Experiments on Random Packings of Ellipsoids

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Recent simulations indicate that ellipsoids can pack randomly more densely than spheres and, remarkably, for axes ratios near 1.25:1:0.8 can approach the densest crystal packing (fcc) of spheres, with a packing fraction of 74%. We demonstrate that such dense packings are realizable. We introduce a novel way of determining packing density for a finite sample that minimizes surface effects. We have fabricated ellipsoids and show that, in a sphere, the radial packing fraction \( \phi(r) \) can be obtained from \( V(h) \), the volume of added fluid to fill the sphere to height \( h \). We also obtain \( \phi(r) \) from a magnetic resonance imaging scan. The measurements of the overall density \( \phi_{\text{avr}} \), \( \phi(r) \) and the core density \( \phi_0 = 0.74 \pm 0.005 \) agree with simulations.

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The structure of liquids and glasses [1], the thermodynamics and kinetics of the crystal-liquid transition [2,3], crystal nucleation and growth [4–6], the structure and flow of granular materials [7,8], the jamming transition [9], the quality of ceramics, and a host of other problems in science, mathematics and technology depend on the packing properties of hard particles [10–12]. In particular, the freezing transition of simple liquids is related to the fact that a jammed random packing of spheres at volume fraction 0.64 [13] is less dense than the fcc crystal packing at 0.74 [2,3,14]. Thermodynamically, at high density, free volume considerations dominate the entropy and favor the configuration with the highest density. Until recently most studies of hard particle packings dealt with spheres. Within the past year, both experiments and simulations have shown the richness of the packing problem when the shape is modified even by the affine deformation of a sphere to an ellipsoid [15,16]. While the papers were inspired by random packing experiments on M&M® candies [17], most of the results were based on simulations. The simulations [15] show that the density of jammed random packings of ellipsoids can approach that of the crystal packing, raising the possibility of an elusive thermodynamically stable glass.

The present study was undertaken to test whether such a dense random packing could be realized experimentally. Simulations use frictionless particles with periodic boundary conditions. Can experiments with a finite number of particles with friction yield information on the bulk packings? We show that injected fluid volume vs height in a packed spherical container provides a good measure of \( \phi_{\text{bulk}} \) for rather small systems. This technique should prove useful for many granular material and packing studies. We confirm these measurements with magnetic resonance imaging (MRI) studies, which provide the most direct method in the case of nonmetallic particles.

In previous work, some of us have used a modified Lubachevsky-Stillinger algorithm to estimate the packing fraction of randomly jammed ellipsoids [15]. The algorithm is an event-driven molecular dynamics simulation with controlled particle growth [18]. The packing fraction, as a function of \( \alpha \), for ellipsoids with aspect ratios \( \alpha:1:1 \) and \( \alpha^{1/2}:1:\alpha^{-1/2} \) is shown in Fig. 1(a). Surprisingly the packing fraction has a cusplike minimum at \( \alpha = 1 \) (spheres, \( \phi = 0.64 \)) and increases sharply with \( |\alpha - 1| \) [15]. What was striking in these early simulations is that for ellipsoids with aspect ratios 1.25:1:0.8 the packing fraction approaches that of the densest sphere packing, which corresponds to the fcc lattice and its stacking variants at \( \phi = 0.7405 \). Later simulations with a small periodic unit cell (32 particles/cell) and very slow growth with the same ellipsoids obtained an apparently disordered packing with \( \phi = 0.7427 \), Fig. 1(b). Moreover, using this procedure, we found crystal ellipsoid packings that are denser than the fcc packing of spheres [16]. Confidence in the simulations was enhanced by the close agreement with the measured value of the density for M&M candies (\( \alpha \sim 1/2 \), \( \phi = 0.71 \)) from simulation and \( \phi = 0.69 \pm 0.01 \) from experiment [15].

The 1.25:1:0.8 ellipsoids used for these experiments were fabricated using a stereolithography machine 3D Systems Model SLA 250. The machine uses a UV laser
this study.

of 0.7427. (c) A photograph of several ellipsoids fabricated for
aspect ratio
increases linearly with
j
/.0001

local density is radially symmetric. We assume that the
minimize the surface to volume ratio, a spherical container
smaller than its bulk value,

has a cusplike minimum at
/.0011

FIG. 1 (color online). (a) The simulated packing fraction for
ellipsoids of ratios \(\alpha:1:1\) and \(\alpha^{-1/2}:1:\alpha^{1/2}\) as a function of \(\alpha\). It
has a cusplike minimum at \(\alpha = 1\) (spheres, \(\phi = 0.64\)) and
increases linearly with \(|\alpha - 1|\). (b) A simulated jammed
configuration for 32 particles/periodic unit cell of ellipsoids with
aspect ratio 1.25:1:0.8, which has a disordered packing density
of 0.7427. (c) A photograph of several ellipsoids fabricated for
this study.

with an absorption length of \(\sim 100 \mu m\) in monomer
CIBATOLL® SL 5170. The computer controlled laser
scans the surface, leaving a solid polymerized layer
100 \mu m deep. This patterned layer is then lowered and
the next layer written by polymerizing the covering liquid.
The beam width, which determines the \(xy\) resolution, is
comparably scaled at 150 \(\mu m\) allowing the feature size of
150 \(\mu m\) in each direction. The size of our ellipsoids was
2.344:1.875:1.5 cm for \(<1\%\) resolution. A picture of a few
ellipsoids used in this study is shown in Fig. 1(c).

