Improving the Density of Jammed Disordered Packings using Ellipsoids

Aleksandar Donev[1,2], Ibrahim Cisse[3,4], David Sachs[3], Evan A. Variano[3,5], Frank H. Stillinger[6], Robert Connelly[7], Salvatore Torquato[1,2,6,*], P. M. Chaikin[2,3]

[1]Program in Applied and Computational Mathematics, Princeton University, Princeton, NJ, 08544
[2]Princeton Materials Institute, Princeton, NJ, 08544
[3]Department of Physics, Princeton University, Princeton, NJ, 08544
[4]North Carolina Central University, Durham, NC, 27707
[5]Department of Civil and Environmental Engineering, Cornell University, Ithaca, NY, 14853
[6]Department of Chemistry, Princeton University, Princeton, NJ, 08544
[7]Department of Mathematics, Cornell University, Ithaca NY 14853

*To whom correspondence should be addressed; E-mail: torquato@princeton.edu.

Packing problems, such as how densely objects can fill a volume, are among the most ancient and persistent problems in mathematics and science. For equal spheres, it has only recently been proved that the face-centered cubic lattice has the highest possible packing fraction $\varphi = \pi/\sqrt{18} \approx 0.74$. It is also well-known that certain random (amorphous) jammed packings have $\varphi \approx 0.64$. Here we show experimentally and with a new simulation algorithm that ellipsoids can randomly pack more densely; up to $\varphi = 0.68 - 0.71$ for spheroids with an aspect ratio close to that of M&M'S®Candies, and even approach $\varphi \approx 0.74$ for general ellipsoids. We suggest that the higher

density is directly related to the higher number of degrees of freedom per particle and thus the larger number of particle contacts required to mechanically stabilize the packing. We support this by measurements of the number of contacts per particle Z, obtaining $Z \approx 10$ for our spheroids as compared to $Z \approx 6$ for spheres. Our results have implications for a broad range of scientific disciplines, ranging from the properties of granular media and ceramics to glass formation and discrete geometry.

The structure of liquids, crystals and glasses, and transitions between these phases is intimately related to volume fractions of ordered and disordered (random) hard-sphere packings (*1*). Packing problems (*2*) are of current interest in dimensions higher than three for insulating stored data from noise (*3*), and in two and three dimensions in relation to flow and jamming of granular materials (*4–6*) and glasses (*7*). Of particular interest is "random packing", which relates to the ancient (economically important) problem of how much grain a barrel can hold. There are many experimental and computational algorithms such that as they proceed rapidly to their limiting density, they produce a relatively robust packing fraction (relative density) $\varphi \approx 0.64$ for randomly packed monodisperse spheres (*8*). This number, widely designated as the random close packing (RCP) density, is not universal but generally depends on the packing protocol (*9*). RCP is an ill-defined concept because higher packing fractions are obtained as the systems becomes ordered, and a definition for randomness has been lacking. A more recent concept is that of the maximally random jammed (MRJ) state, corresponding to the least ordered among all jammed packings (*9*). For a variety of order metrics, it appears that the MRJ state has a density of $\varphi \approx 0.637$ and is consistent with what has traditionally been thought of as RCP (*10*). Henceforth, we will refer to this random form of packing as the MRJ state.

We report on the density of the MRJ state of ellipsoid packings as asphericity is introduced. For both oblate and prolate spheroids, φ and Z (average number of touching neighbors per particle) increase rapidly, in a cusp-like manner, as the particles deviate from perfect spheres. Both reach high densities such as $\varphi \approx 0.71$, and general ellipsoids pack randomly to a remarkable $\varphi \approx 0.735$, approaching the density of the crystal with the highest possible density for spheres (*11*) $\varphi = \pi/\sqrt{18} \approx 0.7405$. The rapid increases are unrelated to any observable increase in order in these systems that develop neither crystalline (periodic) nor liquid crystalline (nematic or orientational) order.

