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Fluctuations, correlations and transitions in granular materials: statistical mechanics for a non-conventional system

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In this work, we first review some general properties of dense granular materials. We are particularly concerned with a statistical description of these materials, and it is in this light that we briefly describe results from four representative studies. These are: experiment 1: determining local force statistics, vector forces, force distributions and correlations for static granular systems; experiment 2: characterizing the jamming transition, for a static two-dimensional system; experiment 3: characterizing plastic failure in dense granular materials; and experiment 4: a dynamical transition where the material 'freezes' in the presence of apparent heating for a sheared and shaken system.

Keywords: granular materials; jamming; disordered solids

1. Introduction

This work begins with a few basic ideas concerning the whats and whys of granular materials. In particular, we consider where granular materials and molecular matter part company, which involves open questions of relevant scales. An important point that we emphasize is that fluctuations in granular systems can be large, although their nature is not well established. Here, we present evidence for the idea that well-defined statistically stationary configurations exist. The idea is that if the external control parameters—things like stresses at the boundaries, or strain rates—are held fixed then there exist well-defined distributions for important internal variables as the granular system is taken through a set of states consistent with the external controls. The extent to which this is generally applicable is unknown. Hence, statistical characterizations of real experimental systems are very important. To the extent that distributions for inter-particle contact forces or correlations between forces, displacements or other standard variables are determined

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solely by a small set of well-defined control parameters, a statistical description should be viable. Then, the task is to determine the relevant ensembles and structures that determine the relations between control parameters and the resulting distributions.

Here, we turn to a series of experiments that emphasize the statistical properties of dense granular systems. These are: experiment 1: determining local force statistics, including vector forces, force distributions and correlations for static granular systems that are subject to simple deformations, pure shear and isotropic compression; experiment 2: characterizing the jamming transition, contact numbers and pressures near the point where a static system becomes mechanically rigid; experiment 3: plastic failure—what happens when a dense granular system is sheared to the point that particles are irreversibly displaced; experiment 4: a dynamical transition: freezing by heating in a sheared and shaken system—how does a dense system respond to different kinds of energy input, and how do these inputs allow the system to explore a phase space of different states. Each of these illustrates some of the recent issues that involve the physics of dense granular materials and, in particular, their statistical nature.

Granular materials are collections of macroscopic 'hard' (but not necessarily rigid) particles whose interactions are dissipative. The particles can be described perfectly well by classical mechanics, although this may lead to mathematical complexity if Coulomb friction between grains is involved. Interactions between grains are dissipative, so that left alone, moving granular systems come to a state of rest. If there is a steady input of energy, motion can be sustained. Such a system may resemble a molecular state. But the granular case is far from equilibrium. Despite this last property, it may be possible to draw on such thermodynamically fundamental concepts as entropy or temperature and to apply or modify these concepts to ensembles of granular particles. It is also clear that fluctuations in granular systems, be they in space or time, can be large. In the dense state, fluctuations in space are associated with force chains, filamentary structures that carry a disproportionately large fraction of the forces within the system. In time-varying systems, these chains form and break. The energy for fluctuations typically comes from the mean flow, which must be sustained by an external source.

There are many fascinating and deep statistical questions associated with granular materials. Perhaps the most important is whether there is indeed an underlying granular statistical description that has the same level of predictive power as statistical mechanics for thermal systems. There are many related questions that need to be addressed. These include: what is the nature of granular friction? What is the nature of granular fluctuations and what is their range? Is there a granular temperature? A granular entropy (Edwards & Oakeshott 1989)? Do fluctuation dissipation relations hold? How does a granular system change from flowing to mechanically rigid? This last question addresses the issue of jamming (Cates *et al.* 1998; Liu & Nagel 1998). Since many systems undergo such a transition, it seems probable that there are connections to other systems, e.g. colloids, foams, glasses, all of which exhibit jamming. Other important questions: what happens at mesoscopic scales? How do we understand granular plasticity, and is it similar to molecular plasticity?

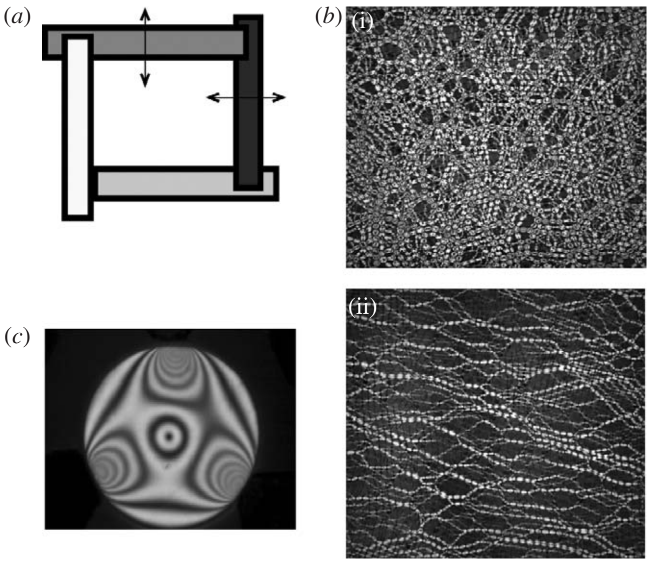

Figure 1. (*a*) Schematic of biaxial tester. Particles are confined within a rectangular enclosure, which has two independently movable walls. (*b*) Images of granular samples that have been (i) isotropically compressed and (ii) subject to pure shear. (*c*) Photoelastic image of a single disc.

