_l - g')\vec{r}_{lg'}$. Therefore, it can be expressed as a *linear combination* of the vectors forming the g and l loops, $\vec{R}_{lg} = \sum_{k=1}^{Z_l+Z_g-2} a_n \vec{r}_n$, and is uniquely defined in terms of the $\vec{r}$ network. On average, an $\vec{R}$ vector depends on $\langle Z_g + Z_l - 2\rangle = 7$ $\vec{r}$ vectors and, since there are two grains to a void, then $\langle R\rangle \approx \sqrt{2}\langle r\rangle$. It follows that $P(R)$ can be safely approximated by a Gaussian around this value. The dependence of the R variables on the r further means that the γ variables can also be expressed in terms of these because $\gamma_n = \sqrt{1 - \vec{R}_n \cdot \vec{r}_n}$. In fact, γ_n depends on average on eight r variables and therefore $P(\gamma_n)$ can also be approximated as a Gaussian. Combining all the above, the VF can be written as

$$\mathcal{W} = \frac{1}{2}\sum_{n,m=1}^{3N/2} a_{nm} r_n^x r_m^y. \tag{15}$$

The coefficients $0 < a_{nm} < 1$ are rational and form a sparse matrix of zeros on the diagonal and, on average, seven finite elements in each row. Although appealing, this quadratic form is useful only with particular forms of $P(\{r\})$ and even then it requires knowledge of the statistics of a_{nm}. Its main disadvantage is in mixing the basic quadrilateral volume units and so losing the clear geometrical interpretation of Eq. (3).

Although the correlations reduce the phase space dimensionality to $3N$, it is difficult to take them all explicitly into consideration. To make progress, we consider all the original $9N$ variables but include the lowest order correlations—those coming from the intragranular loops, which are the smallest. The justification for this approximation is that since the R and γ variables depend on several r degrees of freedom then they are more narrowly distributed and so can be considered as background fluctuations. We believe that this approximation captures the correct physics in granular assemblies.

To illustrate the effects of the intragranular loops, consider a triply coordinated circular grain of diameter D. The three contacts on the circumference form a triangle of vectors $\vec{r}_{lg}$, and we wish to know the probability $P(r_1, r_2, r_3)$ that the lengths of its sides lie inside $(r_1, r_1 + dr_1)$, $(r_2, r_2 + dr_2)$, and $(r_3, r_3 + dr_3)$. The PDF of one of the sides falling between r and $r + dr$ is

$$P(x = r/D) = 1/(\pi\sqrt{1 - x^2}). \tag{16}$$

The first two sides can be chosen independently, but once these are in place, the third side is determined by

$$\begin{aligned} x_3^2 &\equiv f^2(x_1, x_2) \\ &= 2x_1x_2\sqrt{(1 - x_1^2)(1 - x_2^2)} + (x_1^2 + x_2^2) - 2x_1^2x_2^2, \end{aligned} \tag{17}$$

and therefore

$$P(x_1, x_2, x_3) = \frac{\delta[x_3 - f(x_1, x_2)]}{\pi^2\sqrt{(1 - x_1^2)(1 - x_2^2)}}. \tag{18}$$

Although this analysis can be extended to $Z_g > 3$ and to noncircular grains, we do not give it here. We next demonstrate the effect of intragranular correlations on the effective volume.

Consider a large ensemble of N randomly assembled monodisperse circular grains, each contacting exactly three neighbors. Ignoring void loop correlations makes the grains effectively independent but not the quadrilaterals. We take a Gaussian form for $P(\gamma)$ and practically a delta function for R. The one-grain partition function ($z = Z^{1/N}$) is then

$$\begin{aligned} z = \int \prod_{n=1}^{3} &[e^{-\beta r_n R_n \gamma_n/2} P(R_n) P(\gamma_n) dr_n dR_n d\gamma_n] \\ &\times P(r_1)P(r_2)\delta[r_3 - f(r_1, r_2)], \end{aligned} \tag{19}$$

where the lengths r_n are measured in units of the grain diameter D. Integrating over the R and γ variables yields

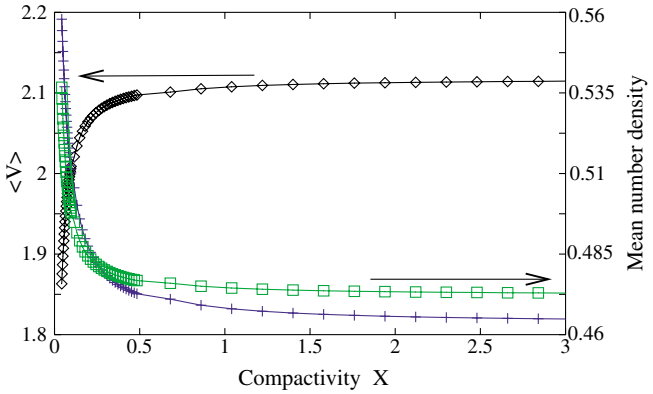

FIG. 2 (color online). The volume (left axis) and density (right axis) of a trivalent granular assembly of monodisperse circular grains. The moduli of the intragranular vectors connecting the contact points are taken to be correlated in the calculation. The PDFs of γ and R are, respectively, Gaussian and a δ function at the periodic lattice value $R_0 = 2D/\sqrt{3}$. Two density plots are presented: one assuming independent r variables (squares) and the other taking the intragranular correlations into account (+). For $X > X_0 = 0.265$, the correlations compactify the system while below this value they lead to an opposite effect for the reason discussed in the text. A significant feature is that the difference between the two plots is quite small. The densities of the two calculations differ only by 0.009 ($\approx 2\%$) at $\lambda X = 5$ and by 0.021 at $\lambda X = 0.04$ ($\approx 4\%$).

