Structure and Novel Physical Properties of Disordered Hyperuniform Materials

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Qualitatively, What is Hyperuniformity?

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- Hyperuniformity provides a unified means of categorizing and characterizing crystals, quasicrystals and special disordered systems.
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Disordered hyperuniform many-particle systems can be regarded to be new ideal states of disordered matter in that they

1. behave more like crystals or quasicrystals in the manner in which they suppress large-scale density fluctuations, and yet are also like liquids and glasses since they are statistically isotropic structures with no Bragg peaks;

2. can exist as both as equilibrium and nonequilibrium phases;

3. come in quantum-mechanical and classical varieties;

4. and, appear to be endowed with unique bulk physical properties.

Understanding such disordered states of matter requires new theoretical tools and present experimental challenges.
Points can represent molecules of a material, stars in a galaxy, or trees in a forest. Let $\Omega \subset \mathbb{R}^d$ represent a \textbf{spherical} window of radius $R$.

Average number of points in window of volume $v_1(R)$: $\langle N(R) \rangle = \rho v_1(R) \sim R^d$

Local number variance: $\sigma^2(R) \equiv \langle N^2(R) \rangle - \langle N(R) \rangle^2$
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For Poisson point patterns and many disordered point patterns, $\sigma^2(R) \sim R^d$.

We call point patterns whose variance grows more slowly than $R^d$ (window volume) hyperuniform. Implies that scattering or structure factor vanishes in infinite-wavelength limit, i.e., $S(k) \rightarrow 0$ for $|k| \rightarrow 0$. 
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S(k) \to 0 \text{ for } |k| \to 0.
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All perfect crystals and many perfect quasicrystals are hyperuniform such that $\sigma^2(R) \sim R^{d-1}$: number variance grows like window surface area.

Hyperuniformity provides a unified means of categorizing and characterizing crystals, quasicrystals and special disordered systems.
SCATTERING AND DENSITY FLUCTUATIONS
Pair Statistics in Direct and Fourier Spaces

For particle systems in $\mathbb{R}^d$ at number density $\rho$, $g_2(r)$ is a nonnegative radial function that is proportional to the probability density of pair distances $r$.

The nonnegative structure factor $S(k) = 1 + \rho \tilde{h}(k)$ is obtained from the Fourier transform of $h(r) = g_2(r) - 1$, which we denote by $\tilde{h}(k)$.
Scaled Number Variance for 3D Systems at Unit Density

\[ \frac{\sigma^2(R)}{R^3} \]

- disordered non-hyperuniform
- ordered hyperuniform
- disordered hyperuniform

\[ R = 0, 1, 2, 3, 4 \]

\[ \sigma^2(R)/R^3 \]
Hidden Order on Large Length Scales

Which is the hyperuniform pattern?
Remarks About Equilibrium Systems

For single-component systems in equilibrium at average number density $\rho$,

$$\rho k_B T \kappa_T = \frac{\langle N^2 \rangle_* - \langle N \rangle_*^2}{\langle N \rangle_*} = S(k = 0) = 1 + \rho \int_{\mathbb{R}^d} h(r) dr$$

where $\langle \rangle_*$ denotes an average in the grand canonical ensemble.

Some observations:

- Any ground state ($T = 0$) in which the isothermal compressibility $\kappa_T$ is bounded and positive must be hyperuniform. This includes crystal ground states as well as exotic disordered ground states, described later.

- However, in order to have a hyperuniform system at positive $T$, the isothermal compressibility must be zero; i.e., the system must be incompressible.