The continuum limit has routinely been assumed in soil mechanics (Nedderman 1992). But there is at best a partial justification for this assumption. Engineers must design, and so need a reliable continuum description. Granular handling devices collapse at an alarming rate, and solids handling devices typically work well below design. Despite much work on continuum models for dense granular systems, current models are complex, and their underlying physical basis is still in considerable need of development. Until the underlying physics is well understood, the design of practical particulate handling devices will remain problematic.

Granular phases can be separated into relatively dilute and dense phases. The dilute phase is typically modelled by the same methods as molecular gases. Kinetic theory provides a rather good description of many phenomena in the granular gas state. Granular solids and dense granular fluids are much less well understood. For dense granular states, theory is far from settled, and under intensive debate and scrutiny. It is this set of states that is the focus of the remainder of this work.

Before turning to the experiments discussed above, we document some of the properties that are important for dense granular materials. First, forces are carried preferentially on force chains. A clear example of force chains, seen in a two-dimensional shearing experiment, has been given by Howell *et al.* (1999). These structures (shown in figure 1) indicate that multiscale phenomena are at play. In addition, as a dense granular system is deformed, force chains form and break, leading to large fluctuations in force. These fluctuations are intrinsically of a spatio-temporal character, as seen for instance in the large stress fluctuations in three-dimensional shear flow reported by Miller *et al.* (1996). For real granular materials, friction and extra contacts, beyond what is needed for mechanical stability, are important issues. Typically, due to these effects, preparation history matters. Indeed, owing to these issues, in most cases, a statistical approach may be the only possible description.

Before turning to our discussion of experiments, we note some interesting approaches and concepts. These include the idea of jamming for behaviour near the solid–fluid transition. This has been discussed by Cates *et al.* (1998), Liu & Nagel (1998), O'Hern *et al.* (2003), Donev *et al.* (2005) and Henkes & Chakraborty (2005) among others. Connections to plasticity in disordered solids have been considered by Falk & Langer (1998), Lemaître (2002) and Maloney & Lemaître (2004). Granular 'elasticity' has been extensively discussed, and here we note several relevant works by Bouchaud *et al.* (1995), Goldenberg & Goldhirsch (2005) and Tighe *et al.* (2005).

2. Experiment 1: determining force statistics

We now turn to an overview of several experiments, beginning with the experiment 1: determining force statistics (see Majmudar & Behringer 2005). Here, the goal is to characterize granular force statistics and correlations. The basic experimental set-up involves two-dimensional particles, discs, that are confined to a biaxial test apparatus (as sketched in figure 1*a*). This apparatus allows us to control the boundaries very precisely and, therefore, to prepare states that have a well-known state of deformation. The experiments use photoelastic particles which allow us to determine forces between the grains. The basic technique involves several parts: we obtain images with and without polarizers and use the second set of images to obtain particle centres and contacts. Using images of individual particles obtained with polarizers, we invoke an exact mathematical solution of stresses within a disc subject to localized forces along its circumference. Specifically, we make a nonlinear fit to photoelastic pattern, whose intensity is given by $I = I_0 \sin 2[(\sigma_2 - \sigma_1)CT/\lambda]$, using the known elastic solution; the contact forces are then the fit parameters. Here, σ_1 and σ_2 are the principal stresses within the disc, C is a material-dependent parameter, T is the thickness of the discs and λ is the wavelength of light. In the previous step, we invoke force and torque balance on each particle. Newton's third law, which requires equal and opposite forces on the two different particles at a contact, provides error checking. In figure 2 we give examples of experimental and 'fitted' images; in general, the agreement is very good.

Several results from these studies are interesting. First, we consider the force distributions, which depend on the preparation history of the sample. Specifically, we contrast inter-particle contact force data for pure shear and isotropic compression in figure 3, where we show the normal and tangential (frictional) forces separately. Notably, only for the case of pure shear do we see an exponential tail at large forces. We note that these results are consistent with Edwards entropy-inspired models for $P(f)$ by Snoeijer *et al.* (2004) and Tighe *et al.* (2005) who consider the effect of anisotropic loading on the force distribution.

