EQUIVALENCE BETWEEN RANDOM CLOSE PACKING IN GRANULAR MATTER AND FREEZING IN THE HARD SPHERE MODEL

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Abstract. The notion of random close packings of a bulk static collection of ball bearings or sand grains was introduced in the 1960's by G.D. Scott and J.D. Bernal. There have been numerous attempts to understand the packings. We give a short argument, following an earlier paper on liquids by Bernal, which explains the packings in purely geometric terms.

In 1960 G.D. Scott and J.D. Bernal began a series of experiments, with others, on large collections of static ball bearings and other 'granular matter' [1, 2] in part studying how the material compacts when shaken repeatedly. They discovered a barrier: the system would not compact beyond a rather sharp volume fraction they called 'random close packing' (RCP), which they determined to be about 0.64.

Many attempts have been made since then to illuminate the phenomenon [3, 4, 5, 6, 7, 8]. The experiments started soon after the first simulation to successfully exhibit the first order phase transition of the hard sphere model of equilibrium statistical mechanics [9], to which they refer and presumably used for intuition. In this paper we extend some of Bernal's ideas about liquids to show a close connection between random close packing at volume fraction 0.64 and the freezing of a hard sphere fluid at density 0.49 [10], a correspondence which we show is useful in both directions. We start with a quick summary of the hard sphere model, and then derive a connection with random close packing.

The hard sphere model of particles in thermal equilibrium uses point particles of mass m constrained to be at least some fixed distance σ apart, with no other interaction. Given $x \in \mathbb{R}^3$ and $s > 1$ we introduce the constraint function $G_{x,s}$, of variables $a, b, \cdots \in \mathbb{R}^3$, to have value 0 if any of the variables $a, b, \cdots$ is further than s from x, or if any distinct pair of $x, a, b, \ldots$ is closer than σ; otherwise $G_{x,s} = 1$. Fix (temperature) $T > 0$, (pressure) $P > 0$, and the number $N + 1$ of particles, and define the (temperature/pressure) relative probability density that N of the particles have position/momentum coordinates $(x_1, p_1), (x_2, p_2), \cdots, (x_N, p_N)$ given that the other has coordinates (x, p), to be

$$\exp\Big[-\frac{1}{2mkT}\Big(p\cdot p+\sum_{j=1}^{N} p_j\cdot p_j\Big)\Big]\int_0^\infty G_{x,s}(x_1,\cdots,x_N)\exp\Big[-\frac{P\sigma^2}{kT}\Big(\frac{4\pi s^3}{3}\Big)\Big]ds. \tag{1}$$

This is the hard sphere model. Notice that the density factors into a part dependent only on the momentum coordinates, controlled by T, and a part dependent only on the position coordinates, controlled by P/kT.

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Simulation shows there is a unique infinite particle state for any pair (P, T) of pressure and temperature except for a special value $P\sigma^3/kT = C^*$, C^* a dimensionless constant, where there is coexistence of the high density phase which exists for $P\sigma^3/kT \geq C^*$ with density ≥ 0.54, and the low density phase which exists for $P\sigma^3/kT \leq C^*$ with density ≤ 0.49.

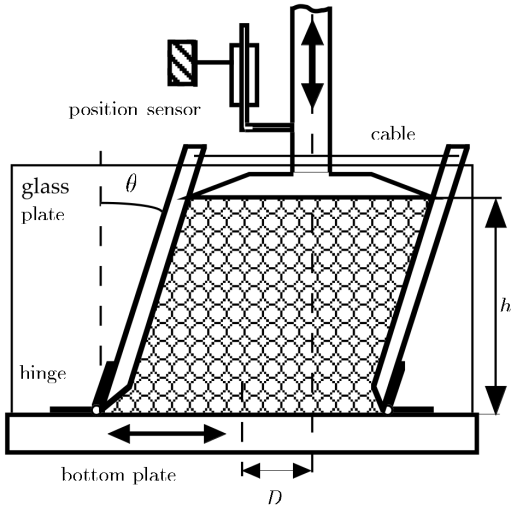

FIGURE 1. Sketch of shear cell used by Nicolas *et al* from [8], using two hinged vertical walls attached to a moving bottom, and pressure applied from above.

The experiments in 1960 came in the midst of a long program of Bernal's investigating the nature of liquids, in particular their particle configurations; see [11] for a review. From his 1959 paper [12], in which he concentrated on liquid argon, we take the assumption that one can picture each argon atom in the liquid as a hard ball of some diameter σ vibrating at high frequency, with a relatively slow drift, and that it is useful to shift focus from the instantaneous ball positions to that of the slowly moving vibrating balls, which we will call 'clouds'. We add the assumption that the clouds are roughly spherical and have a volume which depends only on (P, T), in particular that it is roughly the same size in the coexisting liquid and solid states.

