

# Phases of granular matter

Charles Radin<sup>1</sup> and Harry L. Swinney<sup>2</sup>

<sup>1</sup>Department of Mathematics, University of Texas at Austin, Austin, Texas 78712  
USA

<sup>2</sup>Department of Physics and Center for Nonlinear Dynamics, University of Texas at  
Austin, Austin, Texas 78712 USA

E-mail: radin@math.utexas.edu and swinney@chaos.utexas.edu

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**Abstract.** An understanding of homogeneous nucleation of crystalline structure from a disordered medium such as a liquid remains an important unsolved problem in condensed matter physics. Guided by the results from a number of experiments on granular and colloidal systems in the past two decades, including in particular observations of homogeneous nucleation in colloidal and granular systems, we suggest an alternative to the statistical mechanics approach to static granular matter initiated by Edwards and Oakeshott in 1989.

## 1. Introduction

An important objective for a theoretical model of static, bulk, granular matter is to understand the unusual mechanical properties of the material. For instance, how should one understand why a very heavy vehicle can stand on, or even move slowly over, a deep bed of dry sand? This question was raised many years ago for materials composed of many molecules, where heavy weights can be supported by ice but not by water; matter made of many  $H_2O$  molecules may or may not support a heavy weight. Equilibrium statistical mechanics used the notions of fluid and solid phases to sharpen the question, and centered on the relevance of temperature and pressure.

In 1989 Edwards and Oakeshott [1] developed a theoretical approach to the understanding of granular matter that was inspired by earlier modeling by Flory (see [2]), de Gennes (see [3]), and Edwards (see Doi and Edwards [4]) and others, who adapted the ensemble formalism of equilibrium statistical mechanics to model ‘soft’ *nonequilibrium* matter such as a polymer chain of many segments in solution. One simplification in the adaptation for polymers is to downplay or ignore the need to justify the appropriateness of using ensemble averaging, which in statistical mechanics is based on the dynamics of the particles. Edwards and Oakeshott went further and applied the approach to static granular matter such as bulk sand, using microcanonical and canonical ensembles based on packings of nonoverlapping *stationary* spheres, mechanically stable under gravity and the contact forces among themselves and their container.

A recent comprehensive review by Baule et al. [5] focuses on the statistical model introduced by Edwards and Oakeshott and then generalized. The review did not address the nucleation of grains into macroscopic granular crystals, as observed in a recent experiment [6], which a theory of granular matter should accommodate, if not predict. The recent results on homogeneous nucleation lead us to propose a modification of the Edwards approach, focusing on physical granular matter subject to gravity and a certain form of external energy - a ‘heat bath’ - and we discuss how these ingredients can be used to give a coherent view of a range of granular experiments which are not addressed in the Edwards approach.

## 2. Random close packing

The configuration of grains in physical granular matter is generally disordered but difficult to characterize in detail. In 1960 G.D. Scott [7] conducted an experiment on ball bearings poured repeatedly into a container, and he found that the volume fraction  $\phi$  occupied by the spheres ranged from about 0.60 up to a maximum of 0.64, which he conjectured to be a fundamental upper limit, now called the random close packed (RCP) volume fraction,  $\phi_{RCP}$ . The RCP volume fraction is much smaller than the maximal volume fraction occupied by a collection of identical spheres, which is the volume fraction of hexagonal close packed (HCP) or face centered cubic (FCC) crystalline arrays,  $\phi = \pi/\sqrt{18} \approx 0.74$  [8].

On the theoretical side, in 1959 J.D. Bernal [9] had already conjectured that “there is an absolute impossibility of forming a homogeneous assembly of points of volume intermediate between those of long-range order and closest packed disorder”. There is an unstated probabilistic or entropy qualification, so to be more precise the claim is that, in the sense of the Law of Large Numbers, there is a volume fraction (now called RCP) such that ‘most’ homogeneous configurations of ball bearings with fixed volume fraction below RCP are disordered and most homogeneous configurations with fixed volume fraction above RCP have long range order.

In the half century following Bernal and Scott’s introduction of the RCP concept of a fundamental limit in the volume fraction, many experiments have added increasing support for a fundamental limit in  $\phi$  (see, e.g., [10-14]). A recent example is an experiment by Rietz et al. (2018) [6] on a granular system with an imposed oscillating horizontal shear of small amplitude and low frequency. This experiment revealed a gradual increase in  $\phi$  until a plateau at  $\phi = 0.645$  was reached after 20000 shear cycles, as illustrated in Fig. 1(a). The long plateau persisted for 50000 shear cycles, and then the global volume fraction began to slowly increase as a polycrystalline state developed.

