Unusually Dense Crystal Packings of Ellipsoids

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In this Letter, we report on the densest-known packings of congruent ellipsoids. The family of new packings consists of crystal arrangements of spheroids with a wide range of aspect ratios, and with density φ always surpassing that of the densest Bravais lattice packing $\varphi \approx 0.7405$. A remarkable maximum density of $\phi \approx 0.7707$ is achieved for maximal aspect ratios larger than $\sqrt{3}$, when each ellipsoid has 14 touching neighbors. Our results are directly relevant to understanding the equilibrium behavior of systems of hard ellipsoids, and, in particular, the solid and glassy phases.

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Particle packing problems have fascinated people since the dawn of civilization, and continue to intrigue mathematicians and scientists. Dense packings of nonoverlapping particles have been employed to understand the structure of a variety of many-particle systems, including glasses [1], crystals [2], heterogeneous materials [3], and granular media [4]. The simple hard-sphere system is one of the most intensively studied models because it exhibits a rich thermodynamic behavior, including a well understood liquid-solid transition, and a less understood metastable liquid or glassy phase. An important extension of the hard-sphere model is to include orientational degrees of freedom for the particles, and arguably the simplest such extension is to consider systems of hard ellipsoids. Results reported in a recent paper [5] raise the question of whether the inclusion of orientational degrees of freedom can lead to a thermodynamic (as opposed to kinetic) glass phase. Answering this question necessitates a knowledge of the disordered and ordered phases at very high densities, and, in particular, the densest possible phases. A system in which the density of a disordered phase surpasses the density of the ordered solid would be a candidate for the elusive thermodynamic glass.

In addition to being important for understanding the physics of complex materials, finding the densest packing for a given particle shape is a basic problem in geometry. The famous Kepler conjecture postulates that the densest packing of spheres in three-dimensional Euclidean space has a packing fraction (density) $\phi = \pi/\sqrt{18} \approx 0.7405$, as realized by stacking variants of the face-centered cubic (fcc) lattice packing. It is only recently that this conjecture has been proven [6]. Very little is known about the most efficient packings of convex congruent particles that do not tile three-dimensional space [7]. The only other known optimal three-dimensional result involves infinitely long circular cylindrical particles: The maximal packing density $\phi_{\max} = \pi/\sqrt{12}$ is attained by arranging the cylinders in parallel in the triangular lattice arrangement [8]. Of particular interest are dense packings of congruent ellipsoids (an affine deformation of a sphere) with semiaxes a, b, and c or, equivalently, with aspect ratios $\alpha = b/a$ and $\beta = c/a$. The ratio of the largest to the smallest semiaxes, denoted by δ, is the most important aspect ratio of the ellipsoid. We call δ the *maximal aspect ratio.*

In two dimensions, it can easily be shown that the densest packing of congruent ellipses has the same density as the densest packing of circles, $\phi = \pi/\sqrt{12} \approx 0.9069$ [9]. This maximal density is realized by an affine (linear) transformation of the triangular lattice of circles. Such a transformation leaves the density unchanged. In three dimensions attempts at increasing the packing density yield some interesting structures, at least for needle-like ellipsoids. By inserting very elongated ellipsoids into cylindrical void channels passing through the ellipsoidal analogs of the densest ordered sphere packings (an affinely deformed face-centered cubic or hexagonal close packed lattice), congruent ellipsoid packings have been constructed whose density exceeds 0.7405 and approaches 0.7585 in the limit of infinitely thin prolate spheroids (ellipsoids of revolution), i.e., when $\beta = 1$ and $\alpha \rightarrow \infty$ [8,10].