- Note that a system at a thermal critical point is anti-hyperuniform in the sense that $\lim_{k \to 0} S(k) = +\infty$. 
ENSEMBLE-AVERAGE FORMULATION

For a translationally invariant point process at number density $\rho$ in $\mathbb{R}^d$:

$$\sigma^2(R) = \langle N(R) \rangle \left[ 1 + \rho \int_{\mathbb{R}^d} h(r) \alpha_2(r; R) dr \right]$$

$\alpha_2(r; R)$ - scaled intersection volume of 2 windows of radius $R$ separated by $r$
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For a certain class of systems and large $R$, we can show

$$\sigma^2(R) = 2^d \phi \left[ A \left( \frac{R}{D} \right)^d + B \left( \frac{R}{D} \right)^{d-1} + o \left( \frac{R}{D} \right)^{d-1} \right],$$

where $A$ and $B$ are the “volume” and “surface-area” coefficients:

$$A = S(k = 0) = 1 + \rho \int_{\mathbb{R}^d} h(r) dr, \quad B = -c(d) \int_{\mathbb{R}^d} h(r) r dr,$$

- **Hyperuniform**: $A = 0, B > 0 \implies$ Sum rule: $\rho \int_{\mathbb{R}^d} h(r) dr = -1$
- **Hyposurfical**: $A > 0, B = 0$
- **Degree of hyperuniformity for disordered systems**: Ratio $A/B$ or hyperuniformity index $H = S(k = 0)/S(k_{peak})$

We’ll see that you can have other variance scalings between $R^{d-1}$ and $R^d$. 
Hyperuniformity: Inverted Critical Phenomena

\( h(r) \) can be divided into direct correlations, via function \( c(r) \), and indirect correlations:

\[
\tilde{c}(k) = \frac{\tilde{h}(k)}{1 + \rho \tilde{h}(k)}
\]
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$$\tilde{c}(k) = \frac{\tilde{h}(k)}{1 + \rho \tilde{h}(k)}$$

- For any hyperuniform system, $\tilde{h}(k = 0) = -1/\rho$, and thus $\tilde{c}(k = 0) = -\infty$. Therefore, at the “critical” reduced density $\phi_c$, $h(r)$ is short-ranged and $c(r)$ is long-ranged.

- This is the inverse of the behavior at liquid-gas (or magnetic) critical points, where $h(r)$ is long-ranged (compressibility or susceptibility diverges) and $c(r)$ is short-ranged.
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- For sufficiently large $d$ at a disordered hyperuniform state, whether achieved via a nonequilibrium or an equilibrium route,

$$c(r) \sim -\frac{1}{r^{d-2+\eta}} \quad (r \to \infty), \quad \tilde{c}(k) \sim -\frac{1}{k^{2-\eta}} \quad (k \to 0),$$

$$h(r) \sim -\frac{1}{r^{d+2-\eta}} \quad (r \to \infty), \quad S(k) \sim k^{2-\eta} \quad (k \to 0),$$

where $(2 - d) < \eta < 2$ is a new critical exponent.

- One can think of a hyperuniform system as one resulting from an effective pair potential $v(r)$ at large $r$ that is a generalized Coulombic interaction between like charges. Why? Because

$$\frac{v(r)}{k_B T} \sim -c(r) \sim \frac{1}{r^{d-2+\eta}} \quad (r \to \infty)$$

- However, long-range interactions are not required to drive a nonequilibrium system to a disordered hyperuniform state.
We showed

\[ \sigma^2(R) = 2^d \phi \left( \frac{R}{D} \right)^d \left[ 1 - 2^d \phi \left( \frac{R}{D} \right)^d + \frac{1}{N} \sum_{i \neq j} \alpha_2(r_{ij}; R) \right] \]

where \( \alpha_2(r; R) \) can be viewed as a repulsive pair potential:
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Finding global minimum of \( \sigma^2(R) \) equivalent to finding ground state.

For large \( R \), in the special case of hyperuniform systems,

\[ \sigma^2(R) = \Lambda(R) \left( \frac{R}{D} \right)^{d-1} + \mathcal{O} \left( \frac{R}{D} \right)^{d-3} \]
The surface-area coefficient $\bar{\Lambda}$ for some crystal, quasicrystal and disordered one-dimensional hyperuniform point patterns.

