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Review

Granular statistical mechanics – a personal perspective

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Abstract. The science of granular matter has expanded from an activity for specialised engineering applications to a fundamental field in its own right. This has been accompanied by an explosion of research and literature, which cannot be reviewed in one paper. A key to progress in this field is the formulation of a statistical mechanical formalism that could help develop equations of state and constitutive relations. This paper aims at reviewing some milestones in this direction. An essential basic step toward the development of any static and quasi-static theory of granular matter is a systematic and useful method to quantify the grain-scale structure and we start with a review of such a method. We then review and discuss the ongoing attempt to construct a statistical mechanical theory of granular systems. Along the way, we will clarify a number of misconceptions in the field, as well as highlight several outstanding problems.

1 Introduction

Grains and particulates constitute the second most ubiquitous form of matter on Earth after water. On other planets and stellar objects it is most likely the most prevalent form. Correspondingly, granular materials play a major role in our lives and in human society. The sand around us, the powders we use, the cereals we consume and transport and the snow powder we ski on. Understanding the physics and behaviour of granular matter is essential to a wide range of industries since almost all industrial materials pass, at some stage of their processing, through a particulate form. Consequently, efficient transportation and processing of particulates and powders is essential in our society.

The significance of this form of matter has been realised early on in human history and much scientific and engineering attention focused on it for millennia. In most relevant cases, the modelling of a collection of many grains is an exercise in coarse-graining. This is because the default scientific description of any large system is

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via continuum models. The problem with treating granular systems (GS) discretely occurred already to Joseph son of Jacob: "And Joseph gathered corn as the sand of the sea, very much, until he left numbering; for it was without number" (Genesis 41:49). Arguably, he was the first documented person to anticipate the necessity of descriptions of many-body systems beyond the discrete. Historically, the study of these materials was by engineers and technologists, due to the significance to the industry and commerce. However, the last two decades saw a burst of research on these systems by physicists, due to the rich and often unusual behaviours of these materials, combined with the fundamental theoretical challenges that their modelling present.

Since most interesting granular assemblies consist of large collections of particles, there have been a number of attempts to model these systems statistically [1–6], including by Hans Herrmann, whom this volume honours [7]. In 1989, Edwards and collaborators [8–10] proposed a statistical mechanical formalism to describe them in much the same way that is done for thermal systems. A complete statistical mechanics formalism should be the main route to derivation of equations of state and constitutive properties. Here we describe our perspective on the attempts to formulate such a theory. We review the construction process, describe the insights and understanding that have been gained along the way, and point out some misconceptions that linger in the field.

2 The foundations of granular statistical mechanics (GSM)

Particulate matter is said to be a granular material if the particles are sufficiently large to make thermal fluctuations irrelevant. Under Earth's gravity this criterion translate to the condition that the thermal energy k_BT at temperatures around $T \approx 25\,^{\circ}\mathrm{C}$ is much smaller than the potential energy it takes to move the particle by its own size l, $k_BT \ll mgl$. One implication of this is that, unless agitated externally, granular materials are effectively at zero temperature. As a result, once a granular assembly reaches mechanical equilibrium, it is in a particular metastable state. This has a significant implication for the theory of GSM – it means that ergodicity does not hold. Ergodicity in thermal systems constitutes of the assumption that the statistics of an ensemble of systems at a given time is the same as the statistics of one system at different moments in time. This assumption is well justified in thermal systems, which are never really static – during any experimental measurements almost all particles would have changed position and momentum many times. Using this assumption, we can predict dynamic properties from ensemble statistics and use insight from dynamics to infer static properties and constitutive relations. In contrast, granular systems are generically either stuck in a given configuration or they explore a very limited number of configurations, unless externally excited. It is then irrelevant to consider their statistics at different moments. It follows that GSM is ensemble statistics only.

The presumption that a statistical mechanical approach should work for granular systems at all is based on the apparent reproducibility of the macroscopic behaviour of these systems in a wide range of situations. It is such reproducibility that allows us to walk confidently on the beach without the fear of the underlying sand turning to be quicksand, or to build skyscrapers on soils and occupy them without too much fear for their stability, or to trust the hourglass to clock almost exactly three minutes every time it is turned. That the behaviour of granular matter is reproducible is not only an assumption underlying attempts to construct a predictive theory but it also suggests that, in spite of the astronomical number of possible configurations, there is a typical behaviour and therefore that the grain-scale seeming randomness is underpinned by limit statistics. However, this does not mean granular matter is in equilibrium like

thermal systems; it only means that it reaches well defined steady states. We presume that these steady states can be described by a statistical mechanical formalism.

Since energy is hardly relevant to many of the fundamental issues of static granular matter, the suggestion in [8–10] was to construct GSM, based on entropy alone. A particular configuration of the granular system is then regarded as a micro-state and the entropy is the logarithm of the number of all possible configurations. This immediately highlights another departure from thermal statistical mechanics. Conventionally, we assume a *uniform measure*, namely, that during the duration of any relevant experiment, the system is shuffled through so many micro-states that it samples a typical portion of the phase space. But in granular systems the micro-states cannot be accessed dynamically and it is therefore unclear how transferrable this assumption is to GSM. Without this assumption, we also need a model for the different probabilities of micro-states, which makes the formalism less general and much more difficult to apply. Several numerical and experimental tests of the uniform measure assumption have been carried out on small systems [11–13], but due to the astronomically large number of possible configurations, it is impossible to sample sufficiently many systems to resolve this issue conclusively. Nevertheless, since ergodicity does not apply anyway in most of the systems of interest, then the most practical approach is to ignore the dynamics by which the phase space is explored, assume a uniform measure in the ensemble statistics and then test the resulting predictions against experimental measurements.