The experiments used two varieties of M&M'S®Milk Chocolate Candies, regular and baking or "mini" candies (*12*). Both are oblate spheroids, with small deviations from true ellipsoids, $\Delta r/r < 0.01$. Additionally, M&M'S®Candies have a very low degree of polydispersity with principle axes $2a = 1.34 \pm 0.02$ cm, $2b = 0.693 \pm 0.018$ cm, $a/b = 1.93 \pm 0.05$ for regular, and $2a = 0.925 \pm 0.011$ cm, $2b = 0.493 \pm 0.018$ cm, $a/b = 1.88 \pm 0.06$ for minis. Several sets of experiments were performed to determine the packing fraction. A square box, 8.8×8.8 cm^2 was filled to a height of 2.5 cm while shaking and tapping the container. The actual measurements were performed by adding an additional 9.0 cm to the height and excluding the contribution from the possibly layered bottom. The number of candies and their volume fraction were determined by weighing, having previously determined the average mass, density and volume of the individual candies. These experiments gave $\varphi = 0.665 \pm 0.01$ for regulars and $\varphi = 0.695 \pm 0.01$ for minis. The same technique was used for $1/8''$ ball bearings (spheres) and yielded $\varphi = 0.625 \pm 0.01$. A second set of experiments was performed by filling 0.5, 1 and 5 liter round flasks (to minimize ordering due to wall effects) with candies by pouring them into the flasks while tapping (5 liters corresponds to about 23,000 minis or 7500 regulars). Figure (1A) shows an illustration. The volume fractions found in these more reliable studies were

$\varphi = 0.685 \pm 0.01$ for both the minis and regulars (*13*). The same procedure for 30,000 ball bearings in the 0.5 liter flask gave $\varphi = 0.635 \pm 0.01$, which is close to the accepted MRJ density.

A five liter sample of regular candies similar to that shown in (Fig. 1A) was scanned in a medical MRI at Princeton Hospital. For several planar slices, the direction θ (with respect to an arbitrary axis) of the major elliptical axis was manually measured and the two-dimensional nematic order parameter, $S_2 = \langle 2\cos^2\theta - 1\rangle$ computed with the result $S_2 \approx 0.05$. This is consistent with the absence of orientational order in the packing (*14*).

Our simulation technique generalizes the Lubachevsky-Stillinger (LS) sphere-packing algorithm (*15*) to the case of ellipsoids. The method is a hard-particle molecular dynamics (MD) algorithm for producing dense disordered packings. Initially, small ellipsoids are randomly distributed and randomly oriented in a box with periodic boundary conditions and without any overlap. The ellipsoids are given velocities and their motion followed as they collide elastically and also expand uniformly. After some time a jammed state with a diverging collision rate is reached and the density reaches a maximal value. A novel event-driven MD algorithm (*16*) was used to implement this process efficiently, based on the algorithm used in Ref. (*15*) for spheres, and similar to the algorithm used for needles in Ref. (*17*). A typical configuration of 1000 oblate ellipsoids (the aspect ratio $\alpha = b/a = 1.9^{-1} \approx 0.526$) is shown in (Fig. 1B), with density of about $\varphi \approx 0.70$, and nematic order parameter $S \approx 0.02-0.05$. It is important to note that we have verified that the sphere packings produced by the LS algorithm are jammed according to the rigorous hierarchical definitions of local, collective and strict jamming (*18, 19*). Roughly speaking, these definitions are based on mechanical stability conditions that require that there be no feasible local or collective particle displacements, and/or boundary deformations. Based on our experience with spheres (*10*), we believe that our algorithm (with rapid particle expansion) produces final states that represent the MRJ state well. The algorithm closely reproduces the packing fraction measured experimentally.

The density of simulated packings of 1000 particles is shown in (Fig. 2A). Note the two clear maxima with $\varphi \approx 0.71$, already close to the 0.74 for the ordered FCC/HCP packing, and the cusp-like minimum near $\alpha = 1$ (spheres). Previous simulations for random sequential addition (RSA) (*20*), as well as gravitational deposition (*21*), produce a similarly shaped curve, with a maximum at nearly the same aspect ratios $\alpha \approx 1.5$ (prolate) or $\alpha \approx 0.67$ (oblate), but with substantially lower volume fractions (such as $\varphi \approx 0.48$ for RSA).