The simplest way to measure the packing fraction is to
pack a container with the particles and determine the
volume of fluid that fills the voids [19]. In such measure-
ments, the average packing density \(\phi_{av}\) is significantly
smaller than its bulk value, \(\phi_0\), even for large containers.
This is due to the lower density in the few layers of
particles near the wall (at the wall the density is zero). To
minimize the surface to volume ratio, a spherical container
is preferred. We expect that for a spherical container the
local density is radially symmetric. We assume that the
radial density profile has the form

\[
\phi(r) = \phi_0 - \Delta \phi \left(\frac{R - r}{a}\right),
\]

where \(a\) is the mean particle radius, \(a = (a_1a_2a_3)^{1/3}\) for
ellipsoids with principal axes \(2a_1, 2a_2, 2a_3\), and \(\Delta \phi(x)\) is
an approximately universal function, independent of \(R\), but
dependent on particle shape. The correction \(\Delta \phi(x)\) decays
rapidly to zero for \(x \gg 1\). If we use (1) in \(\phi_{av} = 3 \int_0^R \phi(r)r^2dr/R^3\), we obtain the expansion of \(\phi_{av}\) in
powers of \(\delta = a/R\):

\[
\phi_{av} = \phi_0 - 3\delta (\Delta \phi)_0 + 6\delta^2 (\Delta \phi)_1 - 3\delta^3 (\Delta \phi)_2,
\]

where \(\Delta \phi(x) = \int_0^R \Delta \phi(x) r^2dr = \int_0^\infty \Delta \phi(x) r^2dx\). Thus, for
large spherical containers, the main correction to the mea-
sured total density varies as \(a/R\), and a linear extrapolation
of \(\phi_{av}\) vs \(a/R\) to \(a/R \rightarrow 0\) should give a good estimate of
the bulk density \(\phi_0\) [19,20]. In Fig. 2, we present the re-

sults of experiments with spherical glass marbles of di-

1.1 cm, mini M&M candies (ellipsoids of revolution

or spheroids) and our fabricated ellipsoids with semi-

axes 1.25a:a:0.8a, \(a = 0.938 \pm 0.008 cm\), in spherical

flasks of several sizes. The ellipsoids or spheres were

poured into glass flasks which were then shaken by hand.

More particles were added until further shaking failed to
produce space for the addition of another particle. In order
to overcome friction, we added a small amount of isopro-

panol as a lubricant for the marbles and ellipsoids. Also

shown are computer generated jammed configurations of

frictionless spheres and ellipsoids in geometrically similar

spherical containers [18]. From these data, it is clear that

the density is the largest for the fabricated ellipsoids and

smallest for the spheres. Extrapolating the simulation re-

sults to \(a/R \rightarrow 0\), we find \(\phi_0\) (spheres) = 0.642 \pm 0.002, \(\phi_0\)

(M&M’s) = 0.700 \pm 0.002, and \(\phi_0\) (ellipsoids) = 0.741 \pm 0.002.

Our experimental data for glass marbles and M&M candies agree well with simulated results of

frictionless particles. However, for the fabricated ellip-

soids, the measured packing densities are noticeably lower

than the simulated result for \(a/R > 0.1\). Frictional effects

appear more significant in small containers for objects with

rotational degrees of freedom. Clearly a meaningful ex-

trapolation for the 0.8:1:1.25 ellipsoids to \(a/R \rightarrow 0\) is

FIG. 2. Packing fraction as a function of inverse spherical
container size. Empty symbols and lines are data and extrapo-
lation for simulated packing. The extrapolated values are as
follows: for spheres, \(\phi_0 = 0.642 \pm 0.002\); for mini M&M’s,
\(\phi_0 = 0.700 \pm 0.002\), 1.25:1:0.8 ellipsoids \(\phi_0 = 0.741 \pm
0.002\). Solid symbols are the corresponding experimental results.
Moreover, if $h$ is small enough, the function with the coefficient of the higher-order term remaining $(\pi/3)(1 - \phi_0)$ for a fit in the range $-R_1 < h < R_1$, $V(h)$ is a cubic function with the coefficient of the $h^3$ term remaining $(\pi/3)(1 - \phi_0)$ for a fit in the range $-R_1 < h < R_1$. Therefore, a quick and rather accurate way of obtaining an estimate for the core (bulk) density $\phi_0$ in a packing is to fit a cubic polynomial to $V(h)$ [21]. In Fig. 3, $V(h)$ as measured is shown together with the corresponding cubic fits. For our fabricated ellipsoids we find a core density $\phi_0 = 0.739 \pm 0.005$, which is in excellent agreement with the computational result $\phi_0 = 0.735 \pm 0.01$. For spheres we measure $\phi_0 = 0.635 \pm 0.005$ vs the known bulk density $\phi_0 = 0.64 \pm 0.005$.

