PARTICLE PACKING PROBLEMS IN LOW SPACE DIMENSIONS

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**Hard-Sphere Model**

- Good starting point to describe structure of diverse materials:
  - particulate composites
  - granular media
  - colloids
  - liquids
  - glasses
  - crystals

- Useful model to understand phase transitions (equilibrium) and jamming (nonequilibrium).

- Jamming concepts are intimately related to rigidity transitions in granular media and dynamical arrest in glasses.

- Hard-sphere problems are ancient and hard! Still many unresolved conundrums concerning the structure.
Why Hard Spheres?

Intimately related to classical ground states and glasses.

Most interesting things depend on repulsive part
simplest repulsion is:

\[ U(r) \begin{cases} \infty, & |r| < a \\ 0, & |r| \geq a \end{cases} \]

The essence of the problem -- Hard Spheres
• A quenched liquid stops flowing at a glass transition - system becomes “jammed” on experimental time scales.
• A glass has the disordered structure of a liquid but has the rigidity of a solid.
• For the athermal hard-sphere system, a glass can be created by rapidly increasing the density (like decreasing T).
Growth Algorithm to Make a Hard-Sphere Glass (3D)

VRML animation
OUTLINE

1. Maximally Dense Sphere Packings

2. Is Random Close Packing Well-Defined?

3. To Jam or Not to Jam?

4. Can randomness be quantified? Order Metrics

5. Unified View of Optimal Packings

6. Optimal Packings of Non-Spherical Objects
Definitions

• **Packing** ≡ A collection of congruent copies of a convex particle K in d-dimensional Euclidean space \( \mathbb{R}^d \) such that no two particles overlap.

• **Packing density**, \( \phi \) ≡ The fraction of \( \mathbb{R}^d \) covered by particles.

• **Lattice Packing** ≡ Packing of translates of K such that the translation vectors form a lattice (set of all integral combinations of basis vectors).

• **Periodic packing** ≡ Packing obtained by placing a fixed arrangement of \( n \ (\geq 1) \) convex particles in the fundamental cell.

• **Random packing** ≡ Packing in which pair correlation function \( g_2(r) \) decays to unity sufficiently rapidly.
Maximally Dense Monodisperse Packings

• In 2D, solution is **triangular** lattice (Fejes Toth, 1940).

  \[ \phi_{\text{max}} = \frac{\pi}{\sqrt{12}} = 0.9058... \]

• In 3D, Kepler (1606) conjectured that optimal packing is **FCC lattice** \( \phi_{\text{max}} = \frac{\pi}{18^{1/2}} = 0.74048... \) (Hales, 1998; 2004).

• **Polydispersity**? Other particle **shapes**? Higher D?
Pair Correlation Function $g(r)$

- $g_2(r) \equiv$ prob. density function associated with finding a particle at a radial distance $r$ from a given particle

- Typical “random” jammed packing of spheres of unit diameter in $R^3$
Random Packings of Hard Spheres

- Bernal: `In closing we must not forget the commentary on random packing which Saint Luke attributes to Jesus, “Give and it will be given unto you; good measure, pressed down, and shaken together, and running over … For by your standard of measure it will be measured to you in return.”`

- Prevailing 50-year old view: random close packed (RCP) state is the maximum density that a large, random collection of spheres can attain and is a “universal” quantity - \( \phi \approx 0.64 \) for \( d=3 \) (\( \phi \approx 0.82 \) for \( d=2 \)).

Is Random Close Packing of Spheres Well Defined?

NO! Torquato, Truskett & Debenedetti, PRL (2000)

- What is “random”? Random and “close-packing” are contradictory terms.

- Shown by defining “jamming” and “order metrics”, and generating realistic hard-sphere packings via computer simulations.

- To replace the RCP state, we introduce a new concept: the maximally random jammed (MRJ) state, which can be made precise.
Problems with RCP

• **Dynamical parameters:** pouring rate, and amplitude and frequency of vibration. **Interactions:** interparticle forces, friction (inhibiting densification), and gravity.

• $\phi_c$ value is protocol dependent.

• Terms “random” and “close packed” are at odds with one another.

• There has never been a **rigorous** determination of the RCP packing density.

**GEOMETRIC APPROACH THAT EXAMINES INDIVIDUAL CONFIGURATIONS IS REQUIRED.**
Jamming Categories

Torquato and Stillinger (2001)

• **Locally jammed:** Each particle in the system is individually jammed (d+1 contacting spheres not all in the same hemisphere), while fixing the positions of the remaining particles.

• **Collectively jammed:** A locally jammed configuration in which no finite subset of the particles can be continuously displaced, so that its members move out of contact with the remainder set.

• **Strictly jammed:** A collectively jammed configuration that disallows all uniform volume-nonincreasing deformations.