Why does the packing fraction initially increase as we deviate from spheres? The rapid increase in packing fraction is attributable to the expected increase in the number of contacts due to the additional rotational degrees of freedom of the ellipsoids. More contacts per particle are needed to eliminate all local and collective degrees of freedom and ensure jamming, and forming more contacts requires a denser packing of the particles. In the inset in (Fig. 2B) the central circle is locally jammed. A uniform vertical compression preserves φ but the central ellipsoid can rotate and free itself and the packing can densify. The decrease in the density for very aspherical particles could be explained by strong exclusion-volume effects in orientationally disordered packings (*22*). Results resembling (Fig. 2A) are obtained also for isotropic random packings of spherocylinders (*22, 23*), but an argument based on "caging" (not jamming) of the particles was given to explain the increase in density as asphericity is introduced. Spherocylinders have a very different behavior for ordered packings from ellipsoids (the conjectured maximal density is $\pi/\sqrt{12} \approx 0.91$ as opposed to 0.74 for nearly spherical ellipsoids), and also cannot be oblate and are always axisymmetric. The similar positioning of the maximal density peak for different packing algorithms and particle shapes indicates the relevance of a simple geometrical explanation.

By introducing orientational and translational order it is expected that the density of the packings can be further increased, at least up to 0.74. As shown in (Fig. 3) for two-dimensions, an affine deformation (stretch) of the densest disk packing produces an ellipse packing with the same volume fraction. However, this packing, though densest possible, is not strictly jammed (i.e., it is not rigid under shear transformations). The figure shows through a sequence of frames how one can distort this collectively jammed packing (*19*), traversing a whole family of densest configurations. This mechanical instability of the ellipse packing as well as the three-dimensional ellipsoid packing arises from the additional rotational degrees of freedom and does not exist for the disk or sphere packing.

There have been conjectures (*24, 25*) that frictionless random packings have just enough constraints to completely statically define the system (*26*), $Z = 2f$ (i.e., that the system is isostatic), where f is the number of degrees of freedom per particle ($f = 3$ for spheres, $f = 5$ for spheroids, and $f = 6$ for general ellipsoids) (*27*). If friction is strong, then less contacts are needed, $Z = f + 1$ (*28*). Experimentally, Z for spheres was determined by Bernal and Mason by coating a system of ball bearings with paint, draining the paint, letting it dry, and counting the number of paint spots per particle when the system was disassembled (*29*). Their results gave $Z \approx 6.4$, surprisingly close to isostaticity for frictionless spheres (*30*). We performed the same experiments with the M&M'S®, counting the number of true contacts between the particles (*31*). A histogram of the number of touching neighbors per particle for the regular candies is shown in (Fig. 4). The average number is $Z = 9.82$. In simulations a contact is typically defined by a cutoff on the gap between the particles. Fortunately, over a wide range ($10^{-9} - 10^{-4}$) of contact tolerances, Z is reasonably constant. Superposed in (Fig. 4) is the histogram of contact numbers obtained for simulated packings of oblate ellipsoids for $\alpha = 0.526$, from which we found $Z \approx 9.80$. In (Fig. 2B) we show Z as a function of aspect ratio α (*32*). As with the volume fraction, the contact number appears singular at the sphere value and rises sharply for small deviations. Unlike φ, however, Z does not decrease for large aspect ratios, but rather appears to remain constant.

We expect that fully aspherical ellipsoids, which have $f = 6$, will require even more contacts for jamming ($Z = 12$ according to the isostatic conjecture) and larger φ. Results from simulations of ellipsoids with axes $a = \alpha^{-1}$, $b = 1$ and $c = \alpha$ (where α measures the asphericity) are included in (Fig. 2A). At $\alpha \approx 1.3$ we obtain a suprisingly high density of $\varphi \approx 0.735$, with no significant orientational ordering! The maximum contact number observed in (Fig. 2B) is $Z \approx 11.4$. It is interesting to note that for both spheroids and general ellipsoids, Z reaches a constant value at approximately the aspect ratio for which the density has a maximum. This supports the claim that the decrease in density for large α is due to exclusion volume effects.