Addressing the issue of spatial correlations is also important. In figure 4, we show force–force correlation functions computed for two independent directions. In other words, we maintain directional information in the

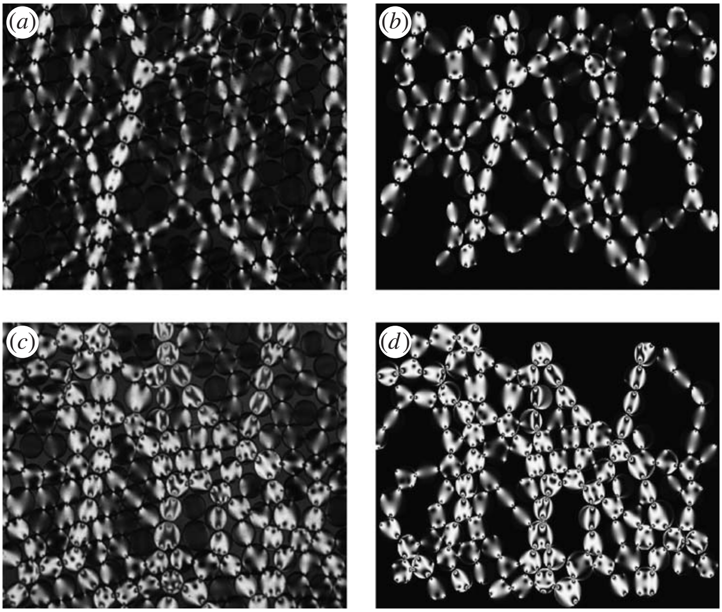

Figure 2. Comparison between (a,c) two experimental photoelastic images and (b,d) the corresponding images, that are computed based on fits to forces at the particle contacts. (a,b) Correspond to a case of pure shear and (c,d) correspond to isotropic compression.

correlation function, rather than averaging over all angles. In the isotropic compressional case, the correlation function is independent of orientation and falls rapidly to zero in a few particle diameters. By contrast, for the case of pure shear, starting from an uncompressed state, we see roughly power-law correlation up to the range of the calculation (approx. $20D$) along the strong force chain direction, and short-range correlation in the direction transverse to the force chains.

3. Experiment 2: the jamming transition

Experiment 2 provides a characterization of the jamming transition. In these experiments, we once again use the two-dimensional biaxial system and slowly move through a range of packings (for solid area fractions of $\phi \simeq 0.84$), where our two-dimensional system just becomes mechanically rigid. Here, one expects from several simulations and models that: (i) the contact number, Z, will increase (nominally discontinuously) at a critical packing fraction ϕ_c, (ii) above the jamming point, Z will continue to increase as a power-law in $\phi - \phi_c$, and (iii) the pressure will also increase as a power-law in $\phi - \phi_c$ above ϕ_c. In results to be detailed elsewhere (Majmudar *et al.* 2007), we do indeed find a rapid increase in Z near $\phi = 0.84$. Above this point, Z continues to increase as a power-law with exponent about 0.55, and the pressure also rises as a power-law with an exponent of about 1.1. These results are consistent with recent simulations by O'Hern *et al.* (2003) and Donev *et al.* (2005) as well as with a novel mean-field model proposed by Henkes & Chakraborty (2005).