As in thermal systems, GSM is based on degrees of freedom (DFs), which are a set of independent variables that describe all the possible micro-states. As we shall see below, these are both structural and stress DFs. The DFs define the phase space. The partition function in thermal systems is a normalisation factor, where the probability of every micro-state to occur is proportional to a Boltzmann factor that depends on its energy ϵ, $e^{-\epsilon/k_B T}$. This energy is determined by a global function – the Hamiltonian – which gives the total energy of the system, given the configuration of the DFs in any particular micro-state. In the original GSM [8–10], the Hamiltonian is replaced by a volume function, $\mathcal{W}$, which gives the total volume of the system. Later on it was realised that stress micro-states need also to be included [8–10,14] and that the volume and stress ensembles are coupled [17]. The substitution of the Hamiltonian by other functions necessitated also different measures of the fluctuations, which in thermal systems are parameterised by the temperature. For the volume function the analogue parameter is a scalar "compactivity" X_0 and for the stress ensemble it is a tensor called "angoricity", whose components are X_{ij} (see below). These parameters, which can be derived as Lagrange multipliers, like the temperature in thermal statistical mechanics, are defined as the derivatives of the relevant expectation values with respect to the entropy S, which is the logarithm of the total number of configurational and stress micro-states that the system can assume,

$$T = \frac{\partial \langle \epsilon \rangle}{\partial S} \tag{1}$$

$$X_0 = \frac{\partial \langle V \rangle}{\partial S} \equiv \beta_0^{-1} \tag{2}$$

$$X_{ij} = \frac{\partial \langle \mathcal{F}_{ij} \rangle}{\partial S} \equiv (\beta_{ij})^{-1} \tag{3}$$

where $\mathcal{F}_{ij}$ is the force moment function, whose volume density is the stress.

The partition function consists of both the volume and stress contribution as follows [17]

$$Z = \int e^{-\mathcal{W}/X_0 - \mathrm{Tr}\{\beta \cdot \mathcal{F}\}} D\{\text{structural DFs}\} \mathrm{D}\{\text{stress DFs}\}. \tag{4}$$

An important decision is the choice of the ensemble, i.e. the collection of systems, whose statistics are studied. Traditionally, one considers the micro-canonical, the canonical and the grand-canonical ensembles. For GSM this choice is doubled because there are two phase sub-spaces, the volume and the stress. This offers more possibilities, e.g. a canonical ensemble in the volume and a micro-canonical ensemble in the stress.

The micro-canonical volume ensemble

This ensemble consists of all the configurations (micro-states) of exact volume V that N grains, of a given shape and size distribution, can assume. The entropy of this ensemble, $S(V)$, is defined as

$$S(V) = \ln \left[C \int \delta \left[W\left(\{u\}\right) - V \right] d\{u\} \right] \tag{5}$$

where C is a system size dependent constant that makes the integral non-dimensional (analogous to the factor $1/N!h^{3N}$ in conventional statistical mechanics), $\{u\}$ is the set of all the structural DFs, and W is a volume function that depends on these DFs. This expression already assumes that the micro-states satisfy the constraints on the ensemble, e.g. that the total number of particles is fixed, that they are in mechanical equilibrium, that rattlers are excluded and so on. Alternatively, one can introduce into the integral in (5) a function Θ, consisting of a set of δ-functions that ensure that these constraints are indeed satisfied [8–10].

Any macroscopic structural feature of the system, A, can be expressed as an expectation value of these statistics

$$\langle A \rangle = \frac{\int A\left(\{u\}\right) \delta \left[W\left(\{u\}\right) - V \right] d\{u\}}{\int \delta W \left[\left(\{u\}\right) - V \right] d\{u\}} \tag{6}$$

where the left hand side is a measurable macroscopic quantity and the integrals on the right hand side involve microscopic variables only. The denominator of (6) is the partition function and acts as a normalisation factor.

The canonical volume ensemble

The canonical ensemble is often more useful than the micro-canonical due to the fluctuations in the volume in real experiments. These are analogous to the fluctuations in the energy of thermal systems when coupled to a heat reservoir. The entire granular system can then be regarded as a reservoir (providing volume fluctuations) plus the pack on which measurements are carried out. Assuming that the occurrence probability of a system n, p_n, is proportional to a Boltzmann-factor-like term with the volume replacing the energy, we have the same result as in conventional statistical mechanics,

$$p_n = \frac{1}{Z_v} e^{-\frac{v_n}{X}} \tag{7}$$

where Z_v is known as the structural (or volume) partition function, v_n the volume of the nth assembly (or structural configuration) and X is the "compactivity" – a scalar factor that characterises volume fluctuations analogously to the way that the temperature quantifies thermal fluctuations. The compactivity is defined as the derivative of the mean volume of an ensemble of such systems, $\langle V \rangle$ with respect to the entropy, $X = \partial \langle V \rangle / \partial S$. This is exactly analogous to the definition of the temperature in thermal systems, $T = \partial \langle E \rangle / \partial S$. The partition function is then

$$Z_v = \int e^{-\frac{W(\mathbf{u})}{X}} d^{N_s}\mathbf{u} \tag{8}$$

where $\mathbf{u}$ is a vector of all the N_s structural DFs. Any structural feature, A, can then be obtained as an expectation value over this partition function,

$$\langle A \rangle = \frac{1}{Z_v} \int A(\mathbf{u})\, e^{-\frac{W(\mathbf{u})}{X}} d^{N_s}\mathbf{u}. \tag{9}$$

