Jammed Systems in Slow Flow Need a New Statistical Mechanics
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Jammed systems in slow flow need a new statistical mechanics

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The slow dynamics of granular flow is studied as an extension of static granular problems, which, as a consequence of shaking or related regimes, can be studied by the methods of statistical mechanics. For packed (i.e. 'jammed'), hard and rough objects, kinetic energy is a minor and ignorable quantity, as is strain. Hence, in the static case, the stress equations need supplementing by 'missing equations' depending solely on configurations. These are in the literature; this paper extends the equilibrium studies to slow dynamics, claiming that the strain rate (which is a consequence of flow, not of elastic strain) takes the place of stress, and as before, the analogue of Stokes's equation has to be supplemented by new 'missing equations' which are derived and which depend only on configurations.

Keywords: jamming; forces; granular; dynamics; colloids

1. The problem

The crucial granular system concerns packed, hard and rough objects: packed means no kinetic problems, hard means no elastic problems and rough means all motion is confined to sliding and rolling. Such a problem, where grains are squeezed out to the right of the granular ensemble depicted in figure 1, has no relationship whatsoever to the kinetic theory of gases. There can be transport equations, but kinetic energy is an irrelevant concept. A prelude to dynamics is statics, and granular statics involves two basic problems: packing and stressing. A granular system can have different densities according to its creation, and the application of external forces causes forces to exist between the grains, but does not cause motion until friction thresholds are overcome.

It has been suggested (Mehta 1993; Liu & Nagel 2001; Edwards & Oakeshott 1989) that in appropriate circumstances the central concept of statistical mechanics, namely entropy, can have validity, and experiments show this is indeed the case (Makse & Kurchan 2002; Behringer 2002; Kurchan 2001; Coniglio *et al.* 2001). The Chicago experiments by Nowak *et al.* (1997) show that initial voids are removed by tapping columns of (spherical) grains with a fixed number of taps of increasing magnitude and take one into the low-density packed limit, $\phi = 0.59$, but repeating the tapping and instead decreasing the magnitude now takes one to the maximum density, $\phi = 0.64$. Thereafter the curve is reversible and confirms an ergodic condition, or at least we believe it does.

One contribution of 14 to a Discussion Meeting 'Slow dynamics in soft matter'.

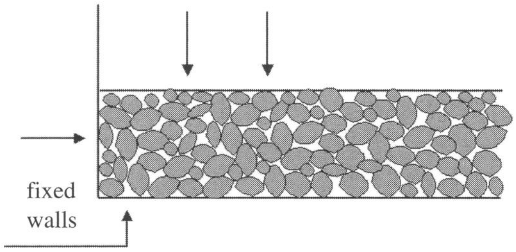

Figure 1. A model problem.

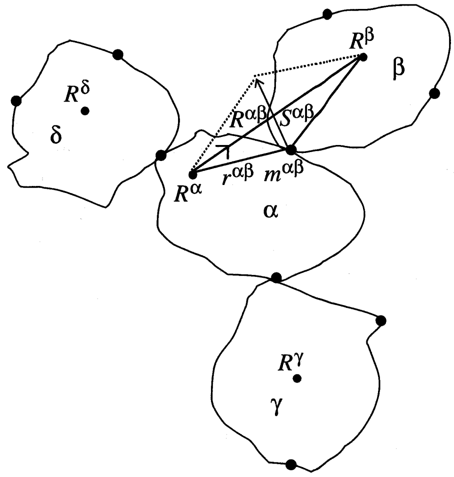

Figure 2. Two-dimensional schematic of configuration tensors in a packed granular assembly. Grains are labelled α–δ.

For infinitely hard bodies there is no enthalpy, and external forces have no effect on distribution functions. This means that if P is the probability distribution of configurations and of forces it must separate into two parts

$$P = P_\mathrm{c} P_\mathrm{f}, \tag{1.1}$$

where 'c' denotes the configurations and 'f' forces. We now derive P_c and P_f.

2. Configurations and entropy

The granular analogue of the Gibbs distribution, $\delta(E - H)\mathrm{e}^{-S/kT}$, can only depend on V and N and must be

$$S = \log \int \delta(V - W)\Theta \, \mathrm{d(all)}, \tag{2.1}$$

where V is the volume, W is the volume expressed as a function of all the coordinates specifying the N particles, i.e. their contacts and orientations, and Θ is a function that guarantees that each grain is in contact with sufficient neighbours to jam the system (Newton's equations show this to be three contacts in two dimensions and four contacts in three dimensions).

