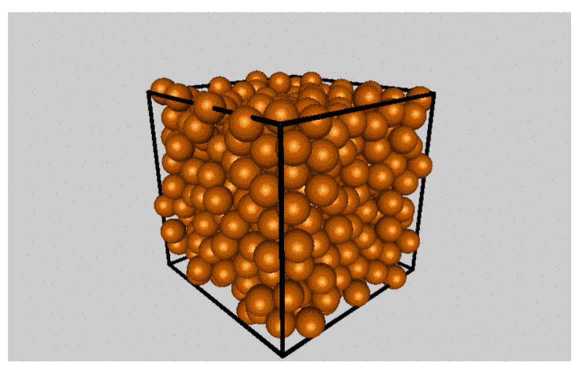
JAMMING IN OPTIMAL PARTICLE PACKINGS

Salvatore Torquato

Department of Chemistry, Materials Institute, and Program in Applied & Computational Mathematics
Princeton University

http://cherrypit.princeton.edu

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.**

OUTLINE

1. Is Random Close Packing Well-Defined?

2. Order Metrics and Optimal Packings

3. To Jam or Not to Jam?

4. Characteristics of the MRJ state

5. Maximally Dense Particle Packings

6. High Dimensions: Cracking a 100-Year Old Problem

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 - $\phi c \approx 0.64$ for d=3 ($\phi c \approx 0.82$ for d=2).
- What is "random"? Random and "close-packing" are contradictory terms.

Is Random Close Packing of Spheres Well Defined?

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

- Shown by defining "order metrics" and "jamming", and generating 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.

Modified Lubachevsky-Stillinger Growth Algorithm (3D)

VRML animation

Jamming Categories

Torquato and Stillinger, J. Phys. Chem. B (2001)

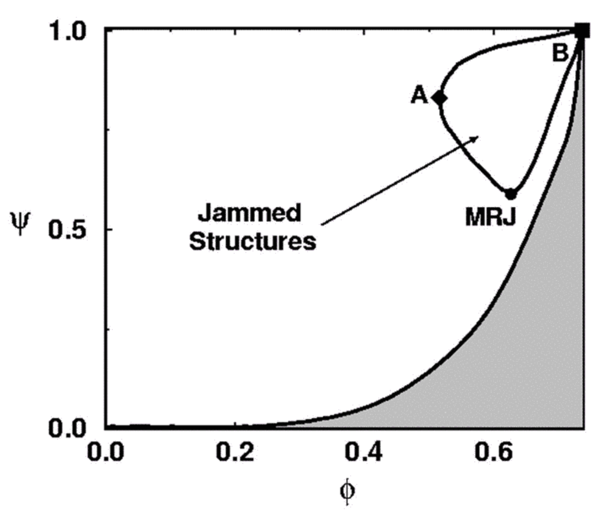
- **Locally jammed:** Each particle in the system is **individually** jammed (**d+1** contacting spheres not all in the same hemisphere), while **fixing** the positions of the remaining particles.
- **Collectively jammed:** A locally jammed configuration in which no **subset** of the particles can be **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!

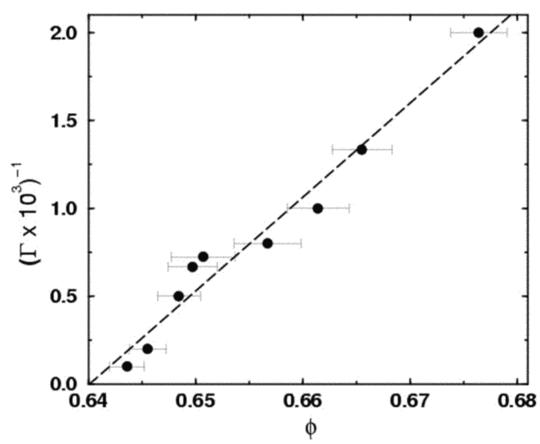
Honeycomb: Collective Unjamming (Hard-Wall BCs)