Particular examples are the mean volume and volume fluctuations, which are, respectively

$$\langle V \rangle = \frac{1}{Z_v} \sum_q V_q e^{-V_q/X} = -\frac{\partial \ln Z_v}{\partial (1/X)} \tag{10}$$

$$\langle V^2 \rangle - \langle V \rangle^2 = -\frac{\partial^2 \ln Z_v}{\partial (1/X)^2} \tag{11}$$

and are the direct analogues of the mean energy and energy fluctuations in conventional thermal systems.

3 Structural description of granular systems

Essential to the GSM is an explicit identification of the structural DFs. However, to do this we first need a quantitative description of disordered granular structures in general. To this end, we use the quadron construction [18–20], to be described below. This method, proposed first for 2D assemblies [18,19] and extended later to 3D [20], is especially useful to the above formalism.

In fact, a local quantitative structural description of disordered granular materials is the starting point for any theory of granular matter, whether statistical mechanics, rheology or mechanics. It is only by coarse-graining from the discrete "micro" structure to the continuum that rigorous and useful prediction can be made. Without a local quantitative description, any discussion of the structure remains abstract, qualitative and phenomenological at best. Many so-called structural descriptors consist of averages over specific structural quantities, such as density-density correlations, diffraction spectra, the mean coordination number, and a host of other correlation functions. Common to all these descriptions is that they constitute volume averages of local structural quantities either over the entire system or over large parts of it.

For a proper theory, the descriptor needs to be local and unambiguous, namely, it must quantify the structure at any arbitrary point within the granular system. A number of works in the literature have attempted to address the structural and

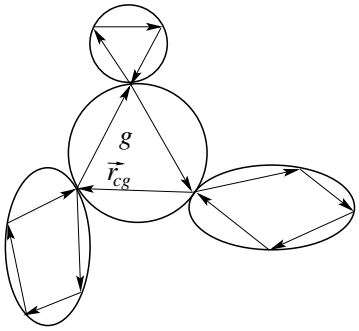

Fig. 1. The contact network of a 2D granular assembly in mechanical equilibrium is a directed graph made of vector edges $\mathbf{r}_{cg}$.

statistical characteristics of granular packings [21–23]. The quadron method, to be described next, appears to us to be superior in its usefulness for a number of aspects of granular science. The method is conceptually the same both in 2D and in 3D and we start by describing it in 2D [18,19] and then show the extension to 3D [20]. The method is based on three conceptual steps.

1. Construction of a connnectivity network: the network is a graph, whose nodes are the inter-granular contact points for rigid particles, or the centroids of contact surfaces for compliant particles. The edges of the graph form polygons (in 2D) and polyhedra (in 3D) around every grain. We focus here on convex grains, in which case these polygons and polyhedra are enclosed inside the space occupied by the grain.
2. Tessellation of the connnectivity graph into elementary volumes – the quadrons – which are generically quadrilaterals in 2D and (non-convex) octahedra in 3D.
3. Quantification of the shape of every quadron by a tensor.

This procedure allows us to quantify the basic structure unambiguously at a sub-granular scale, namely, with volume elements that are smaller than the particles. These volume elements are called *quadrons*. Every grain shares a number of quadrons with the cells surrounding it. While there are other ways to tessellate the space occupied by a granular system, the quadron description has several advantages, which we shall detail below. We review the 2D construction in detail and the 3D one will be described more sketchily. The interested reader can find a detailed description of the latter in the literature [20]. For simplicity, we confine ourselves to description of packs of convex grains, but small non-convexities should not affect the description much [19,24,25].

Quantitative local structural description in 2D

To construct the connectivity network we connect all the contact points around a grain to make a polygon, as shown in Fig. 1. Since the particles are, at most, only slightly non-convex then so are the polygons. The edges of the polygon are then assigned directions, rotating clockwise around a grain. Doing this around every grain generates a directed graph – this is the contact network. In grains with only two contacts the polygon has only two edges, degenerating into two directed lines on top of one another. These directed edges enclose loops around the voids (or cells) in the structure, rotating in the anti-clockwise direction around every loop. Every directed

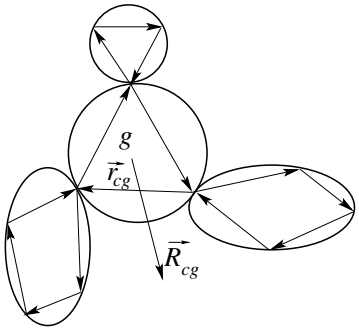

Fig. 2. The conjugate of the contact network is made of the vectors $\mathbf{R}_{cg}$.