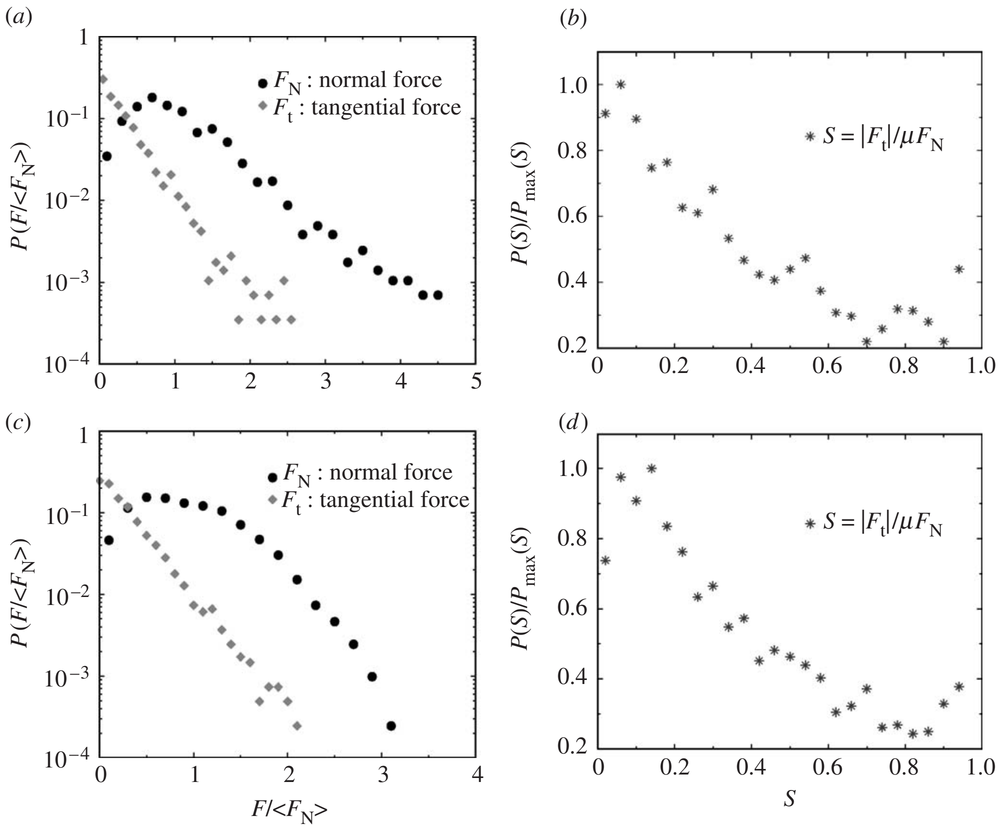

Figure 3. Distributions for contact forces and the mobilization of friction, defined as the ratio $S=|F_t|/\mu F_N$, where F_t is the tangential or frictional force, F_N is the normal force and μ is the inter-particle coefficient of static friction. (*a*,*b*) A system that has been subject to pure shear from an initial state where the particles are just in contact. (*c*,*d*) A similar initial state that has then been subject to isotropic compression. The deformation in a given direction is $\varepsilon = \Delta L/L$. For the pure shear case, the x and y deformations are equal but opposite and have magnitude 0.042. For the compressional case, they are equal and have the value 0.016.

4. Experiment 3: plastic failure

Experiment 3 involves the characterization of plastic failure as a granular sample is subjected to increasing shear. The basic question is: what is the nature of microscopic deformation (plasticity)? This process is described classically for granular materials by (continuum) soil mechanics models. An interesting recent proposition is that the microscopic nature of plastic behaviour in granular materials mimics that seen in other disordered jammed materials (Lemaître 2002; Maloney & Lemaître 2004), such as metallic glasses. For instance, we might expect models, such as shear transformation zone pictures, developed for molecular plasticity to apply to granular plasticity. This type of model, which has been explored recently by Falk & Langer (1998) and developed more fully by Lemaître (2002) for granular-like systems, seeks to