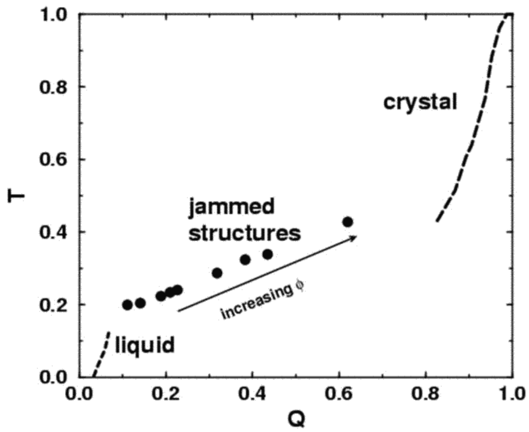
VRML animation

"Order Map" and Optimal Packings



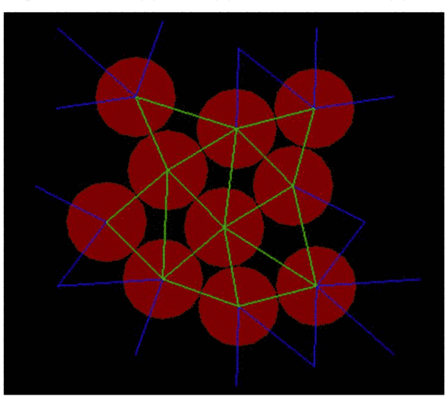
- **B: Jammed state of maximal density.**
- **A: Jammed state of minimal density.**
- **MRJ: Maximally random jammed state is the one that minimizes ψ among all jammed structures.**





To Jam or Not to Jam? -- Linear Programming

(Donev, Torquato, Stillinger, & Connelly 2003)



$G(p)$ (strut framework) = G (graph) + p (embedding)

contact network = ***connectivity*** + ***geometry***

Displacement Formulation:

$\max_{\Delta r} b^T \Delta r$ (*virtual work*)

s.t. $A^T \Delta r \le \Delta l$ (*impenetrability*)

Force Formulation:

$\max_f (\Delta l)^T f$ (*virtual work*)

s.t. $Af = b$ (*static equilibrium*)

and $f \le 0$ (*repulsion only*)

b	particle loads
Δl	interparticle gaps
A	rigidity matrix of $G(p)$
f	***contact forces***
Δr	***displacements*** (flex)

Test collective and strict jamming: Use only 1 *random* load b to find unjamming motions.

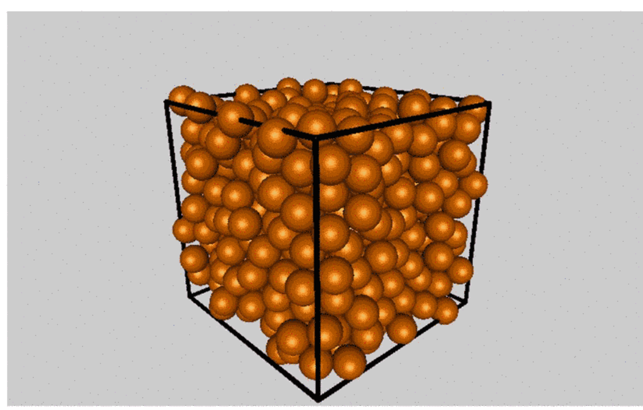
Diversity of Order and Packing Densities

Kansal, Torquato & Stillinger, PRE (2002)

- Generated a large database of strictly jammed packings (2700 packings) in R^3 using different protocols.
- Wide density range over which such packings can be created ($0.52 < \phi < 0.74$), independent of system size.
- At fixed ϕ, variation in order can be substantial. Thus, density alone is not sufficient to characterize packing, showing once again that the RCP state is ill-defined.
- Different order metrics (including local Q_6) yielded a consistent estimate of the MRJ packing density $\phi \approx 0.64$.
- Developed criteria to define more broadly applicable metrics.