From the microcanonical equation (2.1) we can go to the canonical equation by introducing an analogue of free energy, Y, such that

$$e^{-Y/X} = \int e^{-W/X} \Theta \, d(\text{all}) \tag{2.2}$$

with $X = \delta V/\delta S$ so that $V = Y - X \mathrm{d}Y/\mathrm{d}X$.

We call X the compactivity (the analogue of temperature), and Y the effective volume. From these expressions one can mimic the statistical mechanical theories of miscibility of different powders, and the variation in volume mentioned in § 1. There are also successful extensions of the jamming concept to problems which do have internal energy (see Makse & Kurchan 2002).

In two dimensions, an exact theory of W has been given by Ball & Blumenfeld (2002), but we continue with a cruder theory wherein a 'one-particle' theory is offered, knowing the success of conventional statistical mechanics when much can be achieved with a Hamiltonian, $\mathcal{H} = \sum \mathcal{H}^{\alpha}$, which is additive over particles. To do this we need a series of configuration tensors as seen in figure 2, defined below.

With contact points $m^{\alpha\beta}$ and centroids $R^{\alpha} = (1/Z) \sum_{\beta} m^{\alpha\beta}$, we define

$$\begin{aligned} r^{\alpha\beta} &= m^{\alpha\beta} - R^{\alpha}, & F_{ij}^{\alpha} &= \sum_{\beta} R_i^{\alpha\beta} R_j^{\alpha\beta}, \\ R^{\alpha\beta} &= r^{\alpha\beta} - r^{\beta\alpha}, & G_{ij}^{\alpha} &= \sum_{\beta} \tfrac{1}{2}(R_i^{\alpha\beta} S_j^{\alpha\beta} + R_j^{\alpha\beta} S_i^{\alpha\beta}), \\ S^{\alpha\beta} &= r^{\alpha\beta} + r^{\beta\alpha}, & H_{ij}^{\alpha} &= \sum_{\beta} S_i^{\alpha\beta} S_j^{\alpha\beta}. \end{aligned}$$

An approximation to the volume is

$$W = \sum W^{\alpha} = 2\sqrt{\det F^{\alpha}}.$$

This W^{α} is the ratio of area to volume produced by grain α with its nearest neighbour, but it does not accommodate complex topologies (which Ball & Blumenfeld's formulae can). A crude theory (which has some justification, but we do not go into this here) is

$$e^{-Y/X} = \left(\int_{V_0}^{V_1} e^{-W/X} \, dW \right)^N,$$

which gives

$$V = \frac{V_0(V_0 - V_1) - X(V_0 + V_1)}{V_0 - V_1 - 2X} \tag{2.3}$$

as a good approximation, exact at $X = 0$ and $X = \infty$. Note that the minimum volume is V_0, and the maximum $\frac{1}{2}(V_0 + V_1)$, as is to be expected.

3. Forces and stresses

First we introduce the force moment (with $f_i^{\alpha\beta}$ an intergrain force)

$$\sigma_{ij}^{\alpha} = \frac{1}{2} \sum_{\beta} (f_i^{\alpha\beta} r_j^{\alpha\beta} + f_j^{\alpha\beta} r_i^{\alpha\beta}), \tag{3.1}$$

whose macroscopic average will be the stress tensor $\sigma_{ij}(r)$. Newton's laws give (for a grain of mass m in gravity g_i)

$$\sum_\beta f_i^{\alpha\beta} = mg_i^\alpha, \qquad \sum_\beta \epsilon_{ijk} f_j^{\alpha\beta} r_k^{\alpha\beta} = 0, \qquad f_i^{\alpha\beta} + f_i^{\beta\alpha} = 0,$$

which can be solved to give (Edwards & Grinev 1998, 1999)

$$\frac{\partial \sigma_{ij}}{\partial r_j} = \rho g \delta_{ix} + \text{local corrections}, \tag{3.2}$$

