PHASE TRANSITIONS IN GRANULAR PACKINGS

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Abstract

We describe the contact network of granular packings by a frustrated lattice gas that contains steric frustration as essential ingredient. Two transitions are identified, a spin glass transition at the onset of Reynolds dilatancy and at lower densities a percolation transition. We describe the correlation functions that give rise to the singularities and propose some dynamical experiments.

PACS: 05.70, 46.10

Let us consider a packing of granular material, like sand or powder, at rest. Depending on how the packing has been prepared its density and its rheological properties can be
different. If the sand is poured into a recipient its volumetric density is relatively low and it can be moved like a fluid. A stick can be inserted into it and removed again easily since the structure of the grains is rather loose. If the recipient is then shaken at low amplitude the level of sand visibly decreases evidencing an increase of the density. Below a certain level a stick that was put inside before shaking cannot be removed anymore unless a large force is applied[1]. This effect evidences the existence of a critical density, described by Reynolds[2] on 1885, above which the granular structure cannot be sheared anymore. The grains are interlocked so densely that large deformations become impossible unless the grains are separated by some amount, i.e. the global density is decreased again. On the wet beach this is the effect which produces the dry sand around the pressure zone of our foot step. This Reynolds dilatancy has been carefully studied experimentally and numerically by soil mechanicists[3] and has been included into the equations of motion of plastic deformation[4].

Apparently there are many possible configurations of grain positions that lead to a static packing. They can be characterized by their density. Only a subset of these configurations is sufficiently dense to give rise to mechanical locking. To describe these different types of packings Sam Edwards and collaborators[5,6] have formulated a thermodynamic description of a powder which replaced the energy by the volume and the temperature by the compactivity \( \kappa = \partial V / \partial S \). We will describe the ensemble of static packings by adapting to granular media a statistical mechanical model which combines spin glass and lattice gas properties and which appears to describe also the phenomenology of the glass forming liquids[7-11].

The guiding idea of our approach is to consider each static packing as a local minimum in some free energy functional. The rough landscape in configurational space induces slow relaxation phenomena and may exhibit non-ergodic behaviour. The classical mechanism leading to such a complex structure of local minima is based on the concept of frustration. In a granular packing this frustration arises because the grains having different shapes interlock and due to steric hindrance cannot move locally except through large-scale cooperative rearrangements of many particles that usually change the volume.

In this paper we elaborate on a lattice gas model [7-11] containing frustration as an essential ingredient. This model has been called “frustrated percolation” (FP) [8] because it can be viewed as percolation [12] in a frustrated medium. Both static and dynamic properties of the model exhibit complex behaviour with features that are common to
granular packings, structural glasses and spin glasses. This may suggest that, just as
the Ising model provided insight into the physics of critical phenomena near second-order
phase transitions, the frustrated lattice gas model presented here may help to understand
the Reynolds transition through the spin glass transition.

Let us consider a square lattice tilted in such a way that the axis of the lattice forms
$45^\circ$ with the horizontal. Gravity points vertically down. We assign to each edge of the
lattice an interaction $\epsilon_{ij} = -1$ with probability $\pi$ and $\epsilon_{ij} = +1$ with probability $1 - \pi$. We
assume for now that this assignment is fixed in space, i.e. that the disorder is quenched.
We also fix $\pi = 1/2$. A loop is considered frustrated if the product of the sign along the
loop is negative. Next we introduce particles on the vertices of the upper part of the lattice
and let them move down randomly with the constraint that frustrated loops cannot be fully
occupied, i.e. that at least one site must be missing in each frustrated loop. The particle
stops moving when either all vertices below are occupied or if any move downwards would
violate the constraint. So, the constraint induces defects or holes into the system, since a
frustrated loop must necessarily contain at least one empty site. Therefore when no more
particles can be poured into the system from above the system will have a density $\rho_m$ less
than unity, i.e. the density when every site would be occupied by one particle.

One can now allow the particles to diffuse by exchanging their positions with a nearest-
neighbor hole such that no frustrated loop becomes completely occupied. Due to gravity
we consider the probability $p_1$ to move upwards to be less than the probability $p_2$ to move
downwards. We let the system evolve for a time $t$ after which the probability $p_1$ to move
upwards is put equal to zero and the system is left to relax until the particles do not
move anymore. We expect that the final density will be lower than before the exchange
of positions. The smaller the ratio $x = p_1/p_2$, and the larger the time $t$ the lower we
expect the final density. At low particle densities frustration does not significantly inhibit
diffusion. At high densities, however, it can happen that a particle can only move through
the system by long-range collective rearrangements of many particles. On experimental or
numerical time scales this particle will then be frozen within a cluster of particles which
interlock each other. Such a frozen cluster is depicted schematically in Fig. 1. On very long
time scales frozen clusters can dissolve, which is why they have also been called “quasi-
frozen” in the literature [7]. The extremely long life time of frozen clusters is responsible
for the slow relaxation phenomena at high densities.