Important Differences Between 2D and 3D Packings

- LS algorithm yields "random" packings in R^3 that are essentially strictly jammed at about ϕ=0.64 with a low degree of order.



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

Donev, Torquato, Stillinger, & Connelly, JAP, 2003

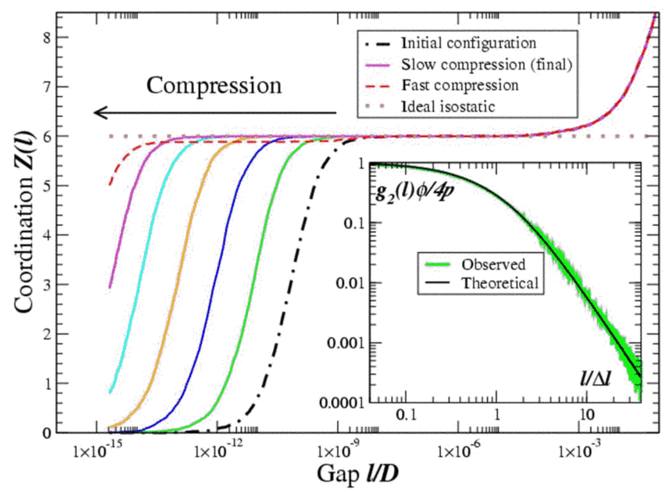
Small-r Behavior of $g_2(r)$ for MRJ Packings

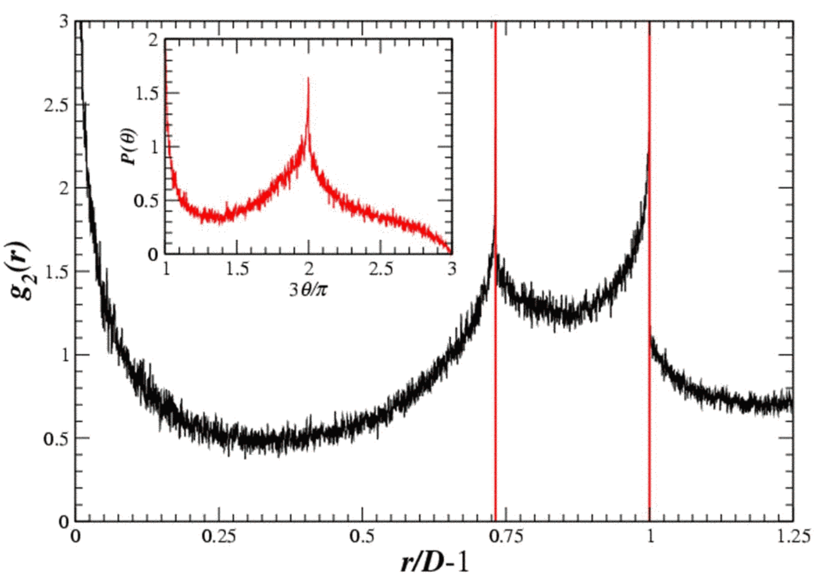
Donev, Torquato & Stillinger, PRE (2005)

- For **nearly jammed packings**, related $g_2(l)$ to $P_f(l)$, where l=r-D.
- Our numerically generated MRJ packings are strictly **isostatic**.
- As jamming is approached, we show clear separation between **δ–function** and background (**near contact**) contributions.