where ρ is the density and gravity acts along $+x$, and

$$(\sigma_{xx} - \sigma_{yy}) \cos\phi = 2 \sin\phi \sigma_{xy}, \tag{3.3}$$

where ϕ is an angle which is random in homogeneous conditions, but which is the angle of repose in, for example, a sandpile (Wittmer *et al.* 1996; Edwards & Oakeshott 1989; Edwards & Mounfield 1996). Equation (3.2) alone is inadequate to solve the statics problem and is normally supplemented by an elastic constitutive equation. Here, though, the 'missing equation', equation (3.3), depends purely on geometry. It can be written approximately in terms of our configuration tensors as (Edwards & Grinev 1998)

$$\begin{vmatrix} \sigma_{xx} & F_{xx} & G_{xx} \\ \sigma_{xy} & F_{xy} & G_{xy} \\ \sigma_{yy} & F_{yy} & G_{yy} \end{vmatrix} = 0 \tag{3.4}$$

or exactly in terms of certain matrices $x^{\alpha\beta}$, $y^{\alpha\beta}$ in $\alpha, \beta, \dots$ space as (Edwards & Grinev 1999)

$$\begin{aligned} 2\sigma_{xy}^\alpha &= y^{\alpha\gamma}[x^{-1}]^{\gamma\beta}\sigma_{xx}^\beta + x^{\alpha\gamma}[y^{-1}]^{\gamma\beta}\sigma_{yy}^\beta \\ &\cong y^{\alpha\gamma}[x^{-1}]^{\gamma\alpha}\sigma_{xx}^\alpha + x^{\alpha\gamma}[y^{-1}]^{\gamma\alpha}\sigma_{yy}^\alpha. \end{aligned}$$

The simplest homogeneous form is a uniform powder under gravity for which the missing equation gives average values of

$$\sigma_{xx} = \sigma_{yy}, \qquad \sigma_{xy} = 0, \qquad \sigma_{xx} = \rho g x = \sigma_{yy},$$

which is the hydrostatic pressure. Thus we have found that P_c is $P_c(F, r)$, and the whole distribution is, in a schematic notation,

$$\mathcal{N}[\delta(V - W)\Theta]\Big[\delta\Big(\sigma - \sum fr\Big)\Big]\Big[\delta\Big(\sum f - g\Big)\delta\Big(\sum f \times r\Big)\Big]. \tag{3.5}$$

Here $\mathcal{N}$ is a normalization and the four δ-functions represent in turn the constraints that the volume W of the system is equal to V; the stress obeys equation (3.1); the forces on a grain balance gravity; and the torques on a grain sum to zero.

4. Forces in granular materials

Although σ_{ij}^α can be averaged into a mean stress tensor, which does lead to important experimental findings, stress is not helpful at the grain level; it is the forces at the surface that matter, and they can be measured.

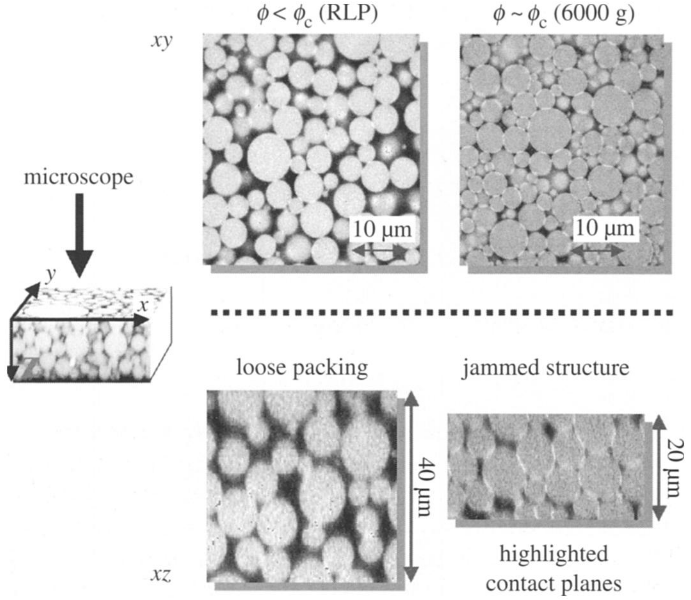

Figure 3. Confocal microscopy images of the emulsion system under gravity and after centrifugation. Upper images are in the horizontal (x, y)-plane; lower images are reconstructions of a slice in x, z.