An earlier experiment by Huerta et al. [15] also imposed a small amplitude horizontal oscillation but at a much higher frequency. The Huerta et al. and Rietz et al. experiments can both be thought of as granular systems in contact with a heat bath, where the frequency of oscillation is analogous to temperature. In this terminology Huerta et al. used a bath at high temperature, 50 Hz, while Rietz et al. used a bath

at low temperature, 0.5 Hz. The two experiments can be interpreted as indicating a fluid/solid phase transition with decreasing temperature, with the RCP volume fraction representing the nucleation barrier at the transition: the need for crystalline clusters to reach a critical size, beyond which macroscopic crystalline matter automatically grows, as will be discussed in the Section 4.

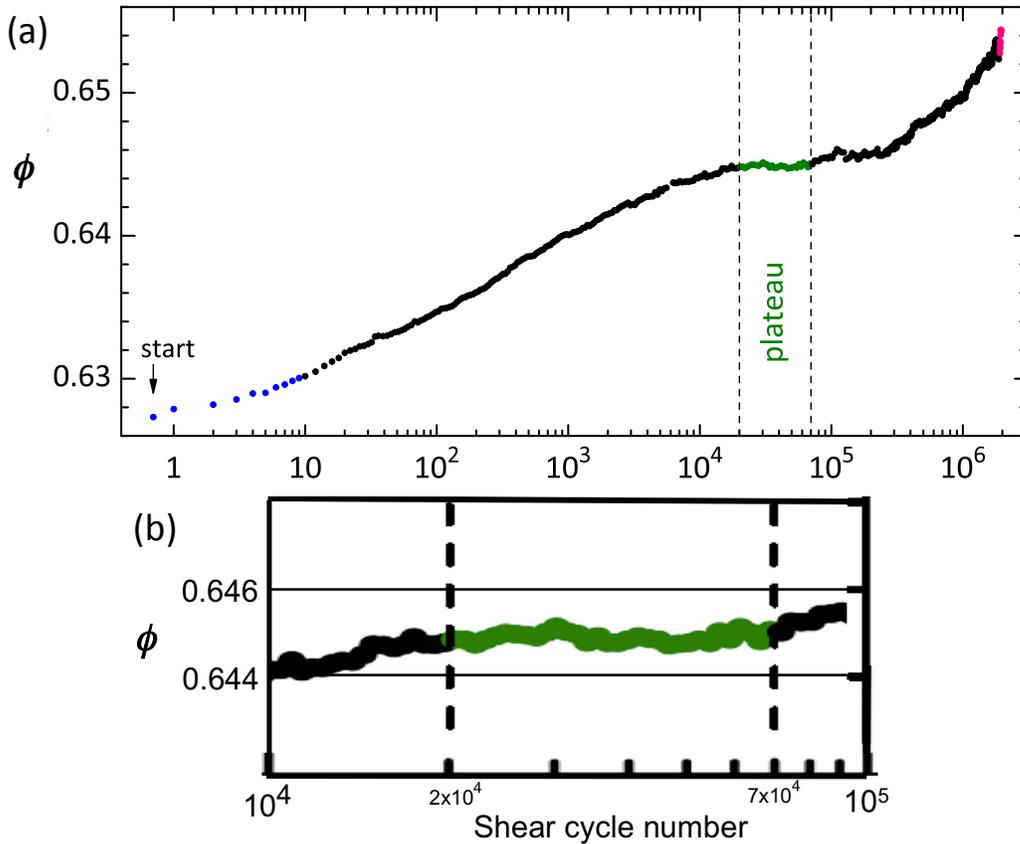


Figure 1: (a) The robustness of the RCP limit is illustrated by these data of Rietz et al. [6] for a packing of 50000 glass spheres (3.00 mm diameter) in a  $(105 \text{ mm})^3$  box with slow horizontal shear imposed by tilting two opposite sidewalls by  $\pm 0.6^\circ$  with an oscillation period of 2 s. The global volume fraction  $\phi$  gradually increases with successive shear cycles until a well-defined plateau is reached at  $\phi = 0.645$ . (b) An expanded graph of the plateau at  $\phi = 0.645$ . The plateau persists for about 50000 shear cycles, and then the volume fraction begins to slowly increase as crystallites emerge in the interior of the sample.

