Packing Nonspherical Particles: All Shapes Are Not Created Equal

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Geometric Structure Approach to Jammed Particle Packings

Torquato & Stillinger, Rev. Mod. Phys. (2010)
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Order Maps for Jammed Sphere Packings
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Order Maps for Jammed Sphere Packings

A: $Z = 7$

MRJ: $Z = 6$ (isostatic)

B: $Z = 12$

• MRJ packings are hyperuniform with quasi-long-range pair correlations with decay $1/r^4$. 
3D Hard Spheres in Equilibrium

Torquato & Stillinger, Rev. Mod. Phys. (2010)
Dense Packings of Nonspherical Particles in $\mathbb{R}^3$

Granular Media

Bucky Ball: $C_{60}$

Truncated Icosahedron
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Granular Media  |  Bucky Ball: $C_{60}$  |  Truncated Icosahedron

Definitions

A collection of nonoverlapping congruent particles in $d$-dimensional Euclidean space $\mathbb{R}^d$ is called a packing $P$.

The density $\phi(P)$ of a packing is the fraction of space $\mathbb{R}^d$ covered by the particles.

Lattice packing $\equiv$ a packing in which particle centroids are specified by integer linear combinations of basis (linearly independent) vectors. The space $\mathbb{R}^d$ can be geometrically divided into identical regions $F$ called fundamental cells, each of which contains just one particle centroid. For example, in $\mathbb{R}^2$:

Thus, if each particle has volume $v_1$:

$$\phi = \frac{v_1}{\text{Vol}(F)}.$$
Definitions

A periodic packing is obtained by placing a fixed nonoverlapping configuration of $N$ particles in each fundamental cell.

Thus, the density is

$$\phi = \frac{N v_1}{\text{Vol}(F)}.$$
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Thus, the density is

$$\phi = \frac{N \nu_1}{\text{Vol}(F)}.$$

A particle is **centrally symmetric** if it has a center $C$ that bisects every chord through $C$ connecting any two boundary points.
Denote by $\sigma^2(R) \equiv \langle N^2(R) \rangle - \langle N(R) \rangle^2$ the number variance.

For a Poisson point pattern and many correlated point patterns, $\sigma^2(R) \sim R^d$.

We call point patterns whose variance grows more slowly than $R^d$ hyperuniform (infinite-wavelength fluctuation vanish). This implies that structure factor $S(k) \to 0$ for $k \to 0$.

The hyperuniformity concept enables us to classify crystals and quasicrystals together with special disordered point processes.

All crystals and quasicrystals are hyperuniform such that $\sigma^2(R) \sim R^{d-1}$ – number variance grows like window surface area.

Many different MRJ particle packings are hyperuniform with $S(k) \sim k$ for $k \to 0$.

Donev, Stillinger & Torquato, 2005; Berthier et al., 2011; Zachary, Jiao & Torquato, 2011; Kurita and Weeks, 2011.
Outline

- Organizing principles for maximally dense packings of nonspherical particles.
- Organizing principles for MRJ packings of nonspherical particles (e.g., isostatic or not; hyperuniformity, etc.).
- Tunability capability via particle shape to design novel crystal, liquid and glassy states.
Packings of the Platonic and Archimedean Solids

- Difficulty in obtaining maximally dense packings of polyhedra: complex rotational degrees of freedom and non-smooth shapes.
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Adaptive Shrinking Cell

- Optimization scheme that explores many-particle configurational space and the space of lattices to obtain a local or global maximal density.

ASC scheme can be solved using a variety of techniques, depending on the particle shape, including MC and linear-programming methods. For spheres, the latter is very efficient [Torquato and Jiao, PRE (2010)].
Kepler-Like Conjecture for a Class of Polyhedra

- **Face-to-face contacts** allow higher packing density.

- **Central symmetry** enables maximal face-to-face contacts when particles are aligned – consistent with the optimal lattice packing.

For any packing of nonspherical particles of volume $v_{\text{particle}}$:

$$\phi_{\text{max}} \leq \phi_{\text{upper bound}} = \min \left[ \frac{v_{\text{particle}}}{v_{\text{sphere}}} \frac{\pi}{\sqrt{18}}, 1 \right],$$

where $v_{\text{sphere}}$ is the volume of the largest sphere that can be inscribed in the nonspherical particle.
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where $v_{\text{sphere}}$ is the volume of the largest sphere that can be inscribed in the nonspherical particle.

These considerations lead to the following conjecture:

*The densest packings of the centrally symmetric Platonic and Archimedean solids are given by their corresponding optimal lattice packings.*

Dense Packings of Icosahedra, Dodecahedra & Octahedra

ASC scheme with many particles per cell yield densest lattice packings for centrally Platonic solids!