Cumulative coordination number

$$Z(r) = \rho 4\pi \int_D^r x^2 g_2(x) dx$$



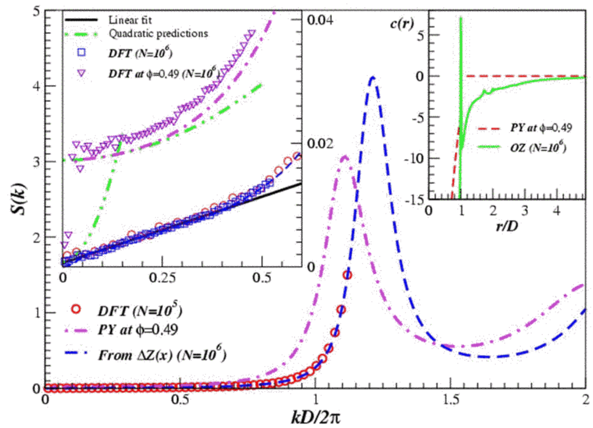


- Background contribution at contact has **divergence with exponent -0.4**, i.e., $g_2(x) \sim x^{-0.4}$ where x=r/D-1.
- **Split second peak** is **not** related to any **crystal** ordering.

Long-Wavelength Density Fluctuations of MRJ Packings

Donev, Stillinger & Torquato (2005)

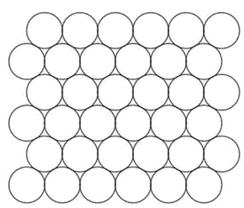
- Verified Torquato-Stillinger conjecture (2003) that MRJ state is **saturated** and **hyperuniform**.
- **Million-particle packings**: $g_2(r) \sim r^{-4}$ for large r or $S(k) \sim |k|$ for small k, i.e., S(k) is **nonanalytic** at origin!



- Same as the **Harrison-Zeldovich spectrum** for early **Universe**, described by Gabrielli et al. (2002) as "**glassy**" Universe.

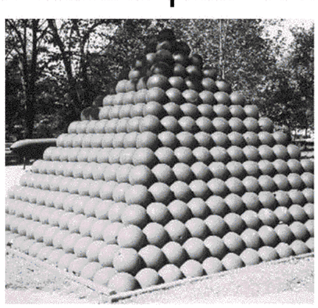
Maximally Dense Monodisperse Packings

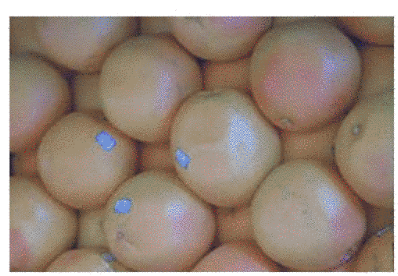
- In **2D**, solution is **triangular** lattice (Fejes Toth, 1940).