<table>
<thead>
<tr>
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<td>Integer Lattice</td>
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<tr>
<td>Step+Delta-Function $g_2$</td>
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</tr>
<tr>
<td>Fibonacci Chain*</td>
<td>0.2011</td>
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*Zachary & Torquato (2009)*
Quantifying Suppression of Density Fluctuations at Large Scales: 1D

The surface-area coefficient $\overline{\Lambda}$ for some crystal, quasicrystal and disordered one-dimensional hyperuniform point patterns.

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More recent work on hyperuniformity of quasicrystals: Ogüz, Socolar, Steinhardt and Torquato (2016).
The surface-area coefficient $\bar{\Lambda}$ for some crystal, quasicrystal and disordered two-dimensional hyperuniform point patterns.

<table>
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<tr>
<th>2D Pattern</th>
<th>$\bar{\Lambda}/\phi^{1/2}$</th>
</tr>
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<tbody>
<tr>
<td>Triangular Lattice</td>
<td>0.508347</td>
</tr>
<tr>
<td>Square Lattice</td>
<td>0.516401</td>
</tr>
<tr>
<td>Honeycomb Lattice</td>
<td>0.567026</td>
</tr>
<tr>
<td>Kagomé Lattice</td>
<td>0.586990</td>
</tr>
<tr>
<td>Penrose Tiling*</td>
<td>0.597798</td>
</tr>
<tr>
<td>Step+Delta-Function $g_2$</td>
<td>0.600211</td>
</tr>
<tr>
<td>Step-Function $g_2$</td>
<td>0.848826</td>
</tr>
<tr>
<td>One-Component Plasma</td>
<td>1.12838</td>
</tr>
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Quantifying Suppression of Density Fluctuations at Large Scales: 3D

Contrary to conjecture that lattices associated with the densest sphere packings have smallest variance regardless of $d$, we have shown that for $d = 3$, BCC has a smaller variance than FCC.

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<tr>
<td>BCC Lattice</td>
<td>1.24476</td>
</tr>
<tr>
<td>FCC Lattice</td>
<td>1.24552</td>
</tr>
<tr>
<td>HCP Lattice</td>
<td>1.24569</td>
</tr>
<tr>
<td>SC Lattice</td>
<td>1.28920</td>
</tr>
<tr>
<td>Diamond Lattice</td>
<td>1.41892</td>
</tr>
<tr>
<td>Wurtzite Lattice</td>
<td>1.42184</td>
</tr>
<tr>
<td>Damped-Oscillating $g_2$</td>
<td>1.44837</td>
</tr>
<tr>
<td>Step+Delta-Function $g_2$</td>
<td>1.52686</td>
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Carried out analogous calculations in high $d$ (Zachary & Torquato, 2009) - of importance in communications. Disordered point patterns may win in high $d$ (Torquato & Stillinger, 2006).
General Hyperuniform Scaling Behaviors

Consider hyperuniform systems characterized by a power-law structure factor

\[ S(k) \sim |k|^\alpha, \quad (|k| \to 0) \]

Limits \( \alpha \to 0 \) and \( \alpha \to \infty \) correspond to Poisson and crystal (or stealthy) systems.

Can prove that the number variance \( \sigma^2(R) \) increases for large \( R \) asymptotically as (Zachary and Torquato, 2011)

\[ \sigma^2(R) \sim \begin{cases} 
R^{d-1}, & \alpha > 1 \quad \text{(CLASS I)} \\
R^{d-1} \ln R, & \alpha = 1 \quad \text{(CLASS II)} \\
R^{d-\alpha}, & 0 < \alpha < 1 \quad \text{(CLASS III)}
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\]

- **Class I:** \( \sigma^2(R) \sim R^{d-1} \): Crystals, quasicrystals, stealthy disordered ground states, charged systems, \( g_2 \)-invariant disordered point processes.

- **Class II:** \( \sigma^2(R) \sim R^{d-1} \ln(R) \): Quasicrystals, classical disordered ground states, zeros of the Riemann zeta function, eigenvalues of random matrices, fermionic point processes, superfluid helium, maximally random jammed packings, density fluctuations in early Universe, prime numbers.