However, there appears to be a widespread belief that for nearly spherical ellipsoids the highest packing fraction is realized by an affine transformation (stretch by α and β along two perpendicular axes) of the densest sphere packing, preserving the density at 0.7405. Mathematicians have often focused on *lattice packings*, where a single particle is replicated periodically on a lattice to obtain a crystal packing. For ellipsoids, a lattice packing is just an affine transformation of a sphere packing, and therefore a theorem due to Gauss [7] enables us to conclude that the densest lattice ellipsoid packing has $\phi \approx 0.7405$. The next level of generality involves nonlattice periodic packings (lattice packings with a multiparticle basis), where a unit cell consisting of several ellipsoids with at least two *inequivalent* orientations is

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periodically replicated on a lattice to fill Euclidean space. We will refer to these as *crystal packings*.

In this Letter, we report on a family of crystal packings of ellipsoids that are denser than the densest Bravais lattice packing for a wide range of aspect ratios in the vicinity of the sphere point $\alpha = \beta = 1$, and for certain aspect ratios yields the densest-known ellipsoid packings with $\phi \approx 0.7707$.

We recently developed a molecular dynamics technique for generating dense random packings of hard ellipsoids [11]. The simulation technique generalizes the Lubachevsky-Stillinger (LS) sphere-packing algorithm [12] to the case of ellipsoids. 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, while the unit cell deforms to better accommodate the packing. After some time, a jammed state with a diverging collision rate is reached and the density reaches a maximal value.

Using this technique, we generated nonequilibrium random close packings of ellipsoids [5], believed to closely represent the maximally random jammed (MRJ) state [13]. The density of the resulting packings for nonspheroidal ellipsoids with $\beta = \alpha^{-1}$ is illustrated in Fig. 1, and it can be seen that for $\alpha \approx 1.25$ ($\beta \approx 0.8$) the random packings have a density as high as 0.735, surprisingly close to what we believed was the densest ordered packing (stretched fcc lattice). This unexpected result brought into question what the maximal density really was for those aspect ratios. Extensive experience with spheres has shown that, for reasonably large packings, sufficiently slowing down the growth of the density, so that the hard-particle system remains close to the equilibrium solid branch of the equation of state, leads to packings near the fcc lattice [13,14]. This, however, requires impractically long simulation times for large ellipsoid packings. By running the simulation for very small unit cells, from 4 to 16 particles per unit cell, we were able to identify crystal packings significantly denser than the fcc lattice, and subsequent analytical calculations suggested by the simulation results led us to discover ellipsoid packings with a remarkably high density of $\phi \approx 0.7707$. This result implies that, among all possible choices of congruent ellipsoids, the maximum density attainable is bounded from below by 0.7707.

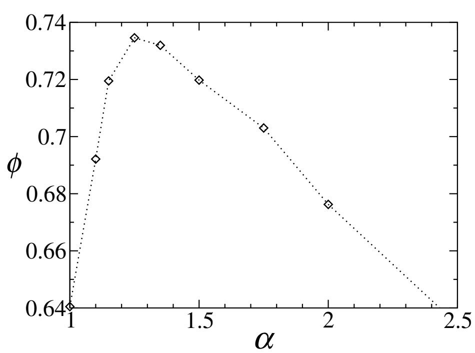

FIG. 1. The packing fraction of the putative MRJ state for nonspheroidal ellipsoids with semiaxes of ratios $1{:}\alpha{:}\alpha^{-1}$ [5]. The maximal density reached is about $\phi \approx 0.735$, which is remarkably close to the density of the fcc crystal.

We now describe the construction of a family of unusually dense crystal packings of ellipsoids. We start from the fcc lattice, viewed as a laminate of face-centered square planar layers of spheres, as illustrated in Fig. 2(a). We similarly construct layers from the ellipsoids by orienting the c semiaxis perpendicular to the layer, while orienting the a and b axes along the axes of the face-centered square lattice defining the layer, as shown in Fig. 2(b). In this process, we maintain the aspect ratio of the squares of side L of the face-centered square lattice defining the layer, i.e., we maintain

$$L = \frac{4\alpha}{\sqrt{1+\alpha^2}}, \tag{1}$$

which enables us to rotate the next layer by $\pi/2$ and fit it exactly in the holes formed by the first layer. This two-layer lamination is then continued *ad infinitum* to fill all space. This can be viewed as a family of crystal packings with a unit cell containing two ellipsoids.