$$z = \int dr_1 dr_2 P(r_1)P(r_2)\psi(r_1)\psi(r_2)\psi[f(r_1, r_2)] \times \exp\left[\frac{\beta^2 R_0^2 \sigma_\gamma^2 (r_1^2 + r_2^2 + f^2)}{8} - \frac{\beta R_0 \gamma_0 (r_1 + r_2 + f)}{2}\right]. \quad (20)$$

Substituting from (18), we can now compute Y and from it the mean volume and its fluctuations. The mean volume is plotted in Fig. 2 as a function of λX. The asymptotic value as $X \to \infty$ is the average of the close and loose random packings, while the value at $X \to 0$ is the close random packing volume. In terms of grain number density, these are 0.473 and 0.537, respectively.

We can now assess the assumption of independence. Using form (18) for $P(r)$ of all three quadrilaterals, we also plot in Fig. 2 the density of the system with and without correlations. It can be observed that the correlations reduce the density of the system at high compactivity, but increase it compared to the correlation-free system at low compactivity. The crossover is at $X_0 = 0.265$. This is expected because the sides cannot all assume simultaneously either $r_{\max}$ or $r_{\min}$. This constrains the high compactivity limit from overexpanding and the low compactivity regime from overshrinking. In spite of this fundamental difference, it is rather surprising how close the densities of the two systems are over the entire range. The densities of the correlated and uncorrelated systems in the high compactivity limit are, respectively, 0.473 and 0.464 (1.9% difference), while at the low compactivity limit ($\lambda X = 0.04$) the values are 0.537 and 0.558 (3.8% difference). This suggests that simple models that do not take the correlations into account may prove sufficiently accurate for various purposes.

In conclusion, we have proposed a new VF for planar granular assemblies that is exactly additive over grains. Nevertheless, we have identified the quadrilaterals, rather than the grains, as the basic units of the system because it is their volumes that cover the entire volume. A sum over grain volumes without involving the quadrilaterals has the disadvantage of requiring an assumption on approximate grain volume. We point out that the VF proposed here applies to any assembly in mechanical equilibrium, not only to isostatic ones. We have found that the relevant phase space is of relatively small dimensionality and identified the sources of geometrical correlations in the system. We have calculated the effective volume and volume fluctuations of several model assemblies both with and without correlations. The identification of quadrilaterals as the basic building blocks is relevant to modeling. For example, a two-volume model for grains, v_1 and v_2 with occurrence probabilities p_1 and p_2, gives a mean volume of [6]

$$\langle V_g \rangle = N\left[\frac{v_1 + v_2}{2} + \Delta v \tanh\left(\beta \Delta v + \ln\sqrt{\frac{p_1}{p_2}}\right)\right]. \quad (21)$$

Applying the same model to quadrilaterals gives a similar expression, but it corresponds to a wider distribution of grain volumes both because a grain has Z_g quadrilaterals and because Z_g itself is distributed. Thus, quadrilaterals as basic units yield more realistic models without investment of extra effort.

We acknowledge discussions with Professor R. C. Ball and Dr. D. V. Grinev.

[1] E. R. Nowak, J. B. Knight, E. Ben-Naim, H. M. Jaeger, and S. R. Nagel, Phys. Rev. E **57**, 1971 (1998); P. Philippe and D. Bideau, Europhys. Lett. **60**, 677 (2002).
[2] H. A. Makse and J. Kurchan, Nature (London) **415**, 614 (2002); A. Barrat, V. Colizza, and V. Loreto, Phys. Rev. E **66**, 011310 (2002); A. Barrat, J. Kurchan, V. Loreto, and M. Sellitto, Phys. Rev. Lett. **85**, 5034 (2001).
[3] S. F. Edwards and R. B. Oakeshott, Physica (Amsterdam) **38D**, 88 (1989); Physica (Amsterdam) **157A**, 1080 (1989).
[4] A. Mehta and S. F. Edwards, Physica (Amsterdam) **157A**, 1091 (1989).
[5] R. C. Ball and R. Blumenfeld, Phys. Rev. Lett. **88**, 115505 (2002).
[6] S. F. Edwards and D. V. Grinev, Phys. Rev. E **58**, 4758 (1998); Chem. Eng. Sci. **56**, 5451 (2001).
[7] This is related to the result in [5] that the Vol. average of the symmetric Pt. of $\sum_g \hat{C}_g$ vanishes as $1/\sqrt{N}$.