The case $p_1 = p_2$ and $t = \infty$ is identical to “site-frustrated percolation” as introduced
Figure 1: Schematic representation of (a) a spanning cluster of particles (b) a spanning cluster of frozen particles. Filled circles are frozen particles and empty circles are particles that can diffuse.

Before [8]. In fact also a “bond-frustrated percolation” has been introduced [7,9] in which the bonds instead of the sites are occupied and which has the same critical properties as the site model. The static properties of bond-frustrated percolation were studied via a renormalization group calculation [9] on a hierarchical lattice, using a Hamiltonian formalism which will be introduced in the Appendix. The results have been confirmed by Monte Carlo simulations on the square lattice [10].

Two critical points were found, one at a density $\rho_p$ which is in the universality class of the ferromagnetic 1/2-state Potts model, while the second critical point at $\rho_R$ is in the same universality class as the $\pm J$ Ising spin glass transition. Each critical point is characterized by a diverging length, associated with the quantities

$$p_{ij} = p_{ij}^+ + p_{ij}^-$$

and

$$g_{ij} = p_{ij}^+ - p_{ij}^- .$$

Here $p_{ij}^+ (p_{ij}^-)$ is the probability that on one hand the sites $i$ and $j$ are connected by at least one path of bonds and on the other hand the product over all signs (or “phase”) $\epsilon_{kl}$ along the path connecting $i$ and $j$ is $+1 (-1)$. In the granular packing a path between $i$ and $j$ corresponds to a chain of touching grains that connect grains $i$ and $j$. The length $\xi_p$ associated with the pair connectedness function $\langle p_{ij} \rangle$ diverges at the percolation density $\rho_p$, while the length $\xi$ associated with $\langle g_{ij}^2 \rangle$ diverges at the higher density $\rho_R$, where $\langle .. \rangle$ is the average over all possible interaction configurations $\{ \epsilon_{ij} \}$. The dynamic properties of site-frustrated percolation with $p_1 = p_2$ have also been studied numerically on a two dimensional square lattice[13]. It was found that the system below the percolation transition, develops a dynamical behaviour in the density-density autocorrelation function, characterized by a two step relaxation function typical of glass forming liquids. Finally as
the temperature approaches the high density critical point $\rho_R$ the system freezes with the diffusion coefficient going to zero.

The rich scenario described above, including the transition at $\rho_R$ occurs due to the freezing of paths, i.e. the interference of different phases. If the particles are free to move the cancelation on average of paths of different phases would make $g_{ij}$ very small. The only way to maintain the asymmetry in the phase of the paths, needed to have a large $g_{ij}$ is by developing frozen paths. The length $\xi$ is a measure of the size of the clusters of frozen sites (see fig. 1). At the critical density $\rho_R$ the length $\xi$ diverges, i.e. the different paths connecting two grains $i$ and $j$ being arbitrarily far apart can block each others motion freezing the entire system mechanically. This is precisely the phenomenology of the Reynolds transition as described in the introduction. It seems therefore plausible that while $\rho_p$ corresponds to the lowest density for which a static packing can be achieved, the density $\rho_R$ corresponds to the onset of Reynolds dilatancy. Since FP close to $\rho_R$ exhibits the same static properties of the spin glass transition and dynamical properties closely related to glass forming liquids, we expect that granular media near the Reynolds transition should have the static properties of a spin glass and the dynamical properties resembling that of a glass forming liquid.

In the rest of this paper we want to elucidate more deeply these properties and propose experimental consequences of this equivalence to granular packings. In the Appendix we establish the relation to the Hamiltonian description of spin glasses.

Spin glasses have been studied extensively [14] and are known to have a “spin glass transition” at temperature $T_{SG}$ in three dimensions. This transition is characterized by a diverging correlation length associated with the square of the spin-spin correlation function, and consequently exhibits a strong divergence in the non-linear susceptibility. The specific heat instead exhibits a weak singularity characterized by a negative exponent $\alpha$, very difficult to detect experimentally or numerically. The FP also exhibits a diverging length associated to $g_{ij}$. However the strong divergence associated to the sum over the sites of $g_{ij}$, is not accessible by an experimental probe. Instead the compressibility associated to density fluctuations has the same weak behaviour as the spin glass specific heat.