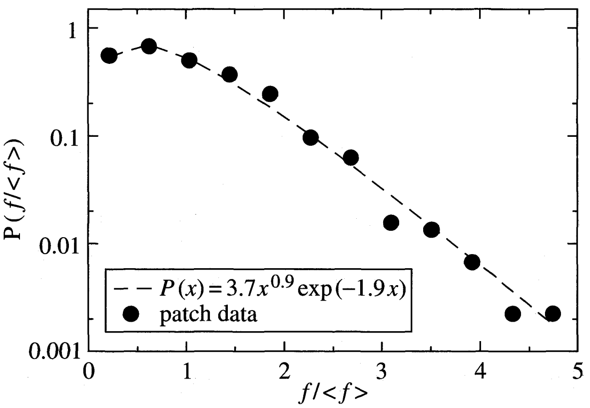

Figure 4. Probability distribution of contact forces in the compressed emulsion system (see figure 3).

Experiments at the Cavendish Laboratory by Brujić *et al.* (2003) used confocal microscopy to image a packing of emulsion particles which have been compressed by ultracentrifugation. In figure 3 the particles are shown in their initial packing under gravity (below the random-close-packing regime) and after compression, i.e. at the onset of particle deformation away from spherical. We have found the refractive index matched combination of the continuous and the dispersed phase appropriate for use in the confocal microscope. Due to the increased fluorescence of the deformed surfaces of particles in contact, the interparticle forces could be extracted using the Princen (1983) model. The probability distribution of the forces is presented in figure 4 and

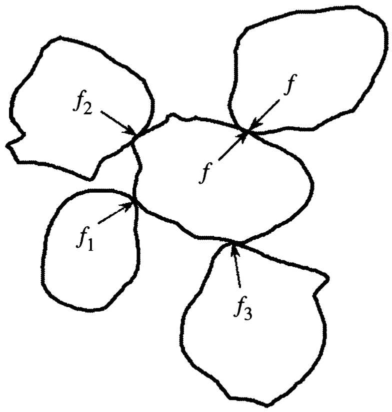

Figure 5. Two-dimensional schematic of contact forces between packed grains.

is consistent with previous studies by Liu *et al.* (1995) which find an exponential distribution of forces.

Recent simulations also support this picture. The geometric complexity is such that a full statistical 'transport' equation is very complex (Bouchaud *et al.* 2001), but it is possible to produce a simple but convincing equation for the distribution (Edwards & Grinev 2003). To be simple one wants a probability in one variable alone. A suitable variable is the magnitude of the force between two grains (see figure 5).

Ignoring gravity we find the relation

$$\boldsymbol{f} = -(\boldsymbol{f}_1 + \boldsymbol{f}_2 + \boldsymbol{f}_3)$$

and consider the scalar f given by

$$f = |\boldsymbol{f}| = \boldsymbol{f} \cdot \frac{\boldsymbol{f}}{|\boldsymbol{f}|}.$$

We now look at the components of $\boldsymbol{f}_1$, $\boldsymbol{f}_2$, $\boldsymbol{f}_3$ in the direction of $\boldsymbol{f}$, and denote these by $\lambda_1 f_1$, etc. We now argue that the probability of finding f, $P(f)$, is related by Newton's second law to the other active forces, hence (in two dimensions for simplicity)

$$P(f) = \int P(f_1, \lambda_1) P(f_2, \lambda_2) \tau(\lambda_1, \lambda_2) \delta(f - \lambda_1 f_1 - \lambda_2 f_2) \, \mathrm{d}\lambda_1 \, \mathrm{d}\lambda_2 \, \mathrm{d}f_1 \, \mathrm{d}f_2,$$

where τ is the weight factor which contains angles and does not allow grains 1, 2 and the grain studied to overlap. The factor τ is a tiresomely complicated function, and its behaviour is well modelled by just integrating λ_1, λ_2 between 0 and 1:

$$P(f) = \int_0^1 \mathrm{d}\lambda_1 \int_0^1 \mathrm{d}\lambda_2 \int_0^\infty \mathrm{d}f_1 \, P(f_1) \int_0^\infty \mathrm{d}f_2 \, P(f_2) \delta(f - \lambda_1 f_1 - \lambda_2 f_2).$$

This equation can be solved analytically by Fourier transform and gives

$$P(f) = \frac{f}{p} \, \mathrm{e}^{-f/p},$$

where $\int P \, \mathrm{d}f = 1$ and $\int P f \, \mathrm{d}f = p^2$. In three dimensions the factor in front of the exponential is replaced by $f^{1/2}$.