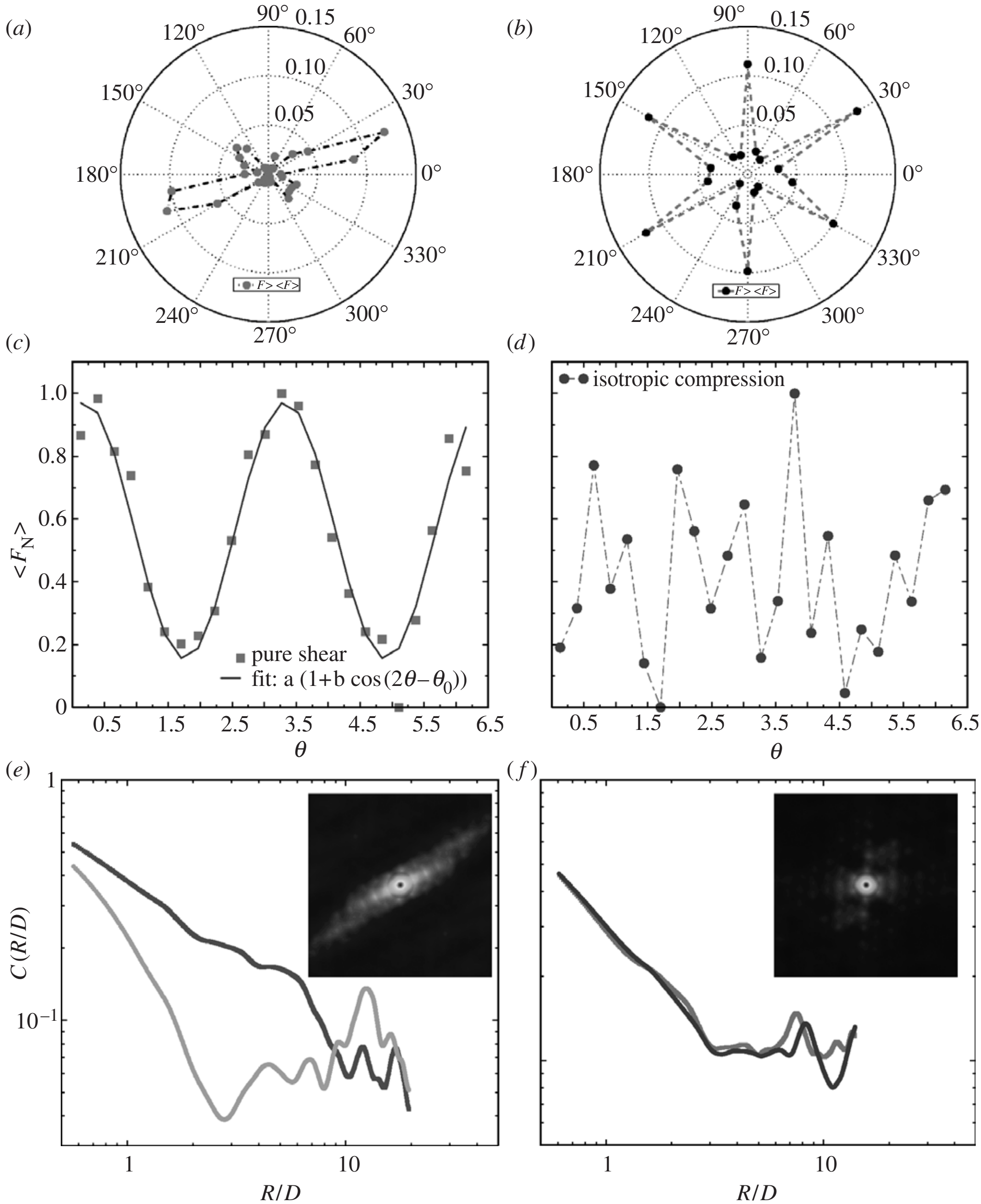

Figure 4. Contact data corresponding to figure 3. (a,b) The angular distribution of contacts for particles carrying forces above the mean, for respectively pure shear and isotropic compression. (c,d) The mean normal force as a function of angle, for pure shear and isotropic compression, respectively. (e,f) Correlation functions of the force for pure shear and isotropic compression, respectively. The two lines in each sub-figure correspond to orthogonal orientations of the relative displacement vector for calculating the correlation function. (e) The dark curve is along the strong force chain direction, and the light curve is normal to that direction. The insets in (e,f) give a greyscale three-dimensional representation of the correlation function. Here, bright corresponds to large values of the correlation function.

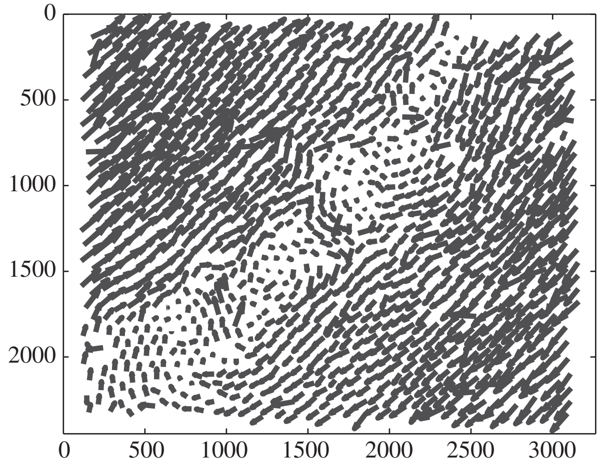

Figure 5. Particle displacement vectors following a small shear deformation of the biaxial experiment.

understand the role of non-affine particle displacements, i.e. the extent to which local micro- or mesoscopic displacements depart from a smooth local transformation. We are currently testing to see whether this type of picture applies to granular systems. At this point, we note some preliminary results, although the story is not yet complete.

In an initial set of measurements, we consider the local displacements and stress changes that occur as our two-dimensional photoelastic granular system is sheared first in a forward and then in a reverse direction. Figure 5 shows representative displacement fields for the particles as the result of small differential shearing. It is immediately obvious that the deformation occurs in a central shear band, in which there are relatively large-scale vortex-like structures. An inspection of the stress–strain curves (figure 6) shows that there are large jumps in stress at localized plastic events.

5. Experiment 4: freezing by heating

The final experiment that we consider here involves a dynamical study of order and disorder in a dense granular system. Results for these experiments have been presented in Daniels & Behringer (2005, 2006). These experiments explore concepts such as temperature and heating in the dense granular state by considering the competition between shearing and vibration. The basic configuration of the experiment is given in figure 7. A layer of polypropylene spherical particles of diameter $D = 2.34 \pm 0.05$ mm is contained in an annular channel. The particles are subject to vibration from below, which is a nominal source of heating, and they are sheared from above by a roughened ring that is rotating at an angular rate, Ω. The strength of the shaking is characterized by the dimensionless acceleration, $\Gamma = A\omega^2/g$, where A is half the peak-to-peak shaker amplitude, ω is the (angular) vibration frequency and g is the acceleration of gravity. Ordinarily, we think of vibratory motion as leading

