# Random Close Packing of Granular Matter

by

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**Abstract**

We propose an interpretation of the random close packing of granular materials as a phase transition.

October 2007

PACS Classification: 45.70.Cc, 81.05.Rm, 45.70.-n

* Research supported in part by NSF Grant DMS-0700120

The phenomenon of random close packing was popularized by Bernal [B] as part of his effort to model liquids. The experiments which clarified many of the issues were performed by Scott et al [S,SK], and showed the following. If a large number of monodisperse hard spheres, for instance steel ball bearings, are gently poured into a container, the volume fraction will be roughly 0.61. If the container is repeatedly shaken vertically, this density rises to about 0.64, and careful protocols lead to reproducible lower and upper limits on the volume fraction, called, respectively, random loose packing ($0.608 \pm .006$) and random close packing ($0.6366 \pm 0.0005$). (Recall that the densest possible packing of monodisperse spheres has volume fraction $\pi/\sqrt{18} \approx 0.74$.) We will not discuss random loose packing any further because of complications due to friction and to the ambient fluid surrounding the spheres – air, water, etc.

Scott et al [SC] noted that volume fractions somewhat above 0.64 (up to approximately 0.66) could be obtained if the material was cyclically sheared, and that this result was accompanied by small crystal-like clusters of spheres. This was confirmed and explored by Pouliquen et al [ND] (who also performed experiments employing horizontal shaking [PN]), obtaining volume fractions up to 0.70, again accompanied by crystal-like clusters of spheres.

In light of the above, the volume fraction 0.64 is generally described as the highest volume fraction at which (static) granular matter can be randomly prepared, in the sense that higher volume fractions require the introduction of some crystal-like clusters (which are "ordered" rather than "random"). This general understanding has recently been questioned in an influential paper by Torquato et al [TT], in which the authors claim to "have shown that the notion of RCP (random close packing) is not well defined mathematically". We will suggest an experimental test of an unambiguous meaning to the intuitive notion of a maximum density for disordered granular matter, together with a mathematical model of traditional form.

Our proposed characterization of random close packing is motivated by properties of the hard sphere model of classical statistical mechanics. In that model point particles, with the usual position and momentum degrees of freedom, interact only through a hard core: no pair may approach closer than some fixed separation $D$, and the particles evolve dynamically through elastic collisions of imaginary spheres of diameter $D$ surrounding their positions. Our interest in the hard sphere model stems from the demonstration by Alder and Wainright [AW], by molecular dynamics simulations, that the model exhibits a first order phase transition between a fluid, which exists at volume fractions below $\approx 0.49$, and a solid, believed to be crystalline, which exists at volume fractions above $\approx 0.54$. Between 0.49 and 0.54 there is a mixed phase. Using the canonical ensemble we integrate out the momentum variables and consider the "reduced" probability distributions on the phase space of the position variables alone, in the infinite volume limit. They are the infinite volume limits of the uniform distributions on packings for fixed volume fraction. In particular the distribution $p(m)_f$ of the mixed phase at volume fraction $f$, $0.49 \leq f \leq 0.54$, is represented by an average of the distributions of the pure phases: $p(m)_f = c\, p_{0.54} + (1-c) p_{0.49}$, where $p_{0.49}$ is the distribution of the highest

density fluid, $p_{0.54}$ is the distribution of the lowest density solid, and $0 \leq c \leq 1$ is such as to produce the volume fraction $f$, namely $c = (f - 0.49)/0.05$. Note that 0.49 is the volume fraction of "freezing". Therefore assuming, as generally believed, that the solid phase is crystalline (in fact face centered cubic [W,BF]), we interpret 0.49 as the "highest random density" among monodisperse spheres. (These structural features have been confirmed not only by many computer simulations but also in experiments with appropriate colloids [RD].) Intuitively, at any volume fraction above the freezing point there is a nonzero probability of (seeing) an infinite, ordered crystal. It is the use of the infinite volume limit, together with the probabilistic formalism, which produces a sharp phase transition between disorder and order in equilibrium statistical mechanics [FR].

We emphasize that there are sphere packings with packing fraction $d > 0.49$ which might well be described as random, for instance packings corresponding to any metastable extension of a fluid isotherm. However the total of all such packings has probability zero with respect to the (infinite volume limit of the) uniform distribution on packings with packing fraction $d$. Our use of this distribution as the touchstone of relevancy is in accord with its common appearance in statistical physics and probability theory; the best justification one can give is that it is commonly found that practical sampling of phase space seems to occur in this way, in particular in the natural dynamics of matter in thermal equilibrium.

The traditional hard sphere model does not include the effects of gravity and cannot represent the properties of granular matter, in particular that of random close packing. However a slightly modified ensemble framework has been proposed to model granular matter. Specifically, in the original proposal of Edwards et al [EO] one uses a uniform distribution on those static monodisperse sphere packings, of fixed volume fraction, which are mechanically stable under gravity. One can add friction to the spheres and perhaps other restrictions besides volume fraction; adding a condition of fixed pressure might be useful, though it is not clear if pressure is isotropic in granular materials.

As is true for solids in thermal equilibrium, in order that an ensemble method be appropriate for (nonequilibrium) granular matter it is important to restrict the protocols used to produce beds of granular matter at fixed volume fraction (and perhaps pressure etc.) One feature that is necessary is that the protocol be "history independent" [NK] in that it give equivalent results starting with beds originally prepared in any manner. There are three types of protocols which claim to produce history independent beds of monodisperse granules with well defined volume fractions: vertical vibration (often called "tapping"); fluidization followed by sedimentation; and cyclic shearing. These methods have produced history independent beds with volume fractions in the following ranges: 0.605 to 0.625 by vertical vibration [NK,RR]; 0.685 to 0.70 by cyclic shearing [NDP]; and 0.57 to 0.63 by fluidization/sedimentation [SG,SN].

It is known from Schröter et al [SN] that granular beds prepared by fluidization/sedimentation undergo a phase transition, as volume fraction is varied, at approximately 0.60 volume fraction, as measured by two different responses to shear.

Given the features of the hard sphere model we conjecture that history independent granular beds would also show a first order phase transition, with a mixed phase for volume fractions between 0.64 and 0.74, again exhibited through the response to shear or other mechanical probe. As for the hard sphere model, the distribution for granular matter in the mixed phase would be a mixture, with one component, at volume fraction 0.74, representing a crystal, and the other, at volume fraction 0.64, representing a disordered phase. This would give a well defined meaning to the phenomenon of random close packing of granular matter just as the freezing density defines a similar concept for the hard sphere model or for hard sphere colloids. It would also suggest that computer simulations may need to more faithfully model physical protocols to see such delicate phenomena.

We note the connection between this proposal and that of Kamien and Liu [KL], which also uses the hard sphere model to understand random close packing. In [KL] random close packing is associated with the end point of a metastable branch in the hard sphere phase diagram, while we use the hard sphere model only to predict behavior in a related but different ensemble, of packings which are mechanically stable under gravity, and in particular we predict a phase transition at volume fraction 0.64.

Experimental protocols have not yet produced history independent beds with volume fraction in any interval containing the volume fraction of interest, 0.64; this will be necessary before our conjecture can be checked against the behavior of granular matter. Alternatively it might be possible to test the conjecture by realistic, history independent computer simulations.

## Acknowledgements

We gratefully acknowledge useful discussions with Persi Diaconis and Matthias Schröter.

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