### 3. A proposed alternative to the Edwards approach to static granular matter

Edwards' fundamental idea of using an ensemble approach to model granular matter was very important as it suggested intuition based on materials in thermal equilibrium.

The Edwards approach has not proved to be helpful in understanding the random close packing phenomenon. A conjecture [16] that RCP might signify a sharp transition analogous to the freezing of fluids in thermal equilibrium was based on physical granular experiments showing crystalline clusters growing on side walls [17], not on calculations using an Edwards ensemble. There are now calculations suggesting that this interpretation of RCP is compatible with the Edwards model [18-19]. However, in subsections 4.1 and 4.3 it will be argued that the homogeneous nucleation observed in the experiment of Rietz et al. [6] indicates that disordered static packings of grains should be considered as supercooled or glassy rather than in equilibrium as in the Edwards approach.

We propose a theoretical approach for granular matter distinct from that of Edwards et al. but consistent with various laboratory observations. An experiment by Huerta et al. [15] revealed buoyancy behavior like that given by Archimedes' Principle for an ordinary fluid in thermal equilibrium. As mentioned, many granular experiments have demonstrated the RCP limit, a well-defined upper bound on the volume fraction of homogeneous disordered granular matter, as illustrated by Fig. 1. We interpret the experiments on buoyancy, the RCP limit, and the emergence of a crystalline state [6] as corresponding to granular matter at consecutively lower 'temperatures', illustrating a fluid/solid freezing transition. We now describe in more detail several experiments that lead us to propose this alternative approach to the Edwards model of granular matter.

### 3.1. Archimedes' law of buoyancy

Huerta et al. [15] studied a bidisperse collection of spheres, of diameters 3 and 4 mm, in a container with two opposite vertical side walls oscillating horizontally with small amplitude (1 mm) but high frequency (50 Hz) and acceleration (up to  $10g$ ). The high frequency oscillation maintained the granular array in a fluidized state. Having the opposite walls oscillate with opposite phase eliminated convection. Low density objects pulled downward into the denser granular medium by a string, and dense test objects placed on top of the granular bed (away from the boundary) were observed to experience buoyant forces like those in an ordinary fluid in thermal equilibrium. Similar behavior was produced by shearing in Nichol et al. [20].

### 3.2. Colloidal systems

Extensive experiments have been conducted using concentrated suspensions of colloidal particles in baths in thermodynamic equilibrium. Colloidal particles have been widely considered as hard spheres, but Poon et al. [21] and Royall et al. [22] explain how in reality colloidal particles are soft, as Fig. 2 illustrates.

Another experiment on which we base our theoretical approach to granular matter was conducted by Rutgers et al. [23], who studied polystyrene spheres in water and used different methods to reduce electrostatic screening in order to mimic hard spheres more closely. There was still an uncertainty in the effective size of the particles, which was

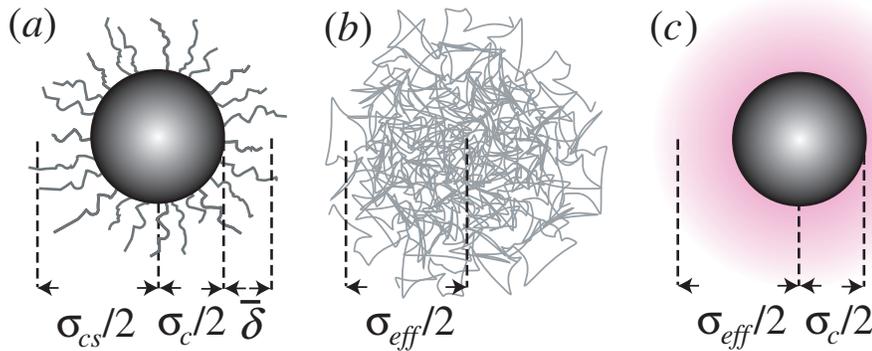


Figure 2: Schematic representations of different models of “hard-sphere” colloids (Fig. 1 in Royall et al. [22]): (a) Sterically stabilized particle with surface ‘hair’ layer of thickness  $\bar{\delta}$  and core radius  $\sigma_c/2$ . (b) Microgel particle with an effective radius  $\sigma_{eff}/2$ , formed by a heavily cross-linked polymer. (c) Charged colloid with an electrical double-layer that gives an effective radius  $\sigma_{eff}/2$ .

handled through an adjustable parameter. Figure 3 (from Fig. 2 of Rutgers et al. [23]) shows a sharp transition in density, which Rutgers et al. interpret as analogous to the fluid/crystal transition of the mathematical hard sphere model [24].