The putative nonanalytic behavior of Z and φ at $\alpha = 1$ is striking and is evidently related to the randomness of the jammed state. Crystal close packings of spheres and ellipsoids show no such singular behavior and in fact φ and Z are independent of α for small deviations from unity. On the other hand, the behavior is not discontinuous as is the number of degrees of freedom which jumps from three to five (or six) as soon as α deviates from one. In several industrial processes such as sintering and ceramic formation interest exists in increasing the density and number of contacts of powder particles to be fused. It is worth noting that using ellipsoidal instead of spherical particles, we may increase the density of a randomly poured and compacted powder to a value approaching that of the densest (FCC) lattice packing.

References and Notes

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11. The highest density is realized only by stacking variants of the FCC and HCP lattices (*33*). This is also a trivial lower bound for the maximal density of ellipsoid packings for any aspect ratio, however, it is known that higher densities are possible for sufficiently aspherical ellipsoids (*34*).

12. M&M'S®Candies are a registered trademark of Mars, Inc.

13. We estimate the correction due to the lower density at the surface of the flasks to be about 0.005.

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26. Namely, that the total number of degrees of freedom is equal to the number of impenetrability constraints (to within a constant of order one), each of which is determined by a contact between two touching particles.

27. It is also often claimed that this is the minimal number of contacts needed to ensure jamming (*24*). However, this claim is based on a counting argument that is only directly applicable to spheres, whereas handling the impenetrability constraints for ellipsoids requires including higher-order corrections due to curvature effects.

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30. More recent simulations and experiments give $Z \leq 6$.

31. Near neighbors (even when very close) leave a spot, touching neighbors leave a spot with a hole in the middle at the contact point.

32. Note that computer-generated packings can have a small percentage of “rattlers” (particles without any contacts that are not observable in our experiments), which we do not exclude when calculating Z.

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38. Continued "tapping" of the candies may further densify the system as happens for granular material (*36*). Furthermore, the somewhat higher density of the computer-generated packings can be explained by taking into account the influence of gravity and friction, which are not included in the simulation. Gravitation-dominated packings always have much lower packing fractions, as low as $\varphi \approx 0.4$, and have significant orientational ordering (*21, 37*).