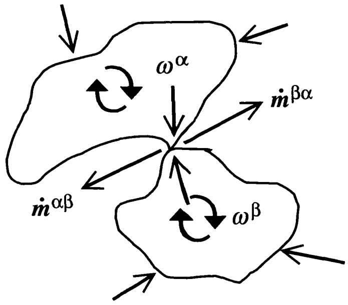

Figure 6. A model for grain dynamics.

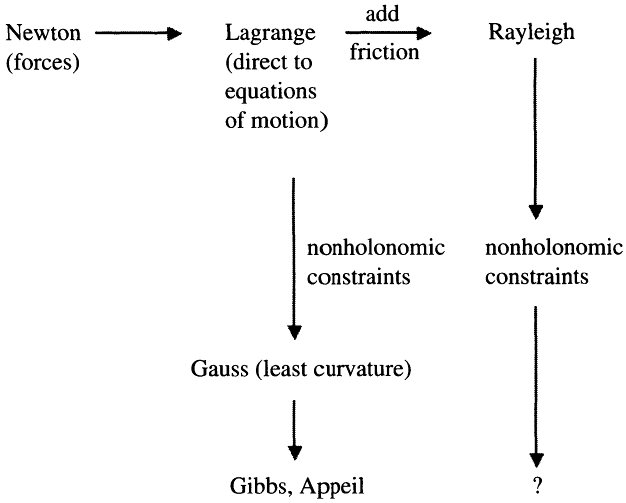

Figure 7. Nineteenth-century resolutions to standard dynamics problems.

A full analysis will need the insertion of (as yet unknown) correlation functions, but could in principle discover how forces are correlated across the material, where it has been claimed that force chains exist which resemble percolation paths. However, this simple treatment bears out the simplest experimental results, and both experiment and theory have obvious extensions. This concludes our discussion of static problems, and now we turn to the much more difficult analysis of slow dynamics.

5. Dynamics

The basic movement is when two grains are subjected to sufficient force to overcome friction when sliding and rolling result. Figure 6 shows a standard problem in 19th century dynamics textbooks, which is very complex. Without friction it is resolved by the Gibbs–Appell equations (Desloge 1982; Pars 1968; Whittaker 1944); with friction it involves putting the Gibbs–Appell equations in the Rayleighan form, which we have not seen in the textbooks. A progression of these ideas is given in figure 7.

The equations are much more complicated than statics (see figure 8) for the following reasons.

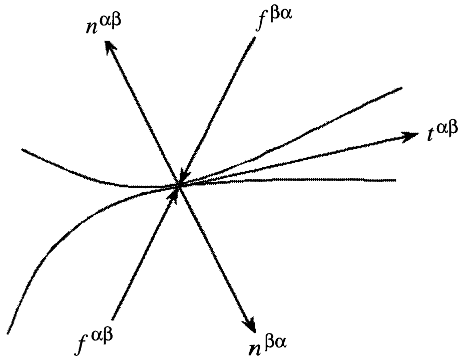

Figure 8. Representation of normal and tangential forces between grains.

(i) They involve tangents and normals to the surface which are often not well defined and also require some law of friction that could be nonlinear. We will assume t and n exist and use the crudest friction law by replacing all normal forces by an average so that the sliding force is $\mu \boldsymbol{v}$.

(ii) The description of dynamics involves both slipping and rotation, i.e. velocities $\boldsymbol{v}^{\alpha}$ and rotations $\boldsymbol{\omega}^{\alpha}$. However, only $\boldsymbol{v}^{\alpha}$ can give rise to a macroscopic variable $\boldsymbol{v}(r)$, for $\boldsymbol{\omega}^{\alpha}$ will vary from grain to grain, indeed for non-slipping motion $\boldsymbol{\omega}^{\alpha} = -\boldsymbol{\omega}^{\beta}$, where α and β are neighbours. It is similar to magnetism in an antiferromagnet. Although it is straightforward to include it as the antisymmetric complement of equation (5.6) below, we omit it for this reason, and the cruder reason that the amount of algebra is vast, so we simply omit ω altogether.