Icosahedra
\[ \phi = 0.836 \]

Dodecahedra
\[ \phi = 0.904 \]

Octahedra
\[ \phi = 0.947 \]

Later showed octahedron packing leads to uncountably infinite number of tessellations by octahedra and tetrahedra (Conway, Jiao & Torquato 2010).
**Superballs**

A $d$-dimensional superball is a centrally symmetric body in $\mathbb{R}^d$ occupying

$$|x_1|^{2p} + |x_2|^{2p} + \cdots + |x_n|^{2p} \leq 1 \quad (p: \text{deformation parameter})$$

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**Superdisks**

Densest packings are lattices and behave quite differently from ellipsoid packings! Jiao, Stillinger & Torquato, PRL (2008); PRE (2009)
Maximally dense packings are certain families of **lattices** for $p \geq 1/2$.

Densest ellipsoid packings are **non-lattices**.

Maximal density is **nonanalytic** at the “sphere” point ($p = 1$) (in contrast to ellipsoids) and increases dramatically as $p$ moves away from unity.

**Rich phase behavior** depending on $p$ (Batten, Stillinger & Torquato 2010; Ni et al. 2012).
Conjecture 2:

The optimal packing of any convex, congruent polyhedron without central symmetry is generally not a (Bravais) lattice packing.

Regular tetrahedra cannot tile space.

Densest lattice packing (Hoylman, 1970): $\phi = 18/49 = 0.3673 \ldots$

Densest packing must be a non-lattice (Conway & Torquato, 2006). Constructed a 20-particle packing with $\phi \approx 0.72$

MRJ isostatic packings of tetrahedral dice (Chaikin et al., 2007): $\phi \approx 0.74$

Many subsequent studies improved on this density with complicated fundamental cells (Chen, 2008; Torquato & Jiao, 2009; Haji-Akbari et al. 2009).

Recently, 3 different groups (Kallus et al. 2010; Torquato and Jiao 2010; and Chen et al. 2010) have found 4-particle packings with $\phi \approx 0.86$. 
Packings of Truncated Tetrahedra

- The Archimedean truncated tetrahedron cannot tile space.
- Densest lattice packing (Betke & Henk, 2000): $\phi = \frac{207}{304} = 0.680 \ldots$
- A dense non-lattice packing with a two-particle (dimer) basis was constructed by Conway and Torquato (2006) with $\phi = \frac{23}{24} = 0.958 \ldots$

Derived analytically packing that nearly fills space: $\phi = \frac{207}{208} = 0.995 \ldots$ Can be obtained by continuously deforming the Conway-Torquato packing. It has small tetrahedral holes and is a new tessellation of space with truncated tetrahedra and tetrahedra (Jiao & Torquato, 2011).

- Two-stage melting process: optimal packing is stable at high densities and the Conway-Torquato packing is stable at lower densities upon melting.
Conjecture 3:

Optimal packings of congruent, centrally symmetric particles that do not possess three equivalent principle axes generally cannot be a Bravais lattice.

Maximally Dense Ellipsoidal Packings

- Densest known packings are non-Bravais lattices.
- With relatively small asphericity, can achieve $\phi = 0.7707$. 

![Graph showing packing fraction $\phi$ vs. aspect ratio $\alpha$.]
# Densest Known Packings of Some Convex Particles

**Table 1:** Densest Known Packings of Some Convex Particles

<table>
<thead>
<tr>
<th>Particle</th>
<th>Packing Density</th>
<th>Central Symmetry</th>
<th>Equivalent Axis</th>
<th>Structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sphere</td>
<td>0.740</td>
<td>Y</td>
<td>Y</td>
<td>Bravais Lattice</td>
</tr>
<tr>
<td>Ellipsoid</td>
<td>0.740 - 0.770</td>
<td>Y</td>
<td>N</td>
<td>Periodic, 2-particle basis</td>
</tr>
<tr>
<td>Superball</td>
<td>0.740 - 1</td>
<td>Y</td>
<td>Y</td>
<td>Bravais Lattice</td>
</tr>
<tr>
<td>Tetrahedron</td>
<td>0.856</td>
<td>N</td>
<td>Y</td>
<td>Periodic, 4-particle basis</td>
</tr>
<tr>
<td>Icosahedron</td>
<td>0.836</td>
<td>Y</td>
<td>Y</td>
<td>Bravais Lattice</td>
</tr>
<tr>
<td>Dodecahedron</td>
<td>0.904</td>
<td>Y</td>
<td>Y</td>
<td>Bravais Lattice</td>
</tr>
<tr>
<td>Octahedron</td>
<td>0.945</td>
<td>Y</td>
<td>Y</td>
<td>Bravais Lattice</td>
</tr>
<tr>
<td>Trun. Tetrah.</td>
<td>0.995</td>
<td>N</td>
<td>Y</td>
<td>Periodic, 2-particle basis</td>
</tr>
<tr>
<td>Cube</td>
<td>1</td>
<td>Y</td>
<td>Y</td>
<td>Bravais Lattice</td>
</tr>
</tbody>
</table>
Generalizations of the Organizing Principles to Concave Particles

Torquato and Jiao, PRE, 2010.