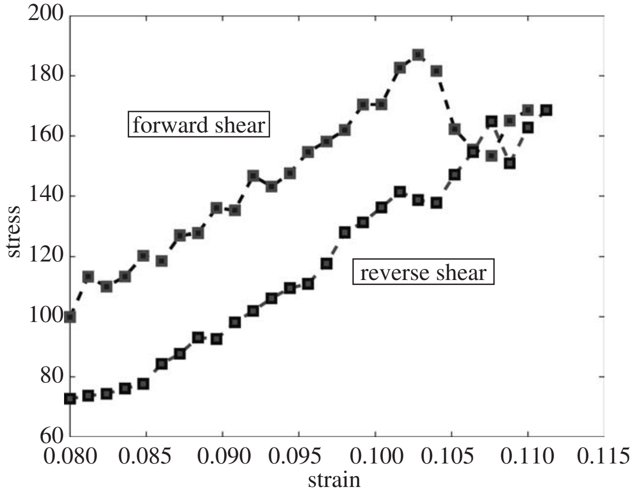

Figure 6. Mean pressure in the biaxial experiment as a function of shear strain, $\varepsilon = \Delta L/L$. The system is deformed from $\varepsilon = 0$ to a value slightly greater than 0.11, and then cycled back to a net strain of $\varepsilon = 0.08$. Note the stress drops at local failure events, and the overall stress drop after the system has been returned to $\varepsilon = 0.08$.

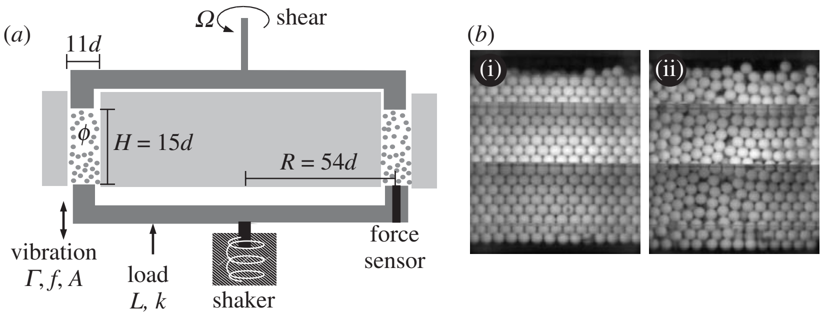

Figure 7. (a) Schematic of combined shear and vibration apparatus. Particles contained in an annulus are sheared from above and vibrated from below. See main text for parameter definitions. (b(i)) Ordered and (b(ii)) disordered states as seen from the outside of the shear apparatus.

to disorder. But in this system, it does something else: it provides a mechanism for the system to explore a broader range of possible states, i.e. packings, than might otherwise occur. In other words, if the system were left undisturbed, it would obviously remain in whatever configuration formed when the particles were put into the container. However, the applied vibration, if it is not too strong, provides enough energy for the particles to explore ‘nearby’ states, i.e. states with slightly different packings. Only for rather vigorous shaking would the vibration be strong enough to lead to disorder. One way to estimate the Γ for which vibrational disorder, e.g. vibrationally induced melting, would occur, is to set the force provided on a particle at the bottom of the layer by shaking equal to the force needed to overcome the overload of particles on top. This leads to an estimate of $\Gamma \simeq H/D$, where H is the height of the layer. In the

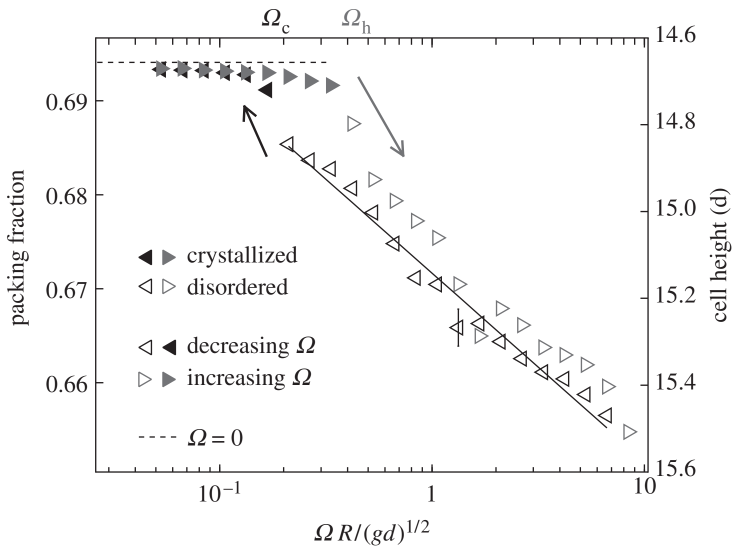

Figure 8. Phase diagram for the combined shaking/shearing system. For a given vibratory strength, the system exists in a three-dimensional crystalline ordered state (with a small number of defects) for low shear rate, Ω, and in a disordered state for high Ω.

experiments described here, $H/D \simeq 20$, which is below any of the Γs that are explored in the present experiments. By contrast, shearing of dense granular materials tends to lead to disorder, although a notable exception is contained in recent studies by Tsai & Gollub (2004). As a result, there is a competition between disordering from the shearing, and ordering which is enhanced by vibration.