The motion of colloidal particles is Brownian (diffusive), not ballistic as assumed in the hard sphere model, but such a simplification is not unusual for modeling soft materials; indeed, the particles in the Edwards model are stationary. A more accurate model would be based on Brownian motion, but such a model would only be appropriate for a local analysis of individual particles, not of the bulk material, and would miss the bulk crystalline state shown in Rutgers et al. unless one takes into account gravity. That experiment shows that gravity produces particle assemblies with a significant vertical density gradient. Gravity and diffusive motion are both used in the model by Burdzy et al. [25], but they cannot analyze the region where Rutgers et al. see a discontinuous transition. Note that the hard sphere model employed by Rutgers et al. assumes ballistic motion but does not include gravity.

There are numerous differences between the behavior of the colloidal system in Rutgers et al. and the granular system in Rietz et al., but there is also the remarkable similarity of the phase transition exhibited in the two. Below we will exploit common elements from Huerta et al. [15], Rietz et al. [6], and Rutgers et al. [23] in developing a heat bath picture of granular matter with ‘typical’ configurations exhibiting a density gradient like those in the colloidal system of Fig. 3.

### 3.3. Dilatancy and the solidity of granular matter

The homogeneous nucleation of crystalline structure in Rietz et al. indicates the creation of a ‘solid’ state of granular matter. Crystallinity, observed for instance through X-ray diffraction for molecular systems, is a standard characteristic of many solids in thermodynamic equilibrium. (Quasicrystals are a notable exception.) However, there