- **Class III:** \( \sigma^2(R) \sim R^{d-\alpha} \ (0 < \alpha < 1) \): Classical disordered ground states, nonequilibrium phase transitions/random organization models.
There are a variety of different systems in $\mathbb{R}$ that are disordered and hyperuniform with the pair correlation function $g_2(r) = 1 - \sin^2(\pi r)/(\pi r)^2$:

1D point pattern is always negatively correlated, i.e., $g_2(r) \leq 1$ and pairs of points tend to repel one another, i.e., $g_2(r) \to 0$ as $r$ tends to zero.

- Eigenvalues of random Hermitian matrices: Dyson 1962, 1970;
- Nontrivial zeros of the Riemann zeta function granting the Riemann hypothesis: Montgomery 1973;

Dyson mapped the GUE solution to a 1D log Coulomb gas at positive temperature: $k_B T = 1/2$. The total potential energy of the system is given by

$$\Phi_N(r^N) = \frac{N}{2} \sum_{i=1}^{N} |r_i|^2 - \sum_{i \leq j}^{N} \ln(|r_i - r_j|).$$
1D Disordered Hyperuniform Systems

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![Graphs showing $g_2(r)$ and $S(k)$](image)

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Recently showed that prime numbers in a distinguished limit are hyperuniform in a similar to but different from quasicrystals (Torquato et al. 2019).
Recent 2D and 3D Examples of Disordered Hyperuniform Systems

Physical Examples

- **Disordered classical ground states**: Uche et al. PRE (2004)
- **Maximally random jammed (MRJ) particle packings**: \( S(k) \sim k \) as \( k \to 0 \) (nonequilibrium states): Donev et al. PRL (2005); Zachary et al. PRL (2011); Dreyfus et al., PRE (2015)
- **Fermionic point processes**: \( S(k) \sim k \) as \( k \to 0 \) (ground states and/or positive temperature equilibrium states): Torquato et al. J. Stat. Mech. (2008); Scardicchio et al., PRE, 2009
- **Charged Hard-Sphere Systems**: Lomba et al. PRE (2017,2018); Chen et al. PCCP (2018)
- **Vortex pinning and states in superconductors**: Reichhardt et al. PRB (2017)
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- **Charged Hard-Sphere Systems**: Lomba et al. PRE (2017, 2018); Chen et al. PCCP (2018)
- **Random organization** (nonequilibrium states):
- **Vortex pinning and states in superconductors**: Reichhardt et al. PRB (2017)

Natural Disordered Hyperuniform Systems

- **Immune-system receptors** (nonequilibrium states): Balasubramanian et al. PNAS (2015)
Recent 2D and 3D Examples of Disordered Hyperuniform Systems

Physical Examples

- Maximally random jammed (MRJ) particle packings: \( S(k) \sim k \) as \( k \to 0 \) (nonequilibrium states): Donev et al. PRL (2005); Zachary et al. PRL (2011); Dreyfus et al., PRE (2015)
- Fermionic point processes: \( S(k) \sim k \) as \( k \to 0 \) (ground states and/or positive temperature equilibrium states): Torquato et al. J. Stat. Mech. (2008); Scardicchio et al., PRE, 2009
- Vortex pinning and states in superconductors: Reichhardt et al. PRB (2017)

Natural Disordered Hyperuniform Systems


Nearly Hyperuniform Disordered Systems

- Amorphous Silicon (nonequilibrium states): Henja et al. PRB (2013)
- Structural Glasses (nonequilibrium states): Marcotte et al. (2013)
Hyperuniformity and Jammed Packings

Conjecture: All strictly jammed saturated sphere packings are hyperuniform (Torquato & Stillinger, 2003).
**Hyperuniformity and Jammed Packings**

- **Conjecture**: All strictly jammed saturated sphere packings are hyperuniform (Torquato & Stillinger, 2003).

- A 3D maximally random jammed (MRJ) packing is a prototypical glass in that it is maximally disordered but perfectly rigid (infinite elastic moduli).