**Generalization of Conjecture 1:**

_Dense packings of centrally symmetric concave, congruent polyhedra with three equivalent axes are given by their corresponding densest lattice packings, providing a tight density lower bound that may be optimal._

**Generalization of Conjecture 2:**

_Dense packings of concave, congruent polyhedra without central symmetry are composed of centrally symmetric compound units of the polyhedra with the inversion-symmetric points lying on the densest lattice associated with the compound units, providing a tight density lower bound that may be optimal._

**Figure 1:** (a) Centrally symmetric concave octapod and the associated optimal Bravais-lattice packing. (b) Concave tetrapods without center symmetry forming a centrally symmetric dimer, which then pack on a Bravais lattice [de Graaf et al, Phys. Rev. Lett. 107, 155501 (2011)].
Nonspherical Particles and Rotational Degrees of Freedom

- **Isostatic (Isoconstrained):** Total number of contacts (constraints) equals total number of degrees of freedom. Conventionally, thought to be associated with the minimal number of constraints for rigidity and random (generic) packings.

  \[ Z = 2f \]

  *Z:* average no. of contacts/particle; *f:* degrees of freedom/particle

  \( f = 2 \) for disks, \( f = 3 \) for ellipses, \( f = 3 \) for spheres, \( f = 5 \) for spheroids, and \( f = 6 \) for general ellipsoids.

- **Hypostatic:**

  \[ Z \leq 2f \]

  Conventionally thought to be unstable.

- **Hyperstatic:**

  \[ Z \geq 2f \]

  True of ordered packings.
There is a competition between translational & rotational jamming.

Rotational degrees of freedom lead to improved density (over spheres) and allows for correlated contacts, which leads to MRJ hypostatic jammed packings.

Donev, Connelly, Stillinger & Torquato, PRE (2007)
Packing density increases monotonically as $p$ deviates from 1.

Disordered superball packings are always hypostatic and do not come close to the isostatic contact number as asphericity increases!

Isostatic disordered superball packings are difficult to construct; they require $Z = 12$, which is associated with crystal packings.
MRJ Packings of Nontiling Platonic Solids

Jiao & Torquato, PRE (2011)

- Hyperuniform with quasi-long-rang (QLR) pair correlations \(1/r^4\) and isostatic.

Figure 2: (a) Structure factor \(S(k)\) of the MRJ packings of the nontiling Platonic solids. The inset shows that \(S(k)\) is linear in \(k\) for small \(k\) values. (b) Local contacting configurations: from left to right, tetrahedra, icosahedra, dodecahedra, and octahedra.
## MRJ Packings of Nontiling Platonic Solids

### Table 2: Characteristics of MRJ packings of hard particles with different shapes.

<table>
<thead>
<tr>
<th>Particle Shape</th>
<th>Isostatic</th>
<th>Hyperuniform QLR</th>
<th>MRJ Packing Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sphere</td>
<td>Yes</td>
<td>Yes</td>
<td>0.642</td>
</tr>
<tr>
<td>Ellipsoid</td>
<td>No (hypostatic)</td>
<td>Yes</td>
<td>0.642 – 0.720</td>
</tr>
<tr>
<td>Superball</td>
<td>No (hypostatic)</td>
<td>Yes</td>
<td>0.642 – 0.674</td>
</tr>
<tr>
<td>Superellipsoid</td>
<td>No (hypostatic)</td>
<td>Yes</td>
<td>0.642 – 0.758</td>
</tr>
<tr>
<td>Octahedron</td>
<td>Yes</td>
<td>Yes</td>
<td>0.697</td>
</tr>
<tr>
<td>Icosahedron</td>
<td>Yes</td>
<td>Yes</td>
<td>0.707</td>
</tr>
<tr>
<td>Dodecahedron</td>
<td>Yes</td>
<td>Yes</td>
<td>0.716</td>
</tr>
<tr>
<td>Tetrahedron</td>
<td>Yes</td>
<td>Yes</td>
<td>0.763</td>
</tr>
</tbody>
</table>
Generalization of Hyperuniformity to Polydisperse Systems

- Structure factor of the MRJ packing of polydisperse particles does not vanish at $k = 0$ (Kurita & Weeks, PRE 2010; Berthier et al., PRL 2011; Zachary, Jiao, & Torquato, PRL 2011).

- Introduced a more general notion of hyperuniformity involving local-volume-fraction fluctuations and associated spectral function $\tilde{\chi}(k)$ for general two-phase media (packings or not) (Zachary & Torquato, J. Stat. Mech. 2009).

- We have shown that MRJ packings of hard-particles are hyperuniform with QLR correlations (i.e., $\tilde{\chi}(k) \rightarrow 0$ as $k \rightarrow 0$), regardless of the particle shapes or relative sizes (Zachary, Jiao & Torquato, PRL 2011; PRE 2011).
CONCLUSIONS

- Non-spherical particles are not created equal! Changing the shape of a particle can dramatically alter its packing attributes.

- We now have some organizing principles for both maximally dense and MRJ packings of nonspherical particles.

- Tunability capability via particle shape could be used to tailor many-particle systems (e.g., colloids and granular media) to have designed crystal, liquid and glassy states.

Collaborators

- Robert Batten, Princeton
- Robert Connelly, Cornell
- John Conway, Princeton
- Paul Chaikin, NYU
- Aleks Donev, Princeton/Courant
- Yang Jiao, Princeton
- Frank Stillinger, Princeton
- Chase Zachary, Princeton

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