$\phi_{max} = \pi/12^{1/2} = 0.9058...$

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

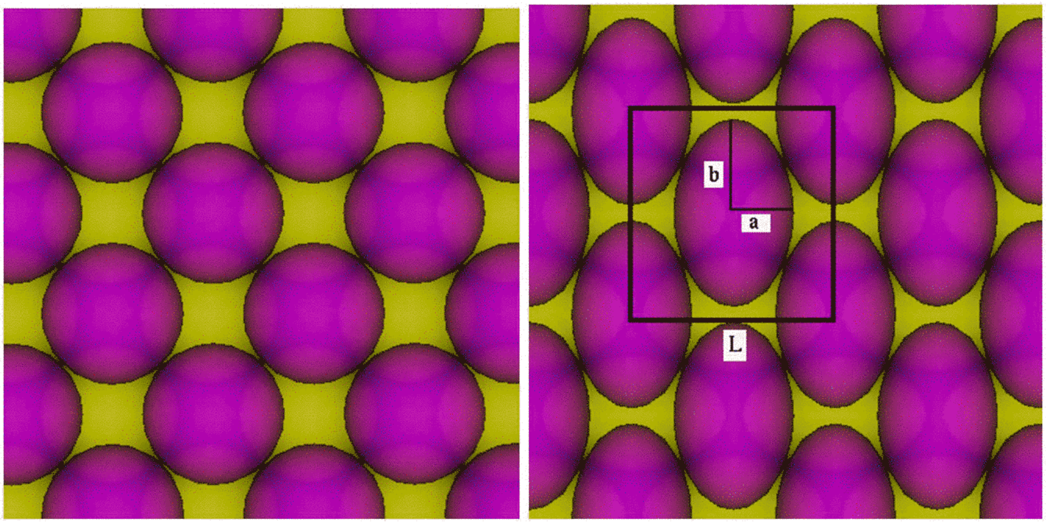




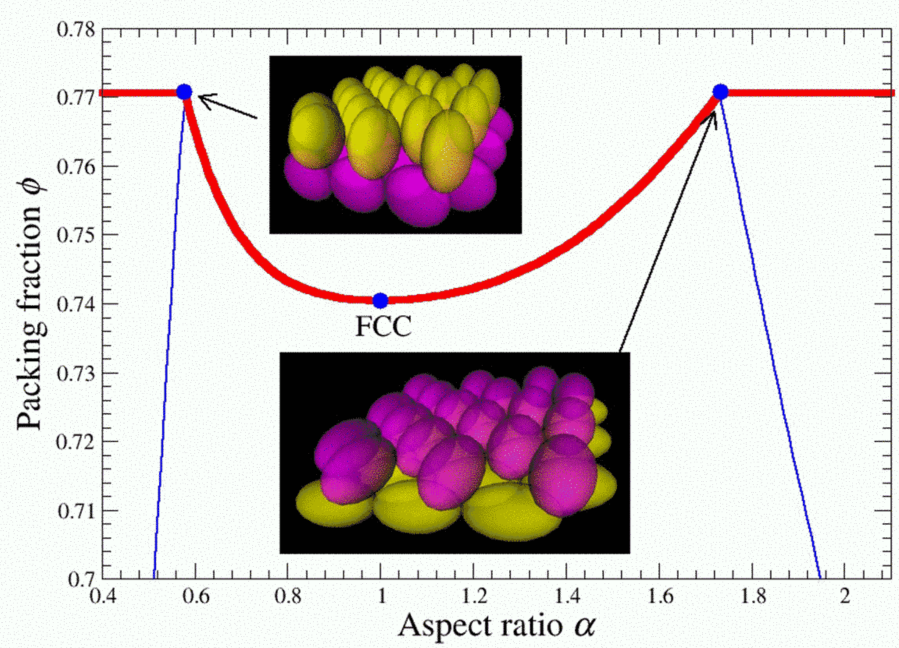
- **D>3**? **Polydispersity**? Other particle **shapes**?

- **Is the FCC ellipsoidal crystal the densest?**
  No!
- **We found densest known ellipsoidal crystals: $\phi \approx 0.77$**

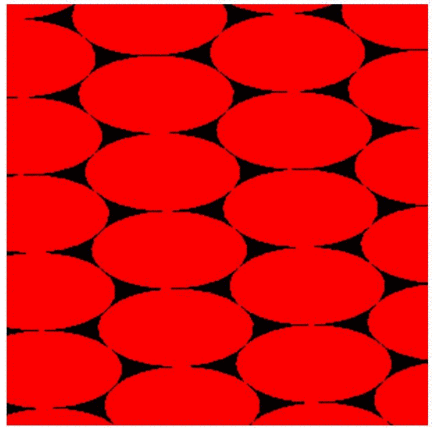
Donev, Stillinger, Chaikin, and Torquato, Phys. Rev. Lett. (2004)



Density of a Family of Crystal Packings

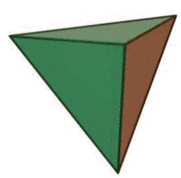


Densest Ellipse Packing Can Be Sheared!



Not True for the Densest Circle Packing!

- **Ulam's 1972 Conjecture: Among all densest packings of convex congruent objects in 3D, sphere gives the lowest packing density.**
- **Our ellipsoid results are consistent with this conjecture but what about the regular tetrahedron?**



- **Best packing is not a Bravais lattice packing: ϕ=0.367…**

Conway (1988): Non-Bravais lattice packing: ϕ=0.703…

Torquato (2005): Non-Bravais lattice packing: ϕ=0.717…

New (Provisional) Lower Bounds on the Optimal Density of Sphere Packings

Salvatore Torquato
Dept. of Chemistry, PRISM,
and Program in Applied & Computational Mathematics
Princeton University
http://cherrypit.princeton.edu

Joint work with Frank Stillinger, Princeton University

Sphere Packings in High Dimension d: Why?