Specifically, there is a hysteretic order–disorder transition, which we document in figure 8. For a non-zero Γ, the low-Ω states are ordered three-dimensional crystals (with a handful of defects), whereas the high-Ω states are disordered throughout the system. Typical states, as seen from the outside of the annulus, are shown in figure 7. The force measured at the bottom of the layer (figure 9) shows several interesting features. For ordered states, there is a two-peaked structure that reflects the sinusoidal oscillations of the base. Thus, the force is varying sinusoidally at the shaker frequency, and the probability distribution of a sinusoid is two peaked. For the disordered higher Ω states, the probability distribution function (PDF) for the force shows a roughly exponential tail, as well as a roll-off at low forces. A particularly interesting aspect of the distribution in this latter case is that the length of the tail stretches out as the transition region is approached from above. This behaviour is reflected in the moments of the force PDF. In this case, the kurtosis is a particularly good example. Here, there is a sharp cusp at the transition point. This same cusp is present in the variance of the height fluctuations. Such fluctuations would be given as a second derivative of the entropy in an appropriate Edwards entropy picture. Hearkening back to traditional phase transitions in molecular matter, it is the second derivatives of an appropriate thermodynamic potential that are singular at ordinary phase transitions.

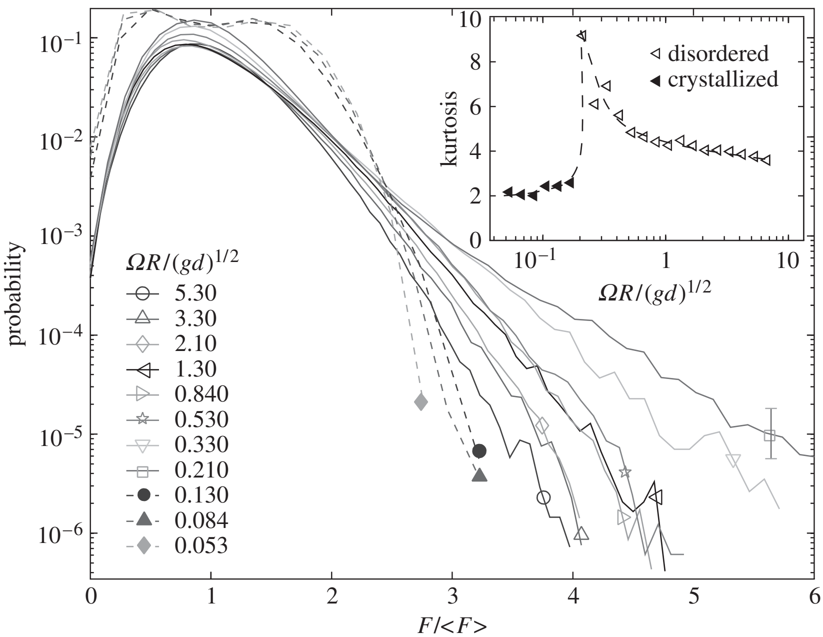

Figure 9. Probability distributions for the force, measured at the bottom of the layer for the indicated shear rates, Ω. Two-peaked distributions pertain to the ordered state, and single-peaked distributions to the disordered state. The tail of the distributions stretches out at the transition point. This feature leads to cusps in various moments for the force distribution, such as the kurtosis, which is given in the inset.

6. Conclusions

To sum up, dense granular systems exhibit rich structures, and in particular, fluctuations in such systems can be large. It is our contention that to provide a basic description of granular matter, one should take a statistical approach. Here, we have worked towards developing a statistical description by obtaining simple measures such as contact force distributions, and force correlations in static and highly controlled experimental systems. For systems near jamming, we find general agreement with theoretical predictions: a jump in contact number at the transition, and power-law variation of the mean contact number, Z, and the pressure as the packing fraction, ϕ, increases above its critical value. We have explored measures of elastic (not discussed here, for space reasons) and plastic deformation. In regard to plastic failure, we have obtained initial results characterizing failure at a microscopic level, where we see a characteristic vortex-like pattern in the displacement field. We have also pursued a dynamic transition between ordered/disordered states in a sheared and shaken system. This system shows a striking ordering of the system under shaking and moderate shear. We understand this transition as being induced by the ability of the system to find a preferred state, which is ordered, and hence densely packed, due to the presence of vibration. This state is not usually accessible to the system if the particles are simply poured into the container. It is interesting to speculate about what drives this transition, i.e. what roles are played by energetic and entropic effects in terms of the system's ability to find this state.