- Such packings of identical spheres have been shown to be hyperuniform with quasi-long-range (QLR) pair correlations in which $h(r)$ decays as $-1/r^4$ (Donev, Stillinger & Torquato, PRL, 2005).

This is to be contrasted with the hard-sphere fluid with correlations that decay exponentially fast. Contradicts frozen-liquid picture of a glass.
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Apparently, hyperuniform QLR correlations with decay $-1/r^{d+1}$ are a universal feature of general MRJ packings in $\mathbb{R}^d$.

Zachary, Jiao and Torquato, PRL (2011): ellipsoids, superballs, sphere mixtures
Berthier et al., PRL (2011); Kurita and Weeks, PRE (2011): sphere mixtures
Jiao and Torquato, PRE (2011): polyhedra
Hyperuniformity and Spin-Polarized Free Fermions

One can map random Hermitian matrices (GUE), fermionic gases, and zeros of the Riemann zeta function to a unique hyperuniform point process on \( \mathbb{R} \).
Hyperuniformity and Spin-Polarized Free Fermions

One can map random Hermitian matrices (GUE), fermionic gases, and zeros of the Riemann zeta function to a unique hyperuniform point process on $\mathbb{R}$.

We provide exact generalizations of such a point process in $d$-dimensional Euclidean space $\mathbb{R}^d$ and the corresponding $n$-particle correlation functions, which correspond to those of spin-polarized free fermionic systems in $\mathbb{R}^d$.

\[ g_2(r) = 1 - \frac{2\Gamma(1 + d/2) \cos^2 (rK - \pi(d+1)/4)}{K \pi^{d/2+1} r^{d+1}} \quad (r \to \infty) \]

\[ S(k) = \frac{c(d)}{2K} k + \mathcal{O}(k^3) \quad (k \to 0) \quad (K : \text{Fermi sphere radius}) \]

Scardicchio, Zachary & Torquato, PRE, 2009
In the Eye of a Chicken: Photoreceptors

- **Optimal** spatial sampling of light requires that **photoreceptors** be arranged in the **triangular lattice** (e.g., insects and some fish).

- **Birds** are highly **visual** animals, yet their cone photoreceptor patterns are **irregular**.
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![5 Cone Types](image_url)

*Jiao, Corbo & Torquato, PRE (2014).*
Avian Cone Photoreceptors

Disordered mosaics of both total population and individual cone types are effectively hyperuniform, which had been never observed in any system before. We call this multi-hyperuniformity (Jiao, Corbo & Torquato, PRE 2014).
Avian Cone Photoreceptors

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Recently showed that multihyperuniformity can be rigorously achieved via hard-disk plasmas (Lomba, Weis and Torquato, PRE 2018).
Slow and Rapid Cooling of a Liquid

Classical ground states are those classical particle configurations with minimal potential energy per particle.
Slow and Rapid Cooling of a Liquid

Classical *ground states* are those classical particle configurations with *minimal* potential energy per particle.

![Diagram showing the relationship between temperature and volume for different states of matter including glass, rapid quench, liquid, and very slow cooling.](image)

- Typically, ground states are *periodic with high crystallographic symmetries*. 
Slow and Rapid Cooling of a Liquid

Classical ground states are those classical particle configurations with minimal potential energy per particle.

Typically, ground states are periodic with high crystallographic symmetries.

Can classical ground states derived from nontrivial interactions ever be disordered?
Consider $N$ particles with configuration $r^{N}$ in a fundamental region $\Omega$ under periodic boundary conditions) with a pair potential $v(\mathbf{r})$ that is bounded with Fourier transform $\tilde{v}(k)$. 
Fourier-Based Design of Hyperuniform Particle Configurations

Uche, Stillinger & Torquato, Phys. Rev. E 2004
Batten, Stillinger & Torquato, Phys. Rev. E 2008

Collective-Coordinate Optimization Procedure

Consider \( N \) particles with configuration \( \mathbf{r}^N \) in a fundamental region \( \Omega \) under periodic boundary conditions) with a pair potential \( v(\mathbf{r}) \) that is bounded with Fourier transform \( \tilde{v}(\mathbf{k}) \).