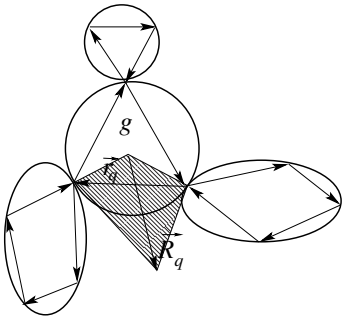

Fig. 3. A quadron q (shaded) is a quadrilateral, whose diagonals are $\mathbf{R}_q(\equiv \mathbf{R}_{cg})$ and $\mathbf{r}_q(\equiv \mathbf{r}_{cg})$.

edge is then described by a vector $\mathbf{r}_{cg}$, uniquely indexed by the grain g, whose contacts it connects, and the cell c that it borders with.

Next we introduce several definitions. For every grain we define a centroid as the mean position vector of the contact points around it,

$$\rho_g = \frac{1}{z_g} \sum_{g'=1}^{z_g} \rho_{gg'} \tag{12}$$

where z_g is the number of contacts (the coordination number) of grain g and the sum is over all the grains g' that are in contact with it. We also define the centroid of each cell, ρ_c, as the mean position vector of the contact points between the grains that surround it. Next, we define a vector $\mathbf{R}_{cg}$, extending from the centroid of g to the centroid of c, $\mathbf{R}_{cg} \equiv \rho_c - \rho_g$. The graph formed by the vectors $\mathbf{R}$ is conjugate (or dual) to the graph formed by the vectors $\mathbf{r}$, namely, every $\mathbf{r}_{cg}$ has one and only one corresponding vector $\mathbf{R}_{cg}$. Here we focus on granular systems in mechanical equilibrium under no external fields, so that there are no body forces – the intergranular forces are generated only by compressive boundary forces. Under these conditions, the polygons around cells must be convex. This, in turn, means that the vectors $\mathbf{r}_{cg}$ and $\mathbf{R}_{cg}$ must cross one another (see Fig. 2).

In Fig. 3 we show a quadrilateral q (shaded), whose diagonals are the vectors $\mathbf{r}_{cg} \equiv \mathbf{r}_q$ and $\mathbf{R}_{cg} \equiv \mathbf{R}_q$. Such a quadrilateral can be constructed for every dual pair $\mathbf{r}_q$ and $\mathbf{R}_q$. The quadrilaterals tile the plane perfectly, without overlaps or gaps, and are the basic volume elements of the structure of the granular assembly. These volume elements play key roles both in GSM and in the stress theory of granular systems and we call them *quadrons*. While generically a quadron is a quadrilateral, for grains

with only two contacts, the quadrilaterals degenerate into triangles. Nevertheless, the quantification described below is not affected by this degeneracy.

We now quantify the shape of every quadron by a local tensor – its shape (or structure) tensor:

$$C_q = \mathbf{r}_q \otimes \mathbf{R}_q. \tag{13}$$

The structure of the granular space associated with grain g is the superposition of the structures of the z_g quadrons that belong to it,

$$C_g = \sum_{q \in g} C_q. \tag{14}$$

The volume (area) of quadron q is

$$\frac{1}{2}\left(C_q - C_q^T\right) = V_q \epsilon; \quad \epsilon \equiv \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix} \tag{15}$$

where C_q^T stands for the transposed of C_q. It is useful to cast the volume as a trace, by rotating $\mathbf{r}_q$, as this form is dimensionally independent and will generalise to 3D,

$$\tilde{C}_q = (\epsilon \cdot \mathbf{r}_q) \otimes \mathbf{R}_q. \tag{16}$$

In terms of this tensor the quadron volume is

$$V_q = \frac{1}{2} Tr\left\{\tilde{C}_q\right\}. \tag{17}$$

The volume associated with every grain is then the sum of the volumes of its quadrons

$$V_g = \frac{1}{2} Tr\left\{\tilde{C}_g\right\} = \frac{1}{2} Tr\left\{\sum_{q \in g} \tilde{C}_q\right\}. \tag{18}$$

In fact, the volume of any region Γ within the granular system, irrespective of how large, is simply the superposition of the quadron volumes that it comprises, $V_\Gamma = \sum_{g \in \Gamma} V_g = \sum_{q \in \Gamma} V_q$. The structure tensor of Γ is $\tilde{C}_\Gamma = \sum_{g \in \Gamma} \tilde{C}_g = \sum_{g \in \Gamma} \sum_{q \in g} \tilde{C}_q = \sum_{q \in \Gamma} \tilde{C}_q$.