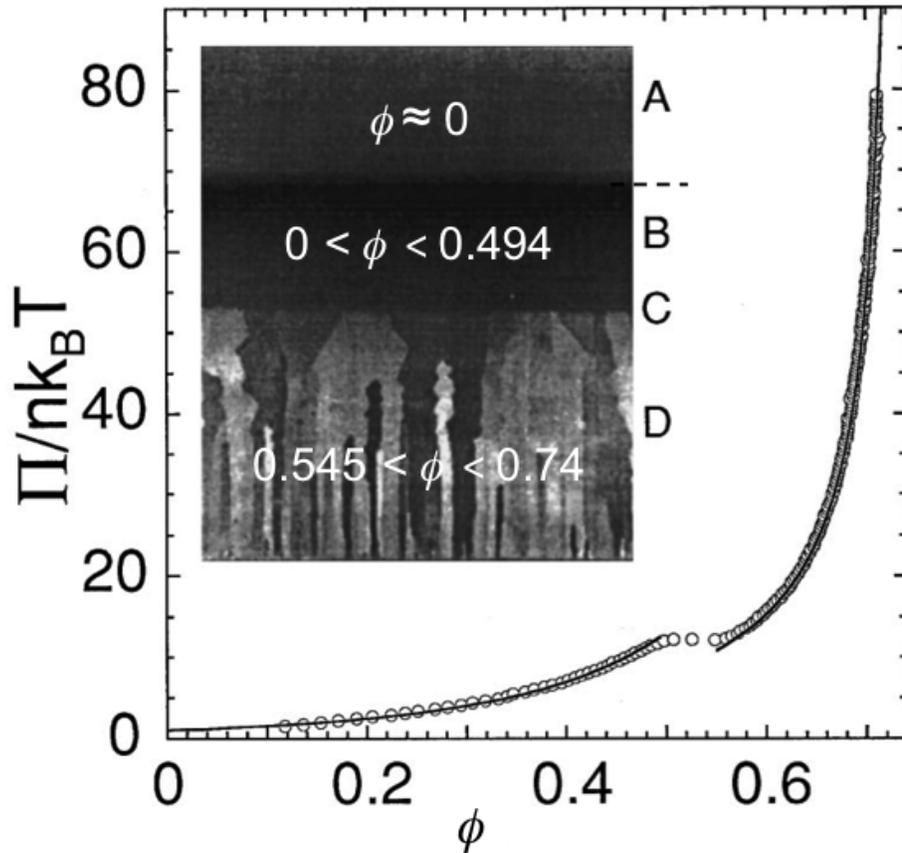


Figure 3: The equation of state for the osmotic pressure  $\Pi$  obtained by Rutgers et al. (Fig. 2 of [23]) for an isothermal system of polystyrene spheres ( $0.720 \mu\text{m}$  diameter) that was allowed to stabilize for three months before the measurements were made. The calculated equation of state, shown by a solid line, agrees well with the measurements of osmotic pressure. The inset shows 10 mm height of a sample: (A) supernatant fluid, (B) liquid phase (the dashed line indicates the faint boundary between the supernatant and the liquid phase), (C) sharp interface between the liquid phase and the phase with crystallites, and (D) phase with crystallites visible due to Bragg scattering of light.

are characteristics other than crystallinity that might provide a clear distinction between solids and fluids, such as the viscosity or the elastic shear modulus [26]. We do not use such characteristics because there have yet to be experiments on mechanical properties of ‘crystalline’ granular matter. However, there is reason to expect that such an approach should work.

In 1885 Osborne Reynolds [27] introduced ‘dilatancy’ to explain several experimental instances of the resistance of granular matter to shear. He imagined a *crystalline* arrangement of sand grains and argued that the arrangement would have to expand to be strained through a small angle, producing a strong force in reaction.

Dilatancy in a horizontally sheared granular system was demonstrated and measured by Nicolas et al. [17]. The system expanded when opposite side walls of

the container were tilted slightly from the vertical position, thus decreasing the volume fraction, as shown in Fig. 4, where compaction and dilation occurs alternately through each successive cycle of shearing. Each successive maximum in the volume fraction was higher, as some particles were pulled down by gravity into space that opened up slightly in the dilated medium during the shearing process. In this experiment the volume fraction continued increasing past the RCP limit as *inhomogeneous* nucleation of a crystal occurred at the cell walls (see Fig. 5 in [17]).

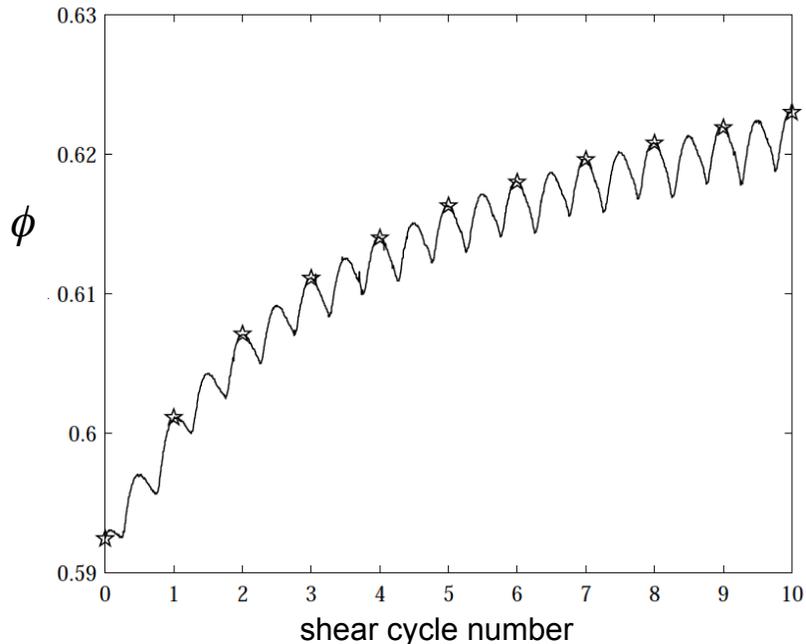


Figure 4: The volume fraction evolution observed by Nicolas et al. [17] during successive cycles of shear of 41000 randomly distributed spheres in a parallelepiped box (Fig. 2 of [17]). Shear was imposed by oscillating the tilt angle of two opposite sidewalls by  $\pm 5.4^\circ$ . The star symbols mark the volume fraction maxima in successive shear cycles as the sidewalls passed through the vertical position with the tilt angle increasing.