- Understanding packings in **high d** has implications for **ground states** and can lead to insights in **low d**.
- **Communications**: Best way to send a digital code over a **noisy channel** corresponds to the **densest sphere packing** in a high-dimensional space.
- **Discrete Geometry**
- **String theory**: d=8

Optimal Packings for d>3 are not known!

Simple Cubic Lattice

Let $\mathbf{Z}$ denote the set of integers.

$$\mathbf{Z}^d = \{(x_1, \ldots, x_d) : x_i \in \mathbf{Z}\}$$

$$\phi = \frac{\pi^{d/2}}{\Gamma(1+d/2)2^d}$$

Checkerboard Lattice D_d

For $d \geq 3$

$$\mathbf{D}^d = \{(x_1, \ldots, x_d) \in \mathbf{Z}^d : x_1 + \cdots + x_d \text{ even}\}$$

$$\phi = \frac{\pi^{d/2}}{\Gamma(1+d/2)2^{(d+2)/2}}$$

D_3, D_4 and D_5 are the best known packings in $\Re^3$, $\Re^4$ and $\Re^5$, respectively.

Minkowski Lower Bound on ϕ_{max} for Lattice Sphere Packings

Minkowski (1905): The maximal packing density ϕ_{max} of a lattice packing of congruent spheres in $\Re^d$ for $d \geq 2$ satisfies

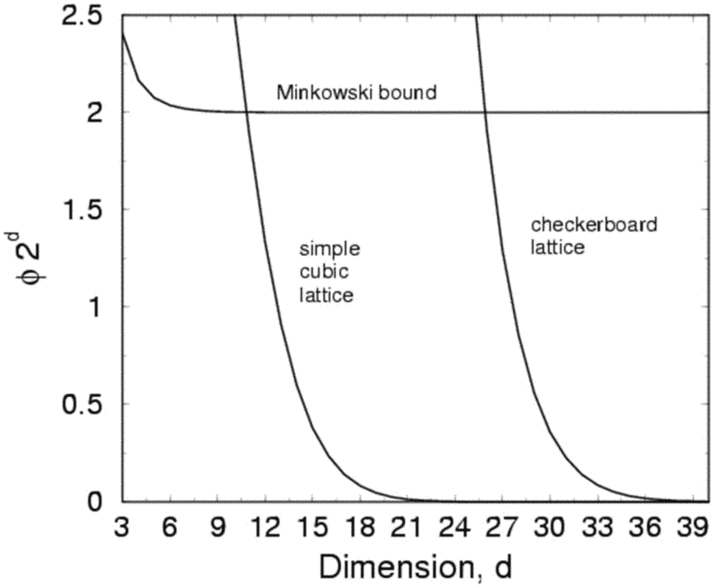
$$\phi_{\text{max}} \geq \frac{\zeta(d)}{2^{d-1}},$$

where $\zeta(d) = \sum_{k=1}^{\infty} k^{-d}$ is the Riemann zeta function.

Remarks:

1. There are no known constructions that realize the bound for any d.
2. No one has been able to provide any exponential improvement on the dominant asymptotic behavior 2^{-d}.
3. Note that any saturated sphere packing in $\Re^d$ has a density that satisfies the inequality

$$\phi \geq \frac{1}{2^d}.$$



New Approach To Obtain Lower Bounds on $\phi_{\max}$

- **Existence Theorem for Point Processes:** A necessary condition for the existence of a nonnegative pair correlation function $g_2(\boldsymbol{r})$ (nonnegative tempered distribution) of a translationally invariant point process at some number (center) density ρ is that $S(\boldsymbol{k}) \equiv 1 + \rho\tilde{h}(\boldsymbol{k}) \geq 0$, where $h(\boldsymbol{r}) \equiv g_2(\boldsymbol{r}) - 1$ and $h(\boldsymbol{k})$ is the Fourier transform of $h(\boldsymbol{r})$.
- **Torquato and Stillinger (2002):** Consider a family of test radial tempered distributions $g_2(r;\mathbf{a})$ at density ϕ, where a denotes a set of parameters. Now consider the optimization problem