Quantitative local structural description in 3D

To construct the 3D quadrons, we follow the same procedure. The granular space is first tessellated into basic volume elements, which we also call quadrons. This is done using the procedure shown in Fig. 4 and explained in great detail in [20]. The 3D quadrons are generically non-convex octahedra, except when a grain has only three contacts, in which case a quadron degenerates into a hexahedron. The structure of every quadron is then quantified by the following 3D shape tensor

$$\tilde{C} = (\xi \times \mathbf{r}) \otimes \mathbf{R} \tag{19}$$

where the vectors ξ, $\mathbf{r}$ and $\mathbf{R}$ are shown in Fig. 4, $\xi \times \mathbf{r}$ is a cross product and $\otimes$ in (19) denotes an outer product. We have seen in 2D that grains with two contacts give rise to degenerate quadrons. Such a degeneracy occurs in general dimension d, whenever

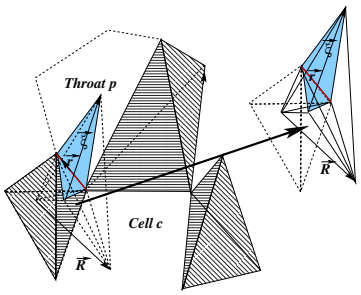

Fig. 4. The quadron description in 3D [20]: unless the grain is under-coordinated with fewer than $d+1$ contacts in d dimensions, the quadron is generically a non-convex octahedron. The quadron's shape is characterised by the structure tensor (19).

a grain has fewer contacts than $d+1$. In 3D this means that grains that have only two or three contact points would give rise to quadrons that are not octahedra but rather either hexahedra (when the number of contacts is 3) or triangles of zero volume when they have only two contacts. Nevertheless, the definition of $\tilde{C}$ remains unambiguous even in these special cases. The 3D volume of the quadron has a similar expression to the 2D version (17),

$$V = \frac{1}{3!} Tr\left\{\tilde{C}\right\}. \tag{20}$$

Advantages of the quadron description

Evidently, it is possible to tessellate the granular space with other methods, the most common of which are the various Voronoi and Delaunay tessellations [26–28]. However, the quadron description has a number of advantages over the traditional methods. The first advantage is that it makes it possible to characterise the basic volume elements non-arbitrarily and unambiguously by the local tensors C. The ambiguity in Voronoi-based methods is that each volume element is a different type of polytope. For example, it would be difficult to quantify the shape and volume of a collection of octahedra, hexahedra, dodecahedra, etc., with one structure tensor without introducing arbitrariness into the definition. In contrast, *all* quadrons, whether degenerate or not, can be described with C. This, however, is also achieved by certain Delaunay and the Bagi tessellation [26–28]. The second advantage is that, unlike conventional methods, the quadrons construction is based on the network of intergranular contacts. Consequently, it takes account of the physical connectivity of the assembly. In contrast, most Voronoi-based tessellations are based only on the proximity of grains. This aspect is essential to theories that aim to model physical properties that depend on the connectivity. Examples are fluid transport through the porous granular material, adsorption, catalysis and stress transmission. To our knowledge, no other tessellation has this advantage combined with the first advantage above. The third advantage is that the quadrons are the natural "quasi-particles" of the GSM. This is because there are fewer grains than structural DFs in the system, but there are more than enough quadrons. To this end, the shape tensor provides an explicit form for the volume function in the partition function

$$W_d = \frac{1}{d!} \sum_{all\ q} Tr\left\{C_d'^q\right\} \tag{21}$$

where $d = 2, 3$ denotes the dimensionality of the system. This aspect and its usefulness to the GSM will be discussed further below. Another advantage is that the tensor C provides a common description to GSM and mechanics of granular materials. It was shown that, at least in $2D$, the shape tensor C plays a key role in a first-principles theory of stress transmission in isostatic granular materials. Specifically, its components couple to the continuous stress field, providing a structure-stress constitutive relation that closes the stress field equations [18].

It should be commented that the quadron method describes only the topology of the granular structure, namely, the connectivity of the graph made by the contact points. This is only one aspect of the structure; another aspect involves all the possible shapes of grains that can be fitted into a given contacts graph. The shapes of the grains can be characterised by an *independent* set of parameters, which we call the geometric DFs. Consequently, the phase space of structural DFs consists of two almost entirely independent subspaces – the topological and the geometric. In the following we focus only on the topological ensemble, but the inclusion of the geometric one is straightforward and will be discussed elsewhere.

4 Quadron statistical mechanics

The topological degrees of freedom

The topological DFs (TDFs) are the independent spatial variables that determine topology, or connectivity, of the packing. To identify these DFs we consider the inter-granular point contacts. We stress again that reducing the contacts to points does not affect the generality of the analysis – in assemblies of compliant grains these points are the centroids of the contact surfaces. Significantly, it also does not affect the application to stressed assemblies, as shown in [29] The volume of the granular assembly is related to the topology, or connectivity, of the structure, i.e. the inter-granular contact positions.

The choice of the statistical ensemble is the first step to doing statistical mechanics and any ensemble is defined by the constraints on it. For the purpose of this discussion, the ensemble consists of all the possible configurations that a given collection of N ($\gg$1) grains can pack into under specified boundary compressive forces. The systems are postulated to be in mechanical equilibrium under these boundary loads. By a given collection of grains, we mean given distributions of grain shapes and sizes, as well as given physical properties of each and every one of them. Examples of physical properties are their inter-granular friction coefficients, their elastic moduli, etc. These properties need not be the same for all grains within a specific system, but all the systems in the ensemble involve exactly the same collection of grains. Specifying the frictional properties is important in that it affects the final structure to some extent (although not some universal properties of the structure topology, as was shown in [30,31]). Another important constraint is that all the members of the ensemble are generated by an identical packing procedure. This is important because the packing process affects the structural organisation and different generation processes lead to different structural statistical characteristics. Finally, to focus the discussion on the canonical ensemble, we constrain all the systems to have the same mean coordination number $\bar{z}$. We comment in passing that relaxing this constraint would necessitate using a grand-canonical ensemble.