(iii) In general, forces cause particles to accelerate, but we can argue that inertia is not significant at a microscopic level and hence argue that the equation (now dropping vector notation)

$$m\ddot{x} = f_{\text{ext}} - f_{\text{reaction}} = f_{\text{ext}} - \mu\dot{x} \tag{5.1}$$

will become $f_{\text{ext}} = f_{\text{reaction}}$ and $f_{\text{reaction}} = \mu\dot{x}$, i.e.

$$\mu(v^{\alpha} - v^{\beta}) = \mu v^{\alpha\beta} = f^{\alpha\beta} - n^{\alpha\beta}(f^{\alpha\beta} \cdot n^{\alpha\beta}) - f^{\beta\alpha} + n^{\beta\alpha}(n^{\beta\alpha} \cdot f^{\beta\alpha}), \tag{5.2}$$

$$f^{\alpha\beta} \cdot n^{\alpha\beta} = f^{\beta\alpha} \cdot n^{\beta\alpha}. \tag{5.3}$$

Hence

$$v^{\alpha\beta} \cdot n^{\alpha\beta} = v^{\beta\alpha} \cdot n^{\beta\alpha} = 0, \tag{5.4}$$

i.e. grains stay in contact along the normal but slide along the tangent.

Thus the equations for the forces $f^{\alpha\beta}$ are as before sliding, but now the velocity difference $v^{\alpha\beta}$ along the tangential directions is given by equation (5.2). (As we have recorded, one should supplement these equations for f and v with the equations for ω which we omit here.)

The basic variable in dynamics which takes the central position of σ_{ij} in statics has to be the flow tensor, which we shall label ϕ_{ij} (having already used f). In normal rheology this is written as

$$\frac{1}{2}\left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i}\right), \tag{5.5}$$

but with our modification and simplification of the full equations we prefer to directly find an equation for $v(r)$. Thus, we define averages, given by equation (5.2), which are related to the static definition of stress

$$\phi_{ij}^{\alpha} = \tfrac{1}{2}\sum (R_i^{\alpha\beta} v_j^{\alpha\beta} + R_j^{\alpha\beta} v_i^{\alpha\beta}), \tag{5.6}$$

which is the analogue of equation (3.1),

$$\sigma_{ij} = \tfrac{1}{2}\sum (f_i^{\alpha\beta} r_j^{\alpha\beta} + f_j^{\alpha\beta} r_i^{\alpha\beta}), \tag{5.7}$$

since $v \propto f$ and $r \propto R$. Substituting for v from equation (5.2) we get

$$\phi_{ij}^{\alpha} = \frac{1}{2\mu}\sum R_i^{\alpha\beta}(f_j^{\alpha\beta} - n_j^{\alpha\beta}(f_k^{\alpha\beta} n_k^{\alpha\beta}) - f_j^{\beta\alpha} + n_j^{\beta\alpha}(n_k^{\beta\alpha} f_k^{\beta\alpha})) + (i \leftrightarrow j). \tag{5.8}$$

Proceeding now as in Edwards & Grinev (1998), covering the same ground that produced equations (3.2), (3.4) we reach

$$\frac{\partial \phi_{ij}}{\partial r_j} = \Lambda_{ik} f_{k(\mathrm{ext})}, \tag{5.9}$$

where

$$\Lambda_{ik} = \frac{1}{2\mu}(\delta_{ik} - \langle n_{i\alpha} n_{k\alpha}\rangle) \tag{5.10}$$

but a new 'missing equation' also emerges, the form of which is

$$\sum_{ij} L_{ij}^{m} \phi_{ij} = 0, \tag{5.11}$$

with $m = 1$ in two dimensions and $m = 1, 2, 3$ in three dimensions. As before, ϕ_{ij} has $d(d+1)/2$ components and the velocity versus force equation has only d components, so there must be $d(d-1)/2$ missing equations.

These equations are purely geometric in origin for incompressible grains. They have a similar structure to equation (3.4), but new tensors appear, in place of F and G, that have averages of the normals built into them. The details will appear in a subsequent publication, but we must catalogue a large number of crudities in the present work.

6. Unresolved problems

Many features are unresolved at the time of writing.