*Boundary conditions matter!*

Jamming can be tested rigorously!
To Jam or Not to Jam? -- Linear Programming

(Donev, Torquato, Stillinger, & Connelly 2003)

Displacement Formulation:

\[
\begin{align*}
\text{max}_{\Delta r} & \quad b^T \Delta r \quad \text{(virtual work)} \\
\text{s.t.} & \quad A^T \Delta r \leq \Delta l \quad \text{(impenetrability)}
\end{align*}
\]

Force Formulation:

\[
\begin{align*}
\text{max}_f & \quad (\Delta l)^T f \quad \text{(virtual work)} \\
\text{s.t.} & \quad Af = b \quad \text{(static equilibrium)} \\
\text{and} & \quad f \leq 0 \quad \text{(repulsion only)}
\end{align*}
\]

Test collective and strict jamming: Use only 1 random load \( b \) to find unjamming motions.
Honeycomb: Collective Unjamming (Hard-Wall BCs)

VRML animation
Quantifying Disorder/Order in Condensed Phase Systems

• Phase diagram for order: “order map” - Can use to classify disordered materials.

• Complete information is out of the question. Must settle for reduced information: set of scalar order metrics $\psi_1, \psi_2, \ldots, \psi_n$ such that $0 \leq \psi_i \leq 1, \forall i.$
“Order Map” and Optimal Packings

- **B**: Jammed state of maximal density.
- **A**: Jammed state of minimal density.
- **MRJ**: Maximally random jammed state is the one that minimizes $\psi$ among all jammed structures.
Simple Order Metrics

- Chose two basic measures of order: bond-orientational order $Q_6$ (Steinhardt et al. 1983) & translational order $T$ (Torquato et al. 2000).

- For example, for 2D

$$Q_6 = |\sum e^{i6\theta}| / N$$

- $Q_6$ measures the persistence of bond orientational order globally.

- $T$ measures the degree of spatial ordering relative to FCC lattice.

- Also considered crystal-independent order metrics.
Important Differences Between 2D and 3D Packings

- Growth algorithm yields “MRJ” packings in $\mathbb{R}^3$ that are strictly jammed with $\phi \approx 0.64$ and mean kissing no. $Z \approx 6$ (isostaticity or $2 \times$ no. of degrees of freedom per particle).

  Kansal, Torquato & Stillinger (2002)

- We have also studied MRJ 4D, 5D & 6D packings.

  Skoge, Donev, Stillinger & Torquato (2006)

- 2D “random” packings are fundamentally different from 3D-6D counterparts. The former are only collectively jammed at about $\phi=0.89$ with a high degree of crystallinity! This illustrates importance of jamming categories in classifying packings.

Conclusions

• RCP state is not well-defined mathematically. A geometric viewpoint is required!

• MRJ state is precisely defined once an order metric $\psi$ is chosen.

• This lays mathematical groundwork for studying randomness in dense particle packings as well as general point patterns.

• We have quantified order (disorder) in molecular systems, including simple liquids, water, and glasses.
MRJ Packings Have Unusual Properties

Donev, Stillinger & Torquato, PRL (2005)

- **They are saturated and hyperuniform** (large-scale density fluctuations – variance vanish), confirming a conjecture of Torquato & Stillinger (PRE, 2003).

- **Minimizing** variance is a ground-state problem and linked to an open problem in number theory.

- **Million-particle packings**: $g_2(r) \sim r^{-4}$ for large $r$ or $S(k) \sim |k|$ for small $k$, i.e., $S(k)$ is nonanalytic at origin!

- Same as the Harrison-Zeldovich spectrum for matter distribution in early Universe and ground states of spin-polarized fermions.
• Discovered an infinite family of 3D crystal packings, which are subpackings of the densest packings, strictly jammed and have anomalously low density (high concentration of self-avoiding “tunnels” that permeate the structures). Torquato & Stillinger (2007)

• Burnell & Sondhi (2008) – studied antiferromagnetic spin interactions on such geometrically frustrated crystals.
Collisional Jamming Stress (Force Chains)


OTHER PARTICLE SHAPES IN $\mathbb{R}^3$

Ellipsoids (Donev et. al., Science 2004)
No orientational order

Nematic order is essentially zero.

Torquato, Chaikin
Ellipse and Ellipsoid Packings

First event-driven MD to generate dense ellipsoid packings
Ellipsoids Density

$\alpha : 1 : \frac{1}{\alpha}$

oblate

prolate

Volume fraction

Aspect ratio, $\alpha$
• Is the FCC ellipsoidal crystal the densest? No!
• Found densest known ellipsoidal crystals: $\phi = 0.77\ldots$
Density of a Family of Crystal Packings

Kissing number = 14
Densest Ellipse Packing Can Be Sheared!

Not True for the Densest Circle Packing!
Packing Regular Tetrahedra

• Ulam’s 1972 Conjecture: Among all densest packings of convex congruent objects in 3-space, sphere gives lowest density.

• Our ellipsoid results are consistent with this conjecture but what about the regular tetrahedron?

• Best packing is not a lattice packing, which has $\phi=0.367…$

• Densest known packing has $\phi \approx 0.72$

Conway & Torquato, PNAS (2006)
Densest Packings of Superdisks

• A superdisk is a 2D shape defined by
  \[ |x_1|^{2p} + |x_2|^{2p} \leq 1 \]

convex shapes \((p \geq 0.5)\) & concave shapes \((0 < p < 0.5)\)

\[ \begin{align*}
  &\text{p = 0.45} & \text{p = 0.75} & \text{p = 1.0} & \text{p = 2.0} \\
\end{align*} \]

• We have found exact constructions for densest known superdisk packings for all convex and concave cases.

Jiao, Stillinger & Torquato, PRL (2008)
Role of Broken Rotational Symmetry
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