Let us consider the topology of the connectivity network. Since N is very large we neglect boundary effects. Alternatively, we can either consider in the following analysis the assembly to live on a $d + 1$ hypersphere or impose periodic boundary

conditions to eliminate boundary corrections. Fundamentally, the TDFs are the position vectors from an arbitrary origin to every inter-granular contact point, $\rho_{gg'}$. Having constrained the mean coordination number to $\bar{z}$, the number of contacts is $N_{cont} = N\bar{z}/2$ and the number of independent variables is therefore $N\bar{z}d/2$. Rigid translation and rotation of the packing do not affect its statistics. With d axes of translation and $d(d-1)/2$ axes of rotation in d dimensions, the number of TDFs in d dimensions is

$$N_{tdf} = \frac{N\bar{z}d}{2} - \frac{d(d+1)}{2} \approx \frac{N\bar{z}d}{2}. \tag{22}$$

Since the vectors $\rho_{gg'}$ depend on the origin, it is more convenient to consider the differences between nearest neighbour $\rho_{gg'}$'s, specifically, the vectors $\mathbf{r}_q$, described in Sect. 3, which extend between neighbouring grain contacts. These vectors have many more components than DFs – twice as many in $2D$ and three to five times as many in $3D$. Of these one can choose many sets of independent vectors as the TDFs. The only requirement on each such choice is that the vectors form a "spanning tree" on the connectivity network [32]. We observe that N_{tdf} is either equal to (in $2D$) or smaller than (in $3D$) the number of quadrons and that it is always *larger* than the number of grains. We will return to the significance of this observation below.

To identify the natural quasi-particles of the volume ensemble we first need to return to the partition function (8). A more in the community is treating the grain volumes as the structural DFs (SDF). Doing this raises a conceptual problem

Quadrons – the quasi-particles of the volume ensemble

We recall that in thermal statistical mechanics there are also particles, or quasi-particles, whose energies are determined by the state of their degrees of freedom. Examples of such entities could be real particles possessing potential, interaction and kinetic energies, or they could be energy-carrying quasi-particles – spins, frequencies, photons, phonons and a variety of excitons. The question is what plays the role of the quasi-particles in GSM. One could argue that the natural choice would be the grains. This is based mainly on the fact that the grains are the smallest moving entities and Newton's conditions of mechanical equilibrium is applied to each individual grain. However, in view of the above discussion we see that this is somewhat misguided. The main property that the fundamental quasi-particles should possess is volume, since dynamics is immaterial to the partition function of static packs. Since grain volumes are made of the volumes of smaller volume elements – the quadrons – then it has been proposed that these are more fundamental and fit the bill better [20,34]. The relation of the quadrons to the grains parallels the relations of the quarks to elementary particles and the relations of internal DFs to larger-scale integrated DFs.

Furthermore, to clinch the identity of the natural quasi-particles of the volume ensemble, let us consider again the partition function (8). This reveals an even more important misconception, i.e. that the grain volumes themselves can be regarded as the structural DFs (SDF). This view is conceptually problematic since the number of grains is *smaller* than the number of SDF we have identified above, $N < N_{SDF} = N\bar{z}d/2$. In contrast, there are sufficiently many quadrons. As shown above, in 2D, their number equals exactly that of the SDF and in 3D $N_q > N_{SDF}$. In 2D this means that we can integrate directly on their volumes, albeit with inclusion of an analogue of the "density of states" (see below). In 3D this means that we can pick and choose a subset of the quadrons to be the quasi-particles of the GSM description. The partition function can then be written as an integral over this subset, expressing the other quadrons in terms of the independent ones. This, in turn, leads us to the realisation that correlations between quadrons must be taken into consideration.

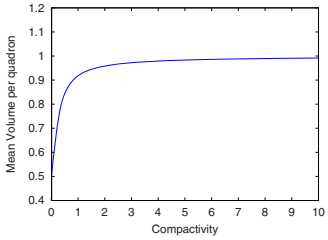

Fig. 5. The mean volume of the ideal quadron gas model as a function of compactivity. Both V_0 and X_0 are measured in units of Δ. As the compactivity increases, the mean volume per grain undergoes a transition at $X_0 = \Delta$ from $V_0 - \Delta$ to V_0.