To pursue the analogy between granular systems and spin glasses it seems useful to consider also the thermal excitations of a packing. A granular temperature $T$ has been introduced by Ogawa [15,16] as being proportional to the kinetic energy of the particles. This can be implemented experimentally by putting the sand into a box which is vibrated
at constant frequency $\omega$. The box should not be too large to ensure that the vibration energy is homogeneously distributed inside the system. Varying the amplitude $A$ of the vibration one can control the temperature via $T \propto A^2$. On can, starting with a given $A_0$, decrease the amplitude linearly with time $t$, like $A(t) = A_0 - rt$ with a rate $r$. It has been shown numerically [13] that in the site-frustrated percolation model, the volume per particle $v$ as function of the temperature exhibits a plateau below a temperature $T_g(\dot{T})$ which is the lower the lower the cooling rate $\dot{T}$. Consequently the final density when the temperature $T \to 0$ is higher for smaller cooling rates. This effect is also found in real glass forming liquids but not reproducible within the simple spin glass model. Since in our analogy $r \sim \dot{T}$ we predict that in the above described experiment of lowering the vibration amplitude with time one should obtain different final densities for different rates $r$. In fact $r$ would be a good control parameter for the Reynolds transition. It would be interesting to experimentally verify this prediction.

Another parameter to control the final density could be the amplitude $A$ itself in the case in which the vibration is suddenly stopped and the system very rapidly falls into a static configuration. Since the quenching takes a time that is of the order of the period $\frac{2\pi}{\omega}$ of the vibration the cooling rate $\dot{T}$ monotonically increases with $A$ so that then the control parameter steering the Reynolds transition would be $A$. A more detailed investigation of this dependence would also be interesting.

It is experimentally not easy to measure dynamical auto-correlation functions or the local response to a mechanical perturbation in particularly in three dimensions. More accessible is the macroscopic relaxation of a granular packing. The analogy to glass forming liquids would suggest that at high densities the restructuring after a perturbation should be slower than logarithmic. We think that doing Reynold’s classic bag experiment [2] in which a bag filled with sand and water is deformed and the water level is seen to go down, might be a candidate to measure this slow relaxation, namely by measuring how fast the level increases again with time. For such an experiment, however, the elastic properties of the bag will have to be carefully controlled.

By describing a granular packing through a lattice model in which particles must fulfill non-local, quenched constraints we found that the configurational ensemble is equivalent to that of frustrated percolation. This later model is known to exhibit a percolation and a spin glass transition and its dynamical properties resemble those of glass forming liquids. It seems plausible to identify the spin glass transition to the onset of Reynolds dilatancy.
In reality, however, the steric constraints in granular systems are not quenched in space but determined by the actual positions of the grains. Considering annealed disorder instead would smoothen the percolation transition but leave the spin glass transition essentially unaltered [7]. Configurations that are obtained starting from a given granular packing (for instance through shaking) do, however, have memory of the original disorder, so that the experimental reality is neither completely quenched nor completely annealed. Further investigations in this direction are under way.

Appendix:

The general Hamiltonian formalism needed to describe the site-bond frustrated percolation problem is given by:

\[ -H = J \sum_{ij} [(\epsilon_{ij} S_i S_j + 1) \delta_{\sigma_i \sigma_j} - 2] n_i n_j + \mu \sum_i n_i \tag{3} \]

where \( n_i \) is the occupancy of site \( i \) (\( n_i = 1 \) if site occupied and \( n_i = 0 \) if site empty), \( S_i \) a spin variable on an occupied site, having the states \( \pm 1 \). \( \sigma_i = 1 ... s \) are Potts variables which assume \( s \) states, \( \mu \) is the chemical potential for a particle and \( J \) is related to the chemical potential for a bond \( \mu_b \) through \( e^{\mu_b/kT} = (1 - e^{-2J/kT})/e^{-2J/kT} \). In the limit \( s \to 1/2 \) the Hamiltonian (3) reproduces site-bond frustrated percolation. In this model the configurations are made of sites connected by bonds. The bonds cannot occupy entirely a frustrated closed loop. As a particular case in the limit \( \mu \to \infty \) (\( \mu_b \to \infty \)) and \( s \to 1/2 \) we obtain frustrated bond (site) percolation while in the limit \( \mu \to \infty \) and \( s \to 1 \), we obtain the \( \pm J \) Ising spin glass model.

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