We can calculate the minimal distance h between two successive layers (that preserves impenetrability) from the condition that each ellipsoid touches four other ellipsoids in each of the layers above and below it. This gives a simple system of equations (two quadratic equations and one quartic equation), the solution of which determines the density to be

$$\phi = \frac{16\pi\alpha\beta}{3hL^2}. \tag{2}$$

The axis perpendicular to the layers can be scaled arbitrarily, without changing the density, because h has the form $\beta f(\alpha)$. We can therefore just consider spheroids with $\beta = 1$. The density of this crystal packing as a function of the aspect ratio α is shown in Fig. 3, and is higher than the density of the fcc sphere packing for a wide range of aspect ratios around the sphere point $\alpha = \beta = 1$, symmetrical with respect to the inversion of α between prolate and oblate ellipsoids (we consider the prolate case in the equations in this section), and quadratic around the sphere point [15]. Two sharp maxima with density of about 0.770 732 are observed when the ellipsoids in the face-centered layers touch six rather than four in-plane neighbors, as shown in Fig. 4, i.e., when $L = 2\alpha$. This corresponds to an in-plane aspect ratio of $\sqrt{3}$, i.e., $\alpha = \sqrt{3}$ for the prolate and $\alpha = 1/\sqrt{3}$ for the oblate case. These two densest-known packings of spheroids are illustrated in the insets in Fig. 3, and in these special packings each ellipsoid touches exactly 14 neighboring ellipsoids (compare this to 12 for the fcc lattice). As illustrated in Ref. [5], an affine deformation of the densest sphere packing gives an ellipsoid packing that is *not* strictly

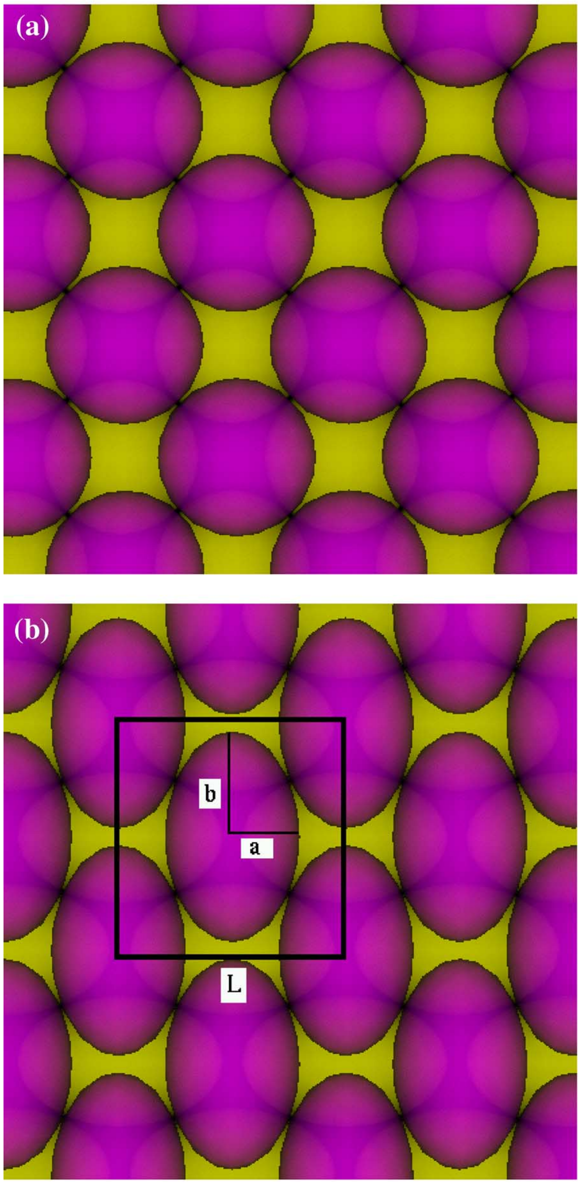

FIG. 2 (color online). Part (a) (top): The face-centered cubic packing of spheres, viewed as a laminate of face-centered layers [in the (001) plane]. The bottom layer is colored purple and the top layer yellow. Part (b) (bottom): A nonlattice layered packing of ellipsoids based on the fcc packing of spheres, but with a higher packing fraction.