Calculations with the volume ensemble

To illustrate the use of quadron volumes as SDF, let us consider the following simple model in 2D – the ideal quadron gas approximation [19]. In this model, as in the traditional ideal gas of molecules, the quadrons are considered independent. Since the number of quadrons, $N\bar{z}$, is the same as the number of SDF, we can write the partition function as

$$Z_v = \left[\int e^{-V/X_0} g(V) dV\right]^{N\bar{z}}. \tag{23}$$

The function $g(V)$ is an analogue of the density of states, namely, it is the frequency of having quadrons with volumes between V and $V + dV$. We suppose a model where the quadrons can take any value between, $V_0 + \Delta$ and $V_0 - \Delta(\Delta > 0)$. The density of states is then $1/2\Delta$ between these two values and zero otherwise. This partition function can be computed exactly

$$Z_v = \left[\frac{X_0 \sinh(\Delta/X_0) e^{-V_0/X_0}}{\Delta}\right]^{N\bar{z}}. \tag{24}$$

The mean volume per quadron is

$$\langle V \rangle = -\frac{1}{N\bar{z}} \frac{\partial \ln Z_v}{\partial(1/X_0)} = V_0 + X_0 - \Delta \coth(\Delta/X_0) \tag{25}$$

(see Fig. 5). As the compactivity increases so does the mean volume, making a transition from $V_0 - \Delta$ at low X_0 to V_0 at high X_0 around $X_0 = \Delta$. The variance of the volume fluctuations per grain can also be calculated exactly,

$$\langle \delta^2 V \rangle = \frac{1}{N} \frac{\partial^2 \ln Z_v}{\partial(1/X_0)^2} = X_0^2 - \frac{\Delta^2}{\sinh^2(\Delta/X_0)}. \tag{26}$$

The variance also shows a transition at $X_0 = \Delta$ (Fig. 6).

Expanding the phase space with the stress ensemble

In the mid 2000's, it was proposed that granular entropy consists not only of the configurational disorder but also of the disorder in the different stress states [14,16,20]. This suggestion was later supported by numerical tests [14,15]. The reason is that the application of external forces on a system's boundary grains cannot be

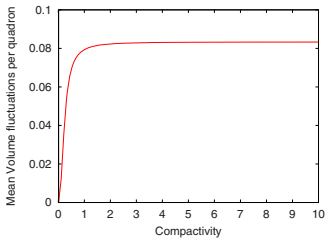

Fig. 6. The mean volume fluctuations of the ideal quadron gas model as a function of compactivity. Both V_0 and X_0 are measured in units of Δ. As the compactivity increases, the fluctuations increase, undergoing a transition at $X_0 = \Delta$.

controlled precisely but only the global boundary stresses can be imposed. Considering only compressive boundary loads in the absence of body forces, the boundary stress, $\Sigma_{\alpha\beta}$ is an average of the total α force component on the boundary, per unit surface area in the β direction. For example, the normal stress on a flat boundary with normal in the n direction is Σ_{nn}, which is the total force applied on it in the n direction. The boundary loading is determined by a collection of single forces applied to the boundary grains. Specifying the boundary loading precisely, requires specifying the position of each of the boundary grains and the external forces applied to them.

There are astronomically many combinations of such grain forces, each potentially leading to a different configurational rearrangement near the boundary. Since stresses in GS are not uniform and since individual force chains emanate exactly from the load points at the boundary then each boundary forces configuration leads to a different stress chain network and hence to a different stress state. It is the statistics of these stress states that the stress ensemble captures. The disorder in the stress micro-states adds to the structural disorder and the stress entropy is the logarithm of the number of such stress micro-states.

We outline the stress ensemble in isostatic systems. The static determinacy of such systems and the linearity of the isostaticity theory means that the number of stress micro-states is exactly the same as the number of possible combinations of individual forces on the boundary. If M is the number of boundary force sources at any one configuration then these forces $\mathbf{g}_m$, $(m = 1, 2, \ldots, M)$ are the independent DFs of the stress ensembles. The inter-granular forces cannot be the DFs because depend linearly on the boundary forces by Newton's equations.

In 2D, the partition function of the stress ensemble is

$$Z_f = \int e^{-\sum_{ij} \frac{1}{X_{ij}} \mathcal{F}_{ij}} \prod_{m=1}^{M} d^M \mathbf{g}_m. \tag{27}$$

Here the indices i, j run over the Cartesian components x, y and $\mathcal{F}_{ij}$ are the components of the force moment function, from which the stress σ_{ij} is derived,

$$\mathcal{F} = \sum_g V_g \sigma_g = \sum_{gg'} \rho_{gg'} \otimes \mathbf{F}_{gg'}. \tag{28}$$

The sum runs over pairs of grains in contact gg', $\mathbf{F}_{gg'}$ is the force that g' applies to g, $\rho_{gg'}$ is the position vector of the contact point between these grains, measured from the centroid of grain g and V_g is the volume associated with grain g, i.e. the sum of the quadron volumes associated with grain g. The tensorial parameter $X_{ij} = \partial \mathcal{F}_{ij} / \partial S$ has been named "angoricity" by Edwards and Blumenfeld [16] and it is the analogue of the temperature and of the compactivity [33,34]. S here is the entire entropy associated with both the volume and the stress ensembles.

Recently, it has been argued that the stress and the volume ensembles are inter-dependent [17]. This corrected a previous misconception in the literature that the stress and the volume ensembles are independent, which had given rise to results obtained from using each of these ensemble alone. The conclusion of inter-dependence was based on three arguments. Firstly, the volume ensemble alone does not capture all the entropy of mechanically stable granular systems because it is presumed to comprise all the possible structural arrangements under a set of identical boundary forces. Yet, no experiment on a collection of many grains can reproduce the same precise forces on every boundary grain – only boundary stresses can be controlled. This means that the statistics of the boundary forces must be taken into consideration. Secondly, the stress ensemble alone certainly does not capture the entire entropy of mechanically stable granular systems because changes in the boundary loading forces, however small, change the internal structure. This means that one cannot consider an ensemble of boundary forces with a fixed structure. Thirdly, the stress ensemble depends on the force moment function $\mathcal{F}$, which in turn depends on the structural DFS. This means that the stress and structural partition functions cannot be calculated independently and $Z \neq Z_v Z_f$. This is shown explicitly below.