(i) Thresholds: friction depends on normal forces and movement only starts when force passes a threshold. In this paper we are far below or far above that threshold, but many real problems have both situations in, say, layered flow.

(ii) The viscosities that arise are related in a complex renormalized way to the bare values, and the averaging will involve the compactivity; also, the accelerations cannot be omitted.

(iii) Rolling can be inserted, and it resolves the fact that, in the treatment above, our 'flow tensor' is not the classical symmetrical velocity gradient.

(iv) We have not discussed fluctuation and not developed transport equations such as the Boltzmann or Fokker–Planck equations, so we do not calculate the time- and space-dependence of the compactivity in terms of the flow.

(v) The shape of the grains (for example, elliptical) leads to a pulsation of the modulus of $R^{\alpha\beta}$ as one grain slides over another, and there will be no flow without Reynolds dilatancy, but this has been omitted so far.

(vi) Grain contacts are always being made and lost.

7. Conclusion

We offer here the simplest picture of the flow of otherwise jammed systems. Although there is no difficulty in visualizing the rolling and sliding of packed particles, and indeed this is much the most familiar many-body problem in everyday life, it is much more difficult to handle mathematically and requires a clean break with conventional statistical mechanics based ultimately on mean free path ideas. We hope this paper is a start on the problem.

The authors have greatly benefited from discussion with Professor Robin C. Ball, Dr Raphael Blumenfeld and Professor Dov Levine on the basic theory, and with Dr John Melrose on simulations. D.C.G. acknowledges the Oppenheimer Fellowship from the University of Cambridge and a research fellowship at Wolfson College.

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Discussion

U. TÜZÜN (*Chemical and Process Engineering, University of Surrey, Guildford, UK*). How confident are you that the compactivity model based on the coordination numbers established between particles of curvilinear boundaries in point contacts will be generically applicable to, say, particles of cubic shape (where there are face–face, point–face and possibly quite a small number of point–point contacts), such as granular sugar and table salt?

Should compactivity models also consider other models of contact (i.e. point, face, etc.) as well as the isotropy of the particle shape?

S. F. EDWARDS. The compactivity concept is not based on the number of contacts, but on the absence of any constraints, apart from the volume and that the system is jammed. However, I need the function (Θ) which tells me what the jammed configurations are, and in my own calculations (but not those of related papers by other authors) I assume that only those configurations which satisfy Newton's equations uniquely will occur, i.e. if there is a larger number of contacts, Newton's equations can be satisfied by having the surplus contacts bearing zero force and can be treated as a non-contact. This is true irrespective of the shape of bodies. For zero friction the number of contacts is higher, so, if bodies have regions of low friction, the statements above become blurred. My firm belief is that until one has solved the unambiguous cases, one should not worry about anomalies, i.e. I am doing pure physics—applied physics and engineering are downstream.

D. SHERRINGTON (*Department of Theoretical Physics, University of Oxford, UK*). Would you comment about compactivity in a fluctuation–correlation relationship?

S. F. EDWARDS. The concept of compactivity implies the equivalent of the zeroth law of thermodynamics, i.e. if there exist three bodies A, B, C and if no heat flows from A to B and from B to C, then no heat will flow from A to C.

The granular equivalent of this seems intuitively correct, but we are doing direct experiments in the Cavendish Laboratory to confirm it. The fluctuation–dissipation theorem is itself a much more subtle experiment, and though such experiments have been done, and do support the compactivity concept to an extent, I am sceptical as to whether they will show anything interesting. However, computer experiments (see Makse & Kurchan 2002) do substantially support the concept, quite emphatically.

D. SHERRINGTON. Would such an experiment show anything interesting?

J. BRUJIĆ. Experiments on glasses have shown that the effective temperature, as shown in corresponding experiments, could be different from that for equilibrium and also depends on the part of the fluctuation–correlation regime one is in.

T. C. B. MCLEISH (*Department of Physics and Astronomy, University of Leeds, UK*). Does your theory apply to experimental cases in three dimensions for which the contact number is greater than four?

S. F. EDWARDS. The compactivity formula is true for any jammed system; it is just that the simplest case is for hard, rough particles. The higher number of contacts arises only when these two conditions are softened. The experiments shown clearly have more than the four contacts which are expected for hard and rough grains, but still yield powerful information which is not available in the ideal theoretical case; ideal theory and ideal experiments do not fit.