### 3.4. Dissipation in driven granular matter

For granular matter a comparison of experiments with simulations of molecular dynamics and granular continuum equations has revealed that dissipation through inelasticity (restitution coefficient less than unity) and sliding friction are both important in strongly driven systems, such as vertically oscillating granular layers. Extensive experiments and analyses on vertically oscillating granular layers has shown that when the downward acceleration of the container exceeds  $g$ , it is necessary to include both forms of dissipation (inelasticity and friction) to obtain agreement between observations and modeling [28-30]. It is not yet clear to what extent dissipation may be a necessary ingredient in

the qualitative near equilibrium or equilibrium behavior we wish to understand in this paper.

A simple picture of the evolution of the granular matter in Rietz et al. is that the effect of shearing is mainly to provide slow motion to the particles and break any contacts between them, which allows mobility of the particles so they can occasionally fall under gravity into holes that develop beneath them due to dilatancy (see Fig. 4). The particles in a colloid only diffuse, and the material as a whole stays somewhat localized (see Fig. 3 and [25]). In the granular experiment of Rietz et al. particles are mostly driven horizontally, and that, together with gravity, also leads to matter remaining near the floor. We will use the similarity of the colloidal experiment of Rutgers et al. and the granular experiments of Huerta et al. and Nicolas et al. to fashion a common picture based on a system of hard particles evolving under external energy input and gravity. We hope that such a picture will capture the essential qualitative features of the experiments, even ignoring dissipation.

## 4. Nucleation and glassy materials

### 4.1. Observations of homogeneous nucleation

Granular experiments with physical hard sphere packings have occasionally observed *inhomogeneous* nucleation at a bounding surface [17, 31-38], where two-dimensional layers easily form a triangular lattice that serves as the base for a three-dimension HCP/FCC cluster, like the oranges stacked in a market. In contrast, the recent experiment by Rietz et al. revealed *homogeneous* nucleation, the spontaneous creation of crystalline clusters of ‘critical size’ far from any bounding wall. An example of a growing crystallite in the granular experiment is shown in Fig. 5(a); crystallites of FCC, HCP, and mixed symmetries were observed to form and grow. Similarly, an experiment on *colloidal* particles by Gasser et al. [39] revealed homogeneous nucleation of crystallites, as illustrated by the example in Fig. 5(b). The homogeneous nucleation of crystallites is a bulk property of the granular and colloidal systems, not a consequence of an interaction of the material with its boundaries, which is a surface phenomenon not necessarily indicative of any bulk property.

Nucleation has been most thoroughly analyzed for materials in thermal equilibrium [40]. A static fluid in thermal equilibrium at fixed pressure and slowly decreasing temperature will freeze at a ‘freezing’ temperature  $T_f$  to a solid, typically a crystalline solid. With a slowly increasing temperature that solid will melt at a ‘melting’ temperature  $T_m$ . With care one finds  $T_f = T_m$ , although a fluid can easily be driven with decreasing temperature to a supercooled (metastable, nonequilibrium) state at some temperature  $T < T_f$ , where  $T$  is the container temperature. Quasistatic freezing of a fluid to its equilibrium solid state generally occurs through nucleation, but unless care is taken to avoid it, the nucleation is inhomogeneous. If such nucleation sites are absent the fluid will supercool until it nucleates spontaneously, so-called homogeneous

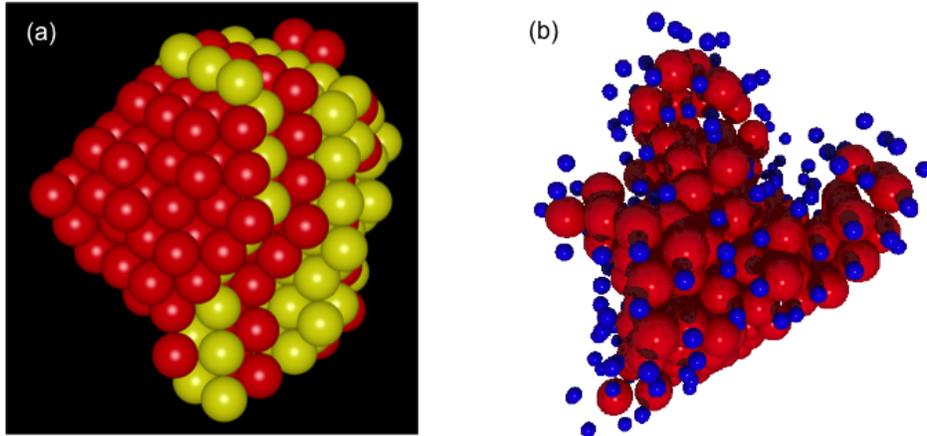


Figure 5: (a) A crystallite with 600 spheres that nucleated in the interior of a sheared granular medium (unpublished figure from the experiment reported in Rietz et al. [6]). The red spheres have FCC local symmetry, and yellow spheres have HCP local symmetry. (b) A crystallite that Gasser et al. observed to nucleate following shear melting of a colloidal system with  $\phi = 0.47$  (Fig. 3C in [39]). The 206 red particles have crystalline order and are drawn to scale, while the neighboring blue particles, reduced in size for clarity, share at least one crystal-like bond.