The implication of these arguments, which hold in any dimension, is that the phase space is made of both the structural and stress DFs. We illustrate this issue in 2D for simplicity, following the work in [17]. We start by considering the ensemble of all 2D N-grain systems ($N \gg 1$), with each system constrained to be prepared by the same process, to be in mechanical equilibrium under M external compressive forces, acting on the boundary grains, and to have the same mean contact number $\bar{z}$.

For this ensemble, the combined partition function is conveniently expressed in terms of two generalised vectors:

$$\mathcal{R} \equiv \left(r_{1x}, r_{2x}, \ldots, r_{N\bar{z}/2x}, r_{1y}, r_{2y}, \ldots, r_{N\bar{z}/2y}\right)$$
$$\mathcal{G} \equiv \left(g_{1x}, g_{2x}, ..., g_{Mx}, r_{1y}, r_{2y}, ..., r_{My}\right)$$

where r_{ni} is the ith component ($i = x, y$) of the nth vector $\mathbf{r}_n$ and similarly for the boundary forces $\mathbf{g}_m$. In terms of these, the volume function is quadratic, $W = \frac{1}{2}\mathcal{R} \cdot A \cdot \mathcal{R}$, where p, q run over quadrons, i, j run over vector components x, y and A is a matrix of rational fractions [17]. The partition function is then

$$Z = \int e^{-\frac{1}{2X_0}\mathcal{R}\cdot A\cdot\mathcal{R} - \mathcal{G}\cdot B\cdot\mathcal{R}} d^{N\bar{z}}\mathcal{R} d^{2M}\mathcal{G}. \tag{29}$$

The matrix B both couples the boundary forces to the loop forces [18] and in inversely linear in the angoricity components [17]. Again, it is the occurrence of the vector $\mathcal{R}$ in both the volume and the force moment functions that couples the volume and the stress phase sub-spaces.

Calculations of expectation values with the combined partition function can be carried out straightforwardly in 2D due to the quadratic form of the exponential in (29) [17]. The mean volume is

$$\langle V \rangle = \frac{N\bar{z} + 2M}{2} X_0. \tag{30}$$

This result assumes that A is not singular, an assumption that has to be carefully checked [35]. Expression (30) is in fact an equipartition principle, analogous to the well known thermal one, $\langle E \rangle = k_B T/2$ per DF. It also highlights the fact that the total number of DFs is $N\bar{z} + 2M$, which includes both the structural and stress DFs. An intriguing interpretation of this result is that the compactivity in itself is not the conjugate variable of the volume function, nor is the angoricity the conjugate of the

force moment function. Rather, the coupling between the two ensemble means that there is a complicated combination of the compactivity and angoricity variables, which is the conjugate of a combined function of the volume and force moment functions. The search for these combination is currently ongoing. One can similarly calculate expectation values that relate to the boundary forces and therefore the boundary stresses. It is this type of calculations that leads to derivation of equations of state [36], but these calculations will not be given here.

Conclusion and discussion

To conclude, the entropy-based granular statistical mechanics of static granular matter is still in the process of development. Although aspects of this formalism have been supported many issues remain open. The absence of ergodicity is particularly handicapping, which makes it difficult to study dynamics with results from the static ensemble. We have pointed out some misconceptions in the field, such as mistaking the grain volumes for degrees of freedom and derivation of results from either the volume or the stress ensembles independently. We have shown here that the quadrons are much better candidates as the quasi-particles and that the two ensembles are inherently coupled.

The results reviewed here highlight the enormous potential of the granular statistical mechanical approach. In particular, a consistent formulation can lead directly to calculations of equations of state, which are much needed in the field of granular matter. In principle, our 2D calculations can be extended straightforwardly to 3D. However, deriving closed form relations in 3D is not as easy as in 2D. This is because the volume function is cubic in the structural DFs, limiting much of the analysis to numerical evaluation of the expectation values. Nevertheless, it would be useful to carry out these calculations and compare with experimental measurements.

This also means that the choice of the ensemble needs to be carefully considered. An ensemble is characterised by its constraints and these constraints must correspond to the modelled experimental system. For example, the ensemble may have either a fixed or a fluctuating number of grains. Even when the number of grains is fixed, it can have either fixed or fluctuating number of quadrons, corresponding to a fixed or fluctuating coordination number $\bar{z}$. The ensemble can also have a given grain size and shape distributions. In particular, one of the most significant constraints, which applies to most of the ensembles we are interested in, is that all the systems should be prepared by the same procedure.

Finally, statistical mechanics of granular matter is far from complete at this stage, making this field an active and exciting research frontier. Much more work is required to bring this field to maturity and to realisation of its potential. This can be done only with a focused cooperative theoretical-computational-experimental effort.

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