We also performed MRI scans of the ellipsoids in the same spherical container to assure that the packing did not show signs of crystal or orientational order. The space between ellipsoids was filled with water. MRI scans were collected on a Siemens 3T Allegra scanner. The images were acquired at 1 mm$^3$ isotropic resolution per voxel using a conventional 3D FLASH sequence (acquisition matrix $= 256 \times 256 \times 256$, FA $= 10^\circ$, TE $= 2.65$ ms, TR $= 5.5$ ms, BW $= 260$ Hz/pixel) in a standard birdcage head coil. The manufacturer’s algorithm for correcting the geometric distortion in the 3D large field of view was applied to the images. Figure 4 inset shows a gray scale slice from the MRI data, and no apparent ordering is present [22]. We also use the MRI data for an independent measurement of $\phi(r)$. Simple thresholding converts the gray scale images into binary ones. Using the binary data, it is straightforward to calculate $\phi(r)$, which is shown in Fig. 4 with a solid line, while the average packing density inside a spherical core of radius $r$, $\phi_{av}(r) = \frac{3}{4\pi r^2} \int_0^r \phi(r') 2\pi r'^2 dr'$, is shown as the line with circles. The oscillations come from the finite particle size and the effects of the walls. Their periodicity is approximately the mean particle diameter, $2a$, $a = (a_1 a_2 a_3)^{1/3}$. The dashed line and triangles are results from a simulated packing with the same geometry. The average density inside the core $r < 0.6R$ is 0.735, and the error due to finite pixel size, uncertainty in the threshold and geometrical distortions in the MRI scan total $\pm 0.01$.

To summarize, we find that it is relatively straightforward to realize experimentally random ellipsoid packings whose density matches simulations of hard frictionless particles. We have also demonstrated a new way to mea-
FIG. 4. (a) Radial density profile \( \phi(r) \). Solid line, \( \phi(r) \) measured from MRI; circles, \( \phi_{\text{ave}}(r) \) measured data averaged from center out to radius \( r \); dashed line, \( \phi(r) \) measured from simulations; triangles, \( \phi_{\text{sim}}(r) \) from simulations. Both measured and simulated average densities in a core \( r < 0.6R \) are \( 0.735 \pm 0.005 \). (b) A gray scale slice from an MRI scan of the 1.25:1:0.8 ellipsoids in water in a spherical container.

sure the bulk density of an infinite random system, using a limited number of particles, which should be useful for packing measurements for other particle shapes. While we will pursue further MRI experiments which can provide more detailed information about interparticle correlations, the new technique can be used for all types of granular material, whether amenable to MRI investigation or not, and is experimentally simple.

While the random packing of our 1.25:1:0.8 ellipsoids at 0.74 ± 0.005 is denser than random sphere packing, as dense or denser than any sphere packing, random, lattice, or crystal, or any lattice packing of ellipsoids, it is not denser than the densest crystal packing of the same ellipsoids. From Ref. [16], we find that the densest crystal packing of these ellipsoids is at least \( \phi = 0.756 \).

This Letter addresses just one aspect of the broad and challenging subject concerning statistical geometry of hard-object packings. One still-unresolved question of interest involves shape dependence of the packing-fraction ratio for amorphous vs crystalline arrangements of identical hard particles. For spheres this ratio is \( \phi_{\text{amp}} / \phi_{\text{crys}} = 0.64/0.7405 = 0.86 \), while the corresponding value for the ellipsoid shape examined herein is substantially larger: \( \phi_{\text{amp}} / \phi_{\text{crys}} = 0.74/0.756 = 0.98 \). It will be enlightening eventually to establish the following: (a) what particle shapes yield the attainable lower and upper limits for this ratio, (b) whether the upper limit can equal or exceed unity, and (c) whether there are useful correlations between this ratio and features of the equilibrium phase diagrams for various particle shapes.

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[17] M&M’s® Candies are a registered trademark of Mars, Inc.
[21] Corrections for surface effects can be made by performing the cubic fit for a core region, e.g., for \( V(h) \) in the range \((-\frac{R}{2}) < h < (\frac{R}{2})\). If a cubic fit is used over the entire range, \(-R < h < R\), we can find the error assuming a form for \( \Delta \phi(h) \) in Eq. (1), \( \Delta \phi(h) = \beta e^{-\alpha h} \), to obtain \( V(h) = A + Bh + h^3[(\pi/3)(1-\phi_0) - 3\pi\beta/(2\alpha^2(R/a)^3) + 315\pi\beta/(2\alpha^4(R/a)^5) + O((h/a)^7)] \).
[22] For the sample core we find the nematic order parameter \( \eta = (3cos^2\Theta - 1)/2 = -0.13 \) slightly above the random value \( \eta \sim 0.05 \pm 0.03 \). There are no peaks in the measured \( S(q) \) above the simulated random \( S(q) \) values.