nucleation, at unpredictable sites. If a fluid is cooled sufficiently quickly, it will fail to nucleate at all and will enter a nonequilibrium long-lived glassy state.

Homogeneous nucleation appears in fluids in thermal equilibrium as the temperature is slowly reduced below the freezing point. Within a thermodynamic phase, mechanical and other responses of matter in thermal equilibrium can be estimated by linear response of the equilibrium state. A different approach, not necessarily part of a theory of bulk matter, is needed to model the nucleation of a new phase in response to a change of state at a phase transition.

The observation in [6] of homogeneous nucleation in static granular matter subjected to gentle cyclic shear suggests that one consider the static material to be either glassy or supercooled, but not in equilibrium.

#### 4.2. Nucleation theory

The observations of nucleation in granular and colloidal matter could perhaps provide insight into nucleation more generally. The current state of the art in nucleation theory is a collection of models called Classical Nucleation Theory (CNT) [40], which has developed over many years to describe the spontaneous development of small crystalline clusters in a fluid. In CNT there exists a ‘critical’ size of a crystalline cluster which, once surpassed, will grow to macroscopic size. Such a critical size has been observed in the recent experiments on nucleation in colloidal and granular systems, as Fig. 6 illustrates: in the colloidal system crystalline clusters with fewer than  $\sim 100$  particles shrank while larger clusters grew (Gasser et al. [39]), and in the granular experiment crystalline

clusters with fewer than  $\sim 10$  spheres shrank while larger clusters predominantly grew.

While Classical Nucleation Theory provides a qualitative picture of nucleation that is consistent with observations, it is not a fundamental theory that can be tested quantitatively in physical experiments. Perhaps detailed measurements of nucleation in colloidal and granular systems will guide the development of a quantitative theory of nucleation that can be tested in experiments on both macroscopic and microscopic systems.

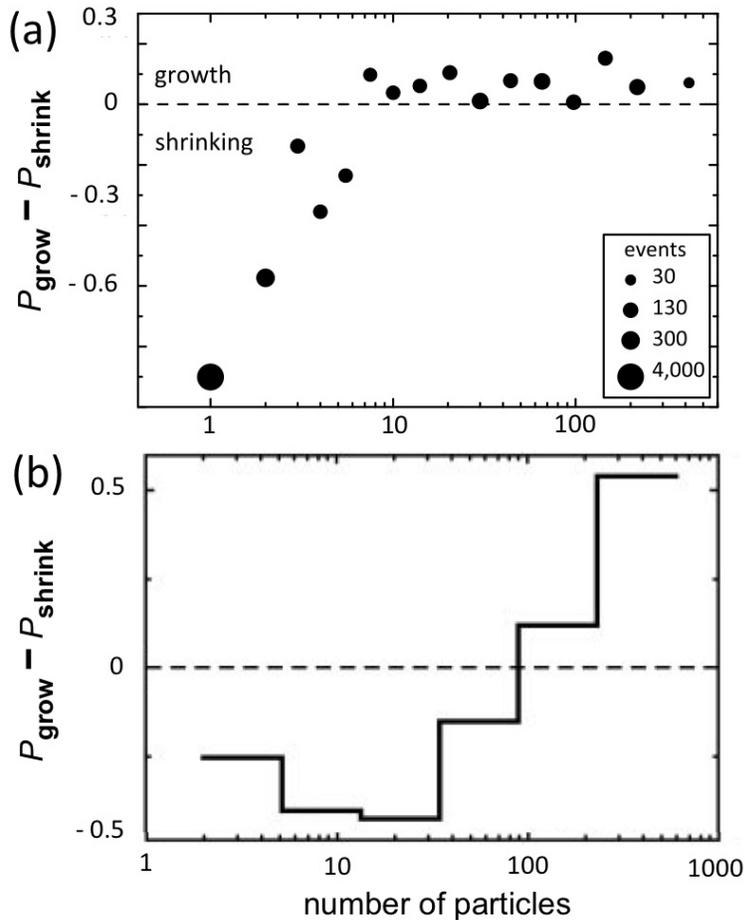


Figure 6: In both the granular and colloidal systems small crystalline clusters usually shrink while crystallites above a critical size grow, as demonstrated by these results: (a) in sheared granular matter (from Fig. 4(a) of Rietz et al. [6]), and (b) in a colloidal system with  $\phi=0.47$  (Fig. 2 of Gasser et al. [39]).