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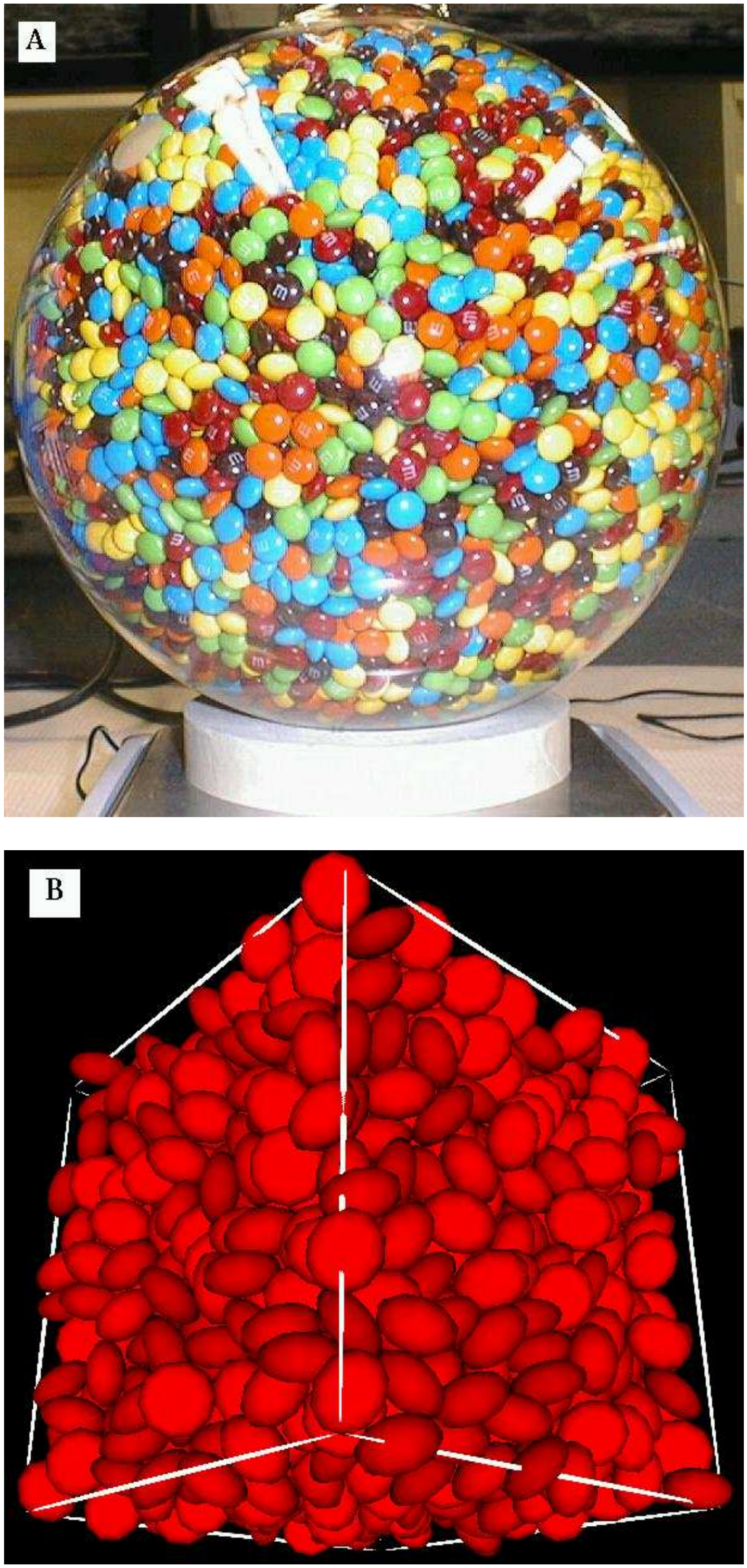
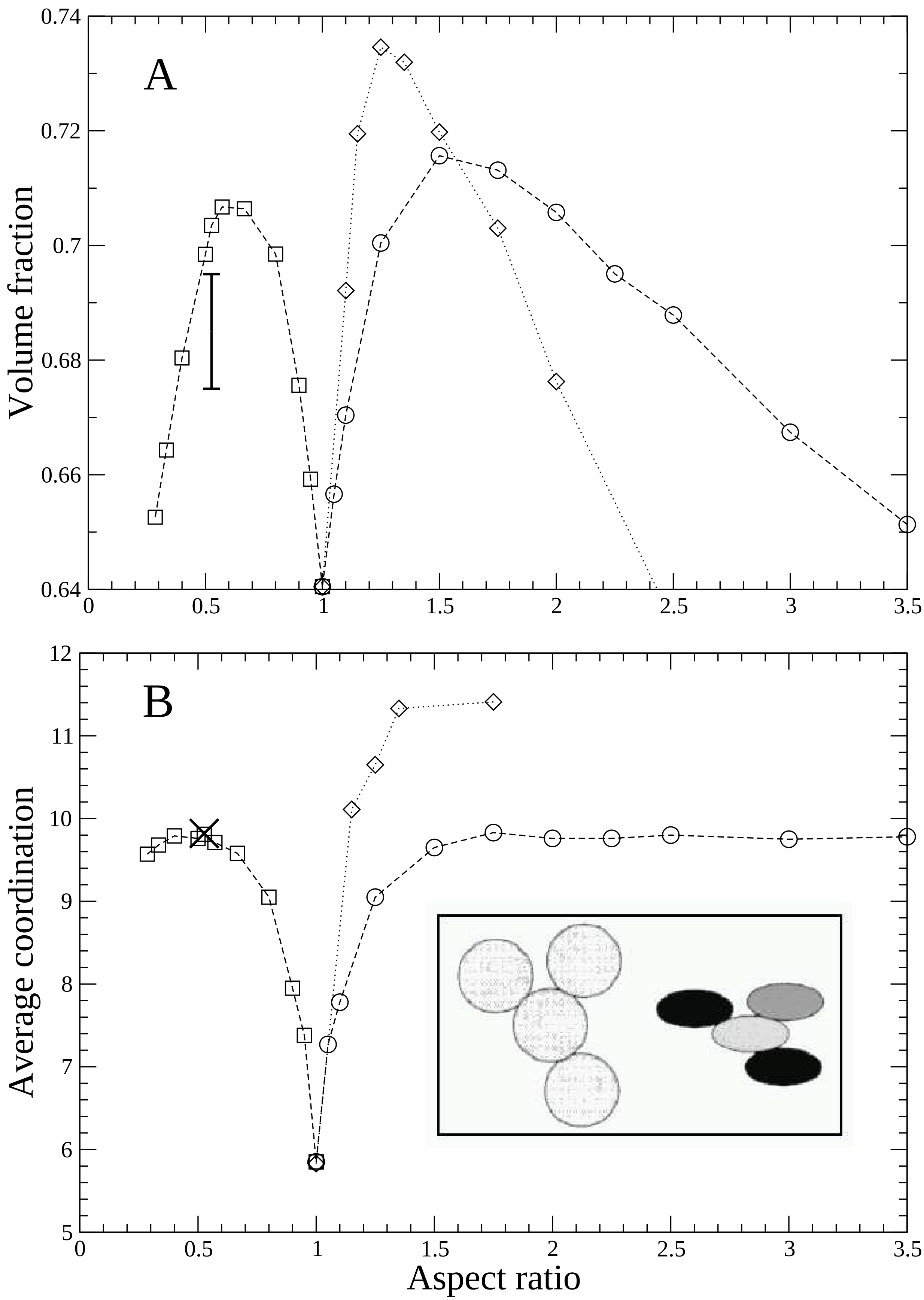
Fig. 1. *Part A (top)*: An experimental packing of the regular candies. *Part B (bottom):* Computer-generated packing of 1000 oblate ellipsoids with $\alpha = 1.9$.