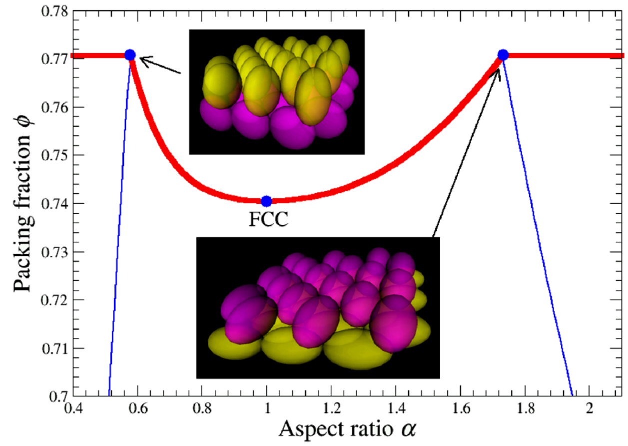

FIG. 3 (color online). The density of the laminate crystal packing of ellipsoids as a function of the aspect ratio α ($\beta = 1$). The point $\alpha = 1$ corresponding to the fcc lattice sphere packing is shown, along with the two sharp maxima in the density for prolate ellipsoids with $\alpha = \sqrt{3}$ and oblate ellipsoids with $\alpha = 1/\sqrt{3}$, as illustrated in the insets. The presently maximal achievable density is highlighted with a thicker line, and is constant for $\delta \geq \sqrt{3}$, as explained in the text.

jammed [16,17]. It is an interesting open question whether our denser laminated crystals are strictly jammed.

Figure 3 shows a rapid decrease in the packing fraction for large maximal aspect ratios. However, it is a surprising fact that the maximal density of 0.770 732 is also achievable whenever the maximal aspect ratio δ of the ellipsoids is greater than or equal to $\sqrt{3}$. The key observation is that the $x = y$ plane is a mirror symmetry plane in the above packings, so that an affine stretch by an arbitrary factor $s \geq 1$ along a direction in this plane will produce a packing of equal (stretched) ellipsoids, without changing the density. Stretching an ellipse with $\delta = \sqrt{3}$ by a factor of s along the $x = y$ line produces an ellipse with aspect ratio

$$\delta^2 = \frac{(2 + s^2 + 2s^4) + 2(1 + s^2)\sqrt{1 - s^2 + s^4}}{3s^2}, \quad (3)$$

which always gives $\delta \geq \sqrt{3}$ and achieves arbitrarily large values for sufficiently large s. Therefore, by stretching the packing in Fig. 4 along the $(\sqrt{2}/2, \sqrt{2}/2, 0)$ direction, we can obtain a packing with density 0.770 732 for any maximal aspect ratio δ larger than