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References

Bouchaud, J.-P., Cates, M. & Claudin, P. 1995 Elasticity stress distribution in granular media and nonlinear wave equation. *J. Phys. I* **5**, 639–656. (doi:10.1051/jp1:1995157)

Cates, M., Wittmer, J., Bouchaud, J.-P. & Claudin, P. 1998 Jamming, force chains and fragile matter. *Phys. Rev. Lett.* **81**, 1841–1844. (doi:10.1103/PhysRevLett.81.1841)

Daniels, K. & Behringer, R. 2005 Hysteresis and competition between disorder and crystallization in sheared and vibrated granular flow. *Phys. Rev. Lett.* **94**, 168001. (doi:10.1103/PhysRevLett.94.168001)

Daniels, K. & Behringer, R. 2006 Characterization of a freezing/melting transition in a vibrated and sheared granular medium. *J. Stat. Mech.* **7**, P07018. (doi:10.1088/1742-5468/2006/07/P07018)

Donev, A., Torquato, S. & Stillinger, F. 2005 Pair correlation function characteristics of nearly jammed disordered and ordered hard-sphere packings. *Phys. Rev. E* **71**, 011105. (doi:10.1103/PhysRevE.71.011105)

Edwards, S. & Oakeshott, R. 1989 Theory of powders. *Physica A* **157**, 1080–1090. (doi:10.1016/0378-4371(89)90034-4)

Falk, M. & Langer, J. 1998 Dynamics of viscoplastic deformation in amorphous solids. *Phys. Rev. E* **57**, 7192–7205. (doi:10.1103/PhysRevE.57.7192)

Goldenberg, C. & Goldhirsch, I. 2005 Friction enhances elasticity in granular solids. *Nature* **435**, 188–191. (doi:10.1038/nature03497)

Henkes, S. & Chakraborty, B. 2005 Jamming as a critical phenomenen: a field theory of zero-temperature grain packings. *Phys. Rev. Lett.* **95**, 198002. (doi:10.1103/PhysRevLett.95.198002)

Howell, D., Veje, C. & Behringer, R. 1999 Fluctuations in a 2D granular Couette experiment: a critical transition. *Phys. Rev. Lett.* **82**, 5241–5244. (doi:10.1103/PhysRevLett.82.5241)

Lemaître, A. 2002 Origin of a repose angle: kinetics of rearrangements for granular materials. *Phys. Rev. Lett.* **89**, 064303. (doi:10.1103/PhysRevLett.89.064303)

Liu, A. J. & Nagel, S. R. 1998 Nonlinear dynamics. Jamming is just not cool any more. *Nature* **396**, 21–22. (doi:10.1038/23819)

Majmudar, T. & Behringer, R. 2005 Contact force measurements and stress-induced anisotropy in granular materials. *Nature* **435**, 1079–1082. (doi:10.1038/nature03805)

Majmudar, T. S., Sperl, M., Luding, S. & Behringer, R. P. 2007 Jamming transition in granular systems. *Phys. Rev. Lett.* **98**, 058001. (doi:10.1103/PhysRevLett.98.058001)

Maloney, C. & Lemaître, A. 2004 Universal breakdown of elasticity at the onset of material failure. *Phys. Rev. Lett.* **93**, 195501. (doi:10.1103/PhysRevLett.93.195501)

Miller, B., O'Hern, C. & Behringer, R. 1996 Stress fluctuations for continuously sheared granular materials. *Phys. Rev. Lett.* **77**, 3110–3113. (doi:10.1103/PhysRevLett.77.3110)

Nedderman, R. 1992 *Statics and kinematics of granular materials*. Cambridge, UK: Cambridge University Press.

O'Hern, C., Slibert, L., Liu, A. & Nagel, S. 2003 Jamming at zero temperature and zero applied stress: the epitome of disorder. *Phys. Rev. E* **68**, 011306. (doi:10.1103/PhysRevE.68.011306)

Snoeijer, J., Vlugt, J., van Hecke, M. & van Saarloos, W. 2004 Force network ensemble: a new approach to static granular matter. *Phys. Rev. Lett.* **92**, 054302. (doi:10.1103/PhysRevLett.92.054302)

Tighe, B., Socolar, J., Schaeffer, D., Mitchener, W. & Huber, M. 2005 Force distributions in a hexagonal lattice of rigid bars. *Phys. Rev. E* **72**, 031306. (doi:10.1103/PhysRevE.72.031306)

Tsai, J. & Gollub, J. 2004 Slowly sheared dense granular flows: crystallization and nonunique final states. *Phys. Rev. E* **70**, 031303. (doi:10.1103/PhysRevE.70.031303)