$$\max_{\mathbf{a}} \phi$$

subject to the constraints

$$g_2(r;\mathbf{a}) \geq 0 \ \ \forall r,$$
$$\text{supp}(g_2) \subseteq \{r : r \geq 1\},$$
$$S(k;\mathbf{a}) \geq 0 \ \ \forall k.$$

We call $\phi_* \equiv \max_{\mathbf{a}} \phi$ the **terminal density** and noted that if such a g_2 at ϕ_* is realizable by a packing, then

$$\phi_{\max} \geq \phi_*.$$

- We recently discovered that our optimization problem is the dual of a primal linear program devised by Cohn and Elkies (2003) for an upper bound on $\phi_{\max}$ [see Cohn (2002)].

Existence of Disordered Packings in High Dimensions

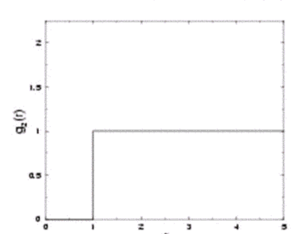
Definition: A disordered packing is one in which $g_2(\mathbf{r})$ decays to unity faster than $|\mathbf{r}|^{-d}$.

- **Conjecture:**

  A hard-core nonnegative tempered distribution $g_2(\boldsymbol{r})$ is a pair correlation function of a translationally invariant disordered sphere packing in $\Re^d$ at number density ρ for sufficiently large d if and only if $S(\boldsymbol{k}) \equiv 1 + \rho\tilde{h}(\boldsymbol{k}) \geq 0$. The maximum achievable density is the **terminal density** ϕ_*.

Can Disordered Packings Beat Best Lattice Packings?

- In sufficiently high dimensions, the answer is likely to be **yes**!
- For example, for the checkerboard lattice D_d, can show that $Z_n \sim d^{2n}$, where Z_n is # of centers in nth coordination shell at squared distance n.
- On the other hand, for sphere packings with $g_2(r) = \Theta(r-1)$ and $\phi = 1/2^d$



the cumulative coordination number is given by

$$Z(r) = \rho s_1(1)\int_1^r x^{d-1} g_2(x)dx = \frac{r^d}{d} - 1 \qquad \text{for} \quad r \geq 1.$$

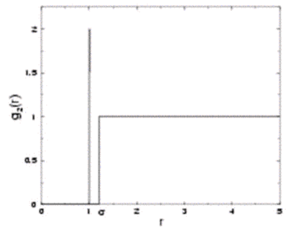
- At $r = \sqrt{3}$ and $d > 6$

$$\frac{Z(\sqrt{3})}{Z_3} \sim \frac{3^{d/2}}{4d^6/45} \qquad (Z(\sqrt{3})/Z_3 = 10^{13} \quad \text{for} \quad d = 100)$$

Step Plus Delta Function With a Gap

- Consider using the first and second attributes only, but allowing the location of the step discontinuity to vary:

$$g_2(r) = \Theta(r-\sigma) + \frac{Z}{s_1(1)\rho}\delta(r-1).$$



- We set $Z = (2\sigma)^d\phi - 1$ and optimize σ, which leads to a a terminal density ϕ_* that provides exponential improvement over previous result.
- This time the conjecture leads to a provisional lower bound that putatively gives the first exponential improvement over Minkowski's 100-year-old bound.