Thus one understands the microscopic origin of macroscopic properties of fluid argon to derive from essentially static clouds made of rapidly vibrating hard balls. In particular these give rise to internal pressure of the liquid, supporting for instance the bulk modulus, by vibrating against neighboring clouds. Such clouds should be large enough to touch other clouds, to provide the pressure. It is not a big leap for us to introduce clouds in the hard sphere model to model *static* sand grains, supporting the bulk modulus by contact forces rather than the vibration in the clouds.

In the crystalline state of the hard sphere model the size of the cloud must be such as to touch the 12 neighbors, giving a volume fraction about 0.74. Since the ratio of the volume fractions in the coexisting states for the underlying vibrating balls should be the same as the corresponding ratio for the clouds:

$$\frac{0.49}{0.54} = \frac{\text{RCP}}{0.74}, \tag{2}$$

this yields a volume fraction for RCP about 0.67, which is within 4% of the current best estimate of 0.646 [13, 14].

In summary, the clouds of vibrating balls in thermal equilibrium behave like well-mixed static sand grains, with the hard sphere freezing point appearing as granular RCP at about 0.646. Indeed, one might well use the hard sphere model to predict the existence of RCP in sand, which one might say is what Bernal and Scott were exploring.

We now shift direction to examine the granular experiments and see what they might say about the hard sphere model.

In 1964 Scott found a way to drive ball packings well above the barrier of volume fraction 0.64, and this was very revealing [15]. He found that if he cyclically sheared a low volume fraction collection in a box with moving side walls (a 'shear cell'), as illustrated in Figure 1, the system would first compact to volume fraction 0.64, and then slowly rearrange close to a densest packing at volume fraction 0.74.

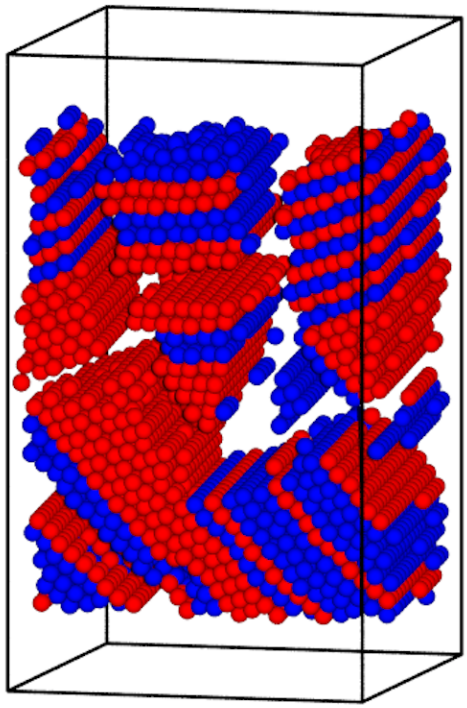

FIGURE 2. Hexagonal layers in growing granular crystallites, made from unpublished simulation data by Jin for [14]. HCP layers are blue and FCC layers are red.

There were difficulties understanding how the rearrangement took place in time, since this would require looking inside the packing, but this was recently done using laser sheets [13, 14] and what was discovered is that in order for the packing to rearrange at volume fraction 0.64 it had to create small clusters of balls (crystalline nuclei), randomly in the middle of the packing, which then grew. This is, of course, how a liquid freezes (homogeneously), so if one considers the nucleating granular system at volume fraction 0.64 as akin to a supercooled

liquid about to freeze, its correspondence with the hard sphere system at density 0.49 suggests that the phase in the hard sphere model at density above 0.54 is not just high density but 'crystalline'. Furthermore, examination of the grain configurations from [13, 14] shows that the high volume fraction granular configurations consist of random layers of hexagonal slabs; they are not regularly layered as would be FCC or HCP. See Figure 2.

Such a random layering might still have *orientational* long range order from the direction normal to the layers, but not full positional long range order as in a true crystal. The correspondence with the hard sphere model suggests the same is true for the high density phase in that model. (For a related question about hard colloids see [16].)

The evidence from the hard sphere model of a singularity in ball packings as a function of volume fraction is remarkable. The recent evidence from [13, 14] that granular matter crystallizes homogeneously is then suggestive that there is a simple connection to freezing in the hard sphere model, so that there really exists only one such ball packing singularity.

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