Fig. 2. *Part A (top)*: Density φ versus aspect ratio α from simulations, for both prolate (circles) and oblate (squares), as well as fully aspherical (diamonds) ellipsoids. The most reliable experimental result for the regular candies (error bar) is also shown, and likely underpredicts the true density (*38*). *Part B (bottom)*: Mean contact number Z versus aspect ratio ratio α from simulations (same symbols as in part a), along with the experimental result for the regular candies (cross). *Inset*: Introducing asphericity makes a locally jammed particle free to rotate and escape the cage of neighbors.

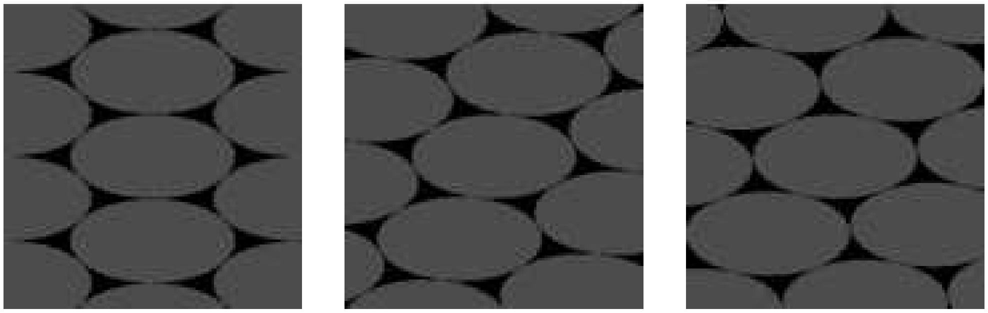
Fig. 3. Shearing the densest packing of ellipses.