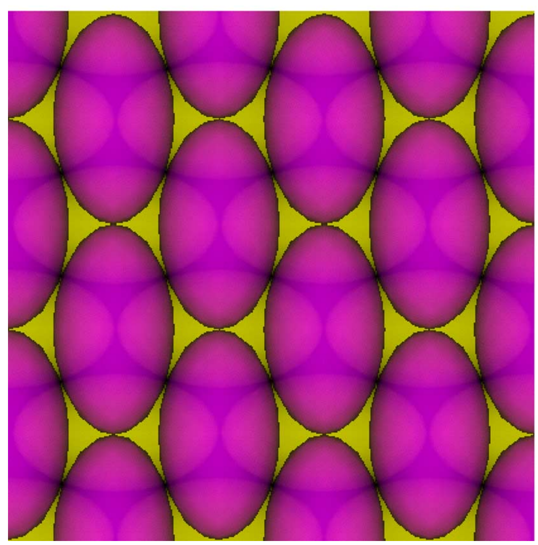

FIG. 4 (color online). The layers of the densest known packing of ellipsoids with aspect ratio $\sqrt{3}$, as illustrated in Fig. 3. The same perpendicular view applies for both prolate and oblate particles. The layers can be viewed as either face centered or triangular.

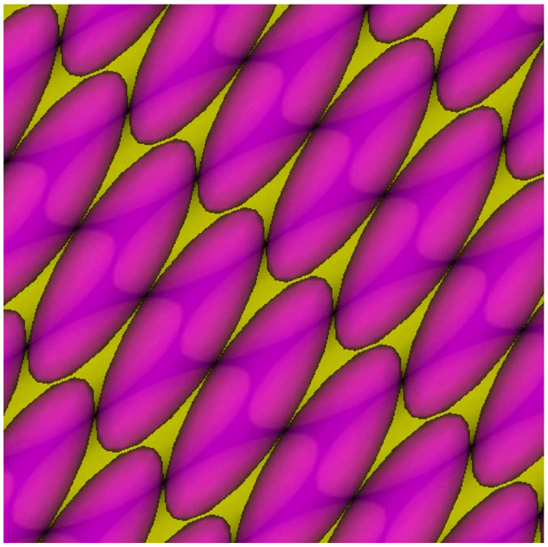

FIG. 5 (color online). The layers of the densest known packing of ellipsoids with maximal aspect ratio $\delta = 3$, as obtained by stretching the packing from Fig. 4 along the $(\sqrt{2}/2, \sqrt{2}/2, 0)$ direction by a factor of 2.4842. The same perpendicular view applies for both prolate and oblate particles.

$\sqrt{3}$ (see Fig. 3). The layers of such a packing of ellipsoids with $\delta = 3$ are illustrated in Fig. 5. As before, scaling the axis perpendicular to the layers can be used to go between the prolate and oblate cases since the c semiaxes remains aligned with the z axes. Notice that the initial stretch can be along a direction with a nonzero z component, which produces alternative packings with the same density and $\delta \geq \sqrt{3}$. The above stretch cannot be used to decrease the aspect ratio so that, for $\delta < \sqrt{3}$, our best results remain as shown in Fig. 3. In the limit of infinitely large stretch s (or infinitely large maximal aspect ratio δ), the particles *approach* perfect alignment that are either needlelike or platelike ellipsoids. However, the packings remain nonlattice arrangements with 14 contacts per particle and a density of 0.770 732.

There is nothing to suggest that the crystal packing we have presented here is indeed the densest for any aspect ratio other than the trivial case of spheres. We believe it is important to identify the densest periodic packings of ellipsoids with small numbers of ellipsoids per unit cell. This may be done using modern global optimization techniques, as has been done for various sphere and disk packing problems. However, this is a challenging project due to the complexity of the nonlinear impenetrability constraints between ellipsoids. In particular, the case of slightly aspherical ellipsoids is very interesting, as the best packing will be a perturbation of the fcc lattice with a broken symmetry, and should thus be easier to identify. In Fig. 3, we see that the density of our crystal packing increases smoothly as asphericity is introduced, unlike for random packings, where a cusplike increase is observed near $\alpha = 1$ [5]. Is there a crystal packing which leads to a sharp increase in density for slightly aspherical ellipsoids? Our initial attempts to answer this question using global optimization have not found such a crystal packing, but have not ruled out the possibility either. Further multidisciplinary investigations are needed to answer this and related questions. The results of such investigations could be used to formulate a Kepler-like conjecture for ellipsoids and understand the high-density phase behavior of the hard-ellipsoid system.

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