#### 4.3. Glassy states of granular matter

Nucleation is significant because of the difficulty of a (disordered) fluid to freeze to an ordered (crystalline) solid. There can also be barriers between different phases of a solid, a well known example being diamond, which is pure carbon and obviously not the equilibrium state at room temperature and pressure, which is graphite. In this sense one

could consider diamond at room temperature and pressure to be a glassy state of carbon. The stability of diamonds implies the barrier is high against nucleation to the graphite phase. Understanding such barriers in their generality is an essential part of the ‘glass problem’. One view of the granular experiment of Rietz et al. [6] is that the experiment revealed an appropriate physical protocol to ‘equilibrate’ supercooled granular matter to a crystalline state, smoothly but with an observable nucleation barrier, which turned out to explain RCP. This suggests that granular matter under cyclic shear can provide an unusual setting to help understand some of the phenomena involved in glass formation. A similar claim has been made [39] for the nucleation of hard colloids, but precision for colloids is limited by the 3-6% uncertainty in the determination of particle size, which is illustrated by the images in Fig. 2 (from [22]).

Any sufficiently large gas such as the atmosphere, or even a typical small solid such as a rock conglomerate, is heterogeneous at the largest scale, and basic physical theory is usually applied first to homogeneous parts and then extended to the heterogeneous material. In particular, we expect typical granular matter, such as a sand pile, to be heterogeneous in density (and composition). At least since the 1996 experiment by Nowak et al. [41], researchers have tried multi-axis shaking [31-33], cyclic shear [17, 34-35], continuous shear [20, 36-37] and sedimentation [38] to prepare homogeneous granular matter to which one could apply the Edwards theory, with its ensembles of *static* spheres. In contrast, when a granular material is fluidized as in [15] or [20] and the external vibration is quickly removed, the static material can be considered ‘glassy’, with mechanical properties different from, and harder to analyze than, those of the fluid. The interpretation that the static state is the result of such a quench derives from the difference of its disordered state from the granular matter at the end of the experiment of Rietz et al., at low but still positive temperature. The result of quenching from such a crystalline state would still be crystalline, and quite different from commonly observed disordered granular matter.

The above discussion on the glassy state of granular matter should be distinguished from those theories which associate a glassy state, or transition to a glassy state, in analyses of hard sphere models without the ingredient of gravity; see for example [42].

## 5. Conclusions

We have sketched a way to understand the properties of granular matter composed of grains moving in response to a certain type of heat bath, explicitly drawing the approach from physical experiments that reveal relevant properties of granular and colloidal matter. The present view uses well-defined properties of granular and colloidal matter to explain how control parameters can turn granular matter from fluid-like to solid-like behavior through a sharp phase transition. Along the way this approach interprets several experiments on RCP that are outside the purview of the Edwards model.

With this perspective on granular matter it is natural to seek an ensemble model

such as that of Edwards, from which one could in principle compute material properties. However, if the interest were ultimately in the properties of static granular matter, as in the solidity of a dry bed of sand as discussed in the Introduction, such a model would only be of use for (rare) systems with volume fraction above RCP, i.e., above  $\phi = 0.645$ . In the heat bath approach static systems with density below RCP are treated as glassy, and an ensemble model of fluidized granular matter would be of little direct use. That said, the experiment of Rietz et al. exhibits a simple physical protocol, namely, very small angle ( $0.6^\circ$ ) and very low rate (0.5 Hz) of shear perpendicular to gravity, which ‘equilibrates’ any static initial state into a crystalline state; hence high density granular matter is now a natural target for study. Aside from the inherent interest in this crystalline state of physical granular matter, this protocol would also be of potential use in investigating the general phenomenon of the nucleation of crystalline structure out of disorder.

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