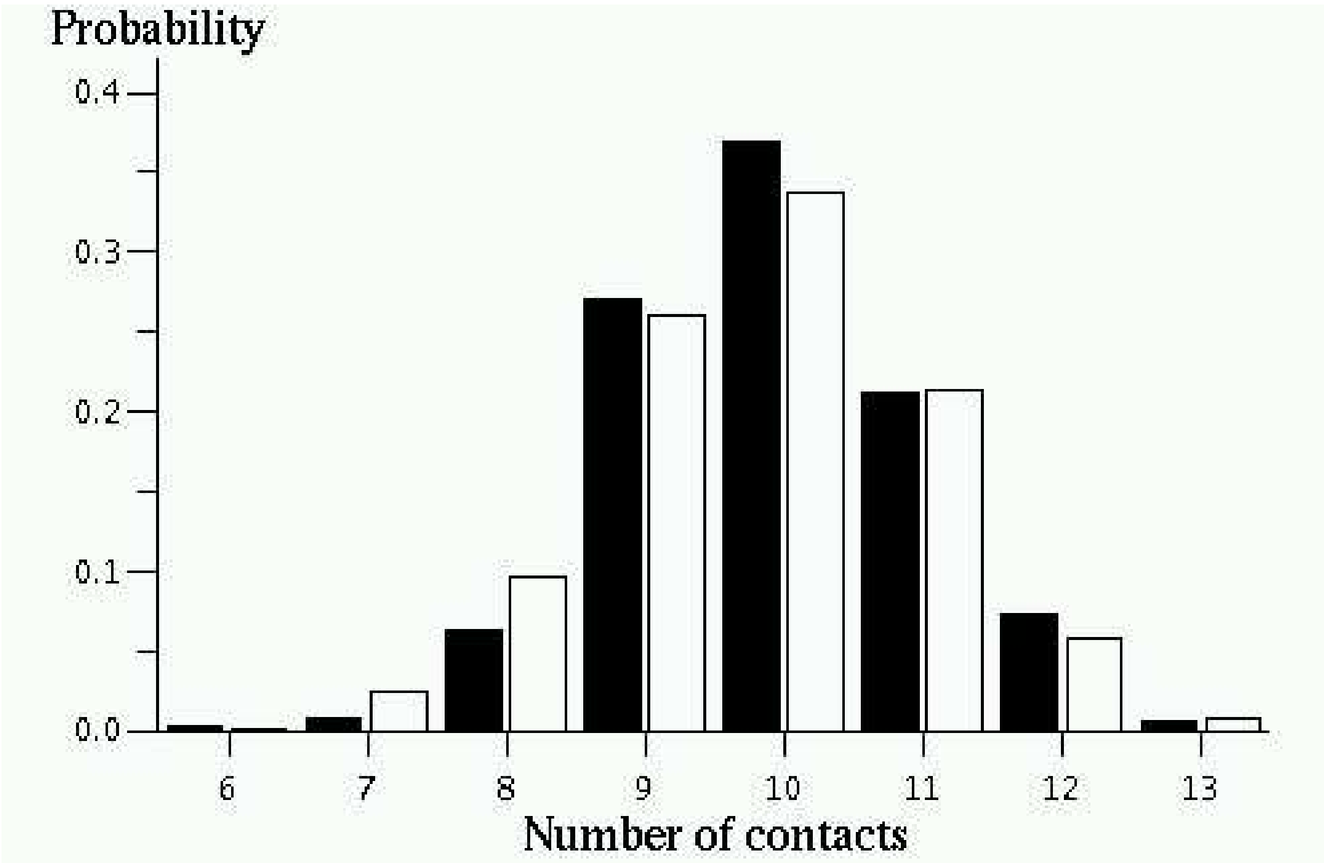
Fig. 4. Comparison of experimental (black bars, from 489 regular candies) and simulated (white bars, from 1000 particles) distribution of particle contact numbers.


A
B



A
Volume fraction
0.64
0.66
0.68
0.7
0.72
0.74
0
0.5
1
1.5
2
2.5
3
3.5
B
Average coordination
5
6
7
8
9
10
11
12
Aspect ratio




Probability
0.4
0.3
0.2
0.1
0.0
6
7
8
9
10
11
12
13
Number of contacts