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Table 1: Optimized Parameters.

d	σ_*	Z_*	ϕ_*	$\frac{2^{d+1}\phi_*}{d+2}$
3	1.246997	7.932582	0.5758254	1.8426412
4	1.212589	9.544727	0.4252470	2.2679840
8	1.137981	70.89055	0.0998509	5.1123635
24	1.058992	5473.545632	0.00008245	106.40950
36	1.041618	76539.66718	2.566299 e-07	928.1827603
56	1.028041	4.249163900 e06	1.253255 e-11	31140.18622
80	1.020210	3.907730115 e08	6.521670 e-17	1.9229793 e06
100	1.016421	1.478803834 e10	2.288485 e-21	5.6882341 e08
125	1.013311	1.246171647 e12	5.610270 e-27	3.7580235 e09
150	1.011214	9.698080982 e13	1.275632 e-32	2.3192902 e11
175	1.009671	7.086019292 e15	2.745830 e-38	1.4858659 e13
200	1.008510	4.959085880 e17	5.667098 e-44	9.0165103 e14

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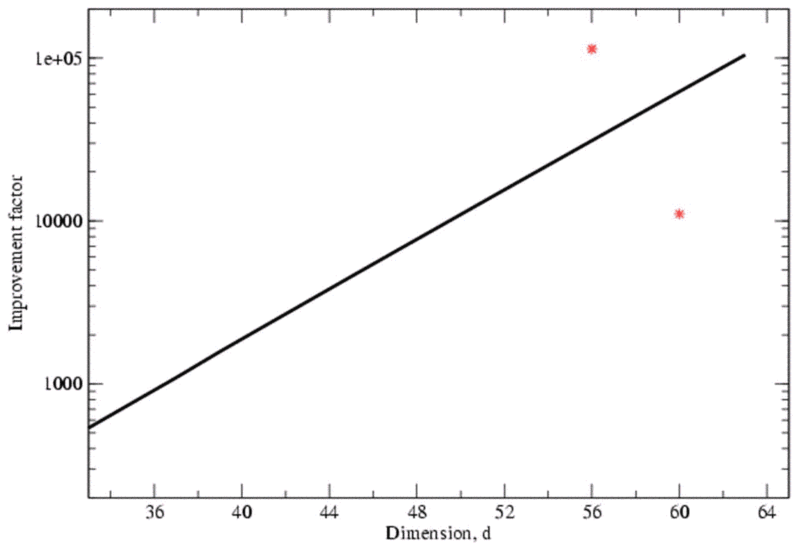
Bound Beats Best Known Packings for $d \geq 59$

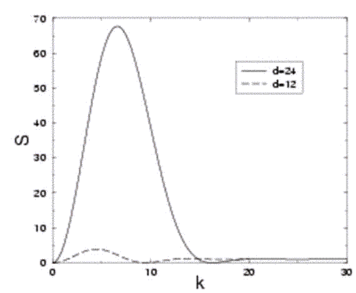
Improvement over density lower bound $(d+2)/2^{(d+1)}$



Asymptotic Analysis

- Structure factor is exactly

$$S(k) = 1 - c_1(d,\sigma)\frac{J_\nu(k\sigma)}{(k\sigma)^\nu} + c_2(d,\sigma)\frac{J_{\nu-1}(k)}{(k\sigma)^{\nu-1}},$$



- Let $\nu = d/2$. We obtain the asymptotic expression

$$\phi_* = \frac{1}{2^{1.557304959\nu - 2.677291565\nu^{1/3} + 0.5570132411}}\left[0.6089457595\nu^{1/6} - \frac{0.4194218702}{\nu^{1/6}} + \frac{0.06159502415}{\nu^{1/2}} + \mathcal{O}\left(\frac{1}{\nu^{5/6}}\right)\right],$$

For $d = 200$, formula predicts $\phi_* = 5.548526 \times 10^{-44}$, which is to be compared to the numerical solution of $\phi_* = 5.667098 \times 10^{-44}$.

Recent Collaborators

Bob Connelly (Cornell)
Paul Chaikin
Aleksandar Donev
Weining Man
Frank Stillinger

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