The total energy is

\[
\Phi_N(\mathbf{r}^N) = \sum_{i<j} v(\mathbf{r}_{ij}) = \frac{N}{2|\Omega|} \sum_{\mathbf{k}} \tilde{v}(\mathbf{k}) S(\mathbf{k}) + \text{constant}
\]

For \( \tilde{v}(\mathbf{k}) \) positive \( \forall \ 0 \leq |\mathbf{k}| \leq K \) and zero otherwise, finding configurations in which \( S(\mathbf{k}) \) is constrained to be zero where \( \tilde{v}(\mathbf{k}) \) has support is equivalent to globally minimizing \( \Phi(\mathbf{r}^N) \).

These hyperuniform ground states are called “stealthy” and when disordered are highly degenerate.
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These hyperuniform ground states are called “stealthy” and when disordered are highly degenerate.

Stealthy patterns can be tuned by varying the parameter $\chi$: ratio of number of constrained degrees of freedom to the total number of degrees of freedom, $d(N - 1)$.
From various initial distributions of $N$ points, found the energy minimizing configurations (with extremely high precision) using optimization techniques.
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For $0 \leq \chi < 0.5$, stealthy ground states are highly degenerate, disordered and isotropic. Success rate to achieve disordered ground states is 100%.

As \( \chi \) increases, short-range order increases.
Creation of Disordered Stealthy Ground States via Collective Coordinates

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Animations
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For $\chi > 1/2$ (no degrees of freedom), the system undergoes a transition to a crystal phase and the energy landscape becomes considerably more complex.

The dimensionality of the configuration space depends on $\chi$. A statistical-mechanical theory for stealthy ground-state thermodynamics and structure has been formulated [Torquato, Zhang and Stillinger, PRX (2015)].
Stealthy Disordered Ground States and Novel Materials

Until recently, it was believed that Bragg scattering was required to achieve cellular solids with complete photonic band gaps.
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These network material designs have been fabricated for microwave regime: Man, Florescu et. al., PNAS (2013).

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High-density transparent stealthy disordered materials: Leseur et al. (2016).

Stealthy materials are nearly optimal wave absorbers: Bigourdan et al. (2018).
Disordered Stealthy Materials with Optimal Transport/Elastic Properties

Optimal Effective Diffusivity in Decorated Stealthy Patterns


Optimal Effective Conductivity and Elastic Moduli in Stealthy Networks

Chen & Torquato, Multifunctional Materials (2018)
WHY DO VERY LARGE DISORDERED STEALTHY HYPERUNIFORM MATERIALS YIELD DESIRABLE PHYSICAL PROPERTIES?
WHY DO VERY LARGE DISORDERED STEALTHY HYPERUNIFORM MATERIALS YIELD DESIRABLE PHYSICAL PROPERTIES?

**ANSWER:** Partly because they are disordered materials with some characteristics of crystals, including unusual “hole” statistics, i.e., holes of arbitrarily large size are prohibited in thermodynamic limit.

**Targeted Spectra** $S(k)$

Configurations are ground states of many-particle systems with up to two-, three- and four-body interactions ([Uche, Stillinger & Torquato, Phys. Rev. E 2006](#)).

![Graph showing various curves representing $S(k)$ vs. $k$]

**Figure 1:** One of them is for $S(k) \sim k^6$ and other for $S(k) \sim k$. 
Can many-body interactions be devised that eliminate the possibilities of crystalline and quasicrystalline phases, while creating mechanically stable, hyperuniform amorphous glasses down to absolute zero.
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This has been accomplished via the perfect-glass paradigm, which represents a soft-interaction analog of the MRJ packing, which is the epitome of a glass.
Can many-body interactions be devised that eliminate the possibilities of crystalline and quasicrystalline phases, while creating mechanically stable, hyperuniform amorphous glasses down to absolute zero.

This has been accomplished via the perfect-glass paradigm, which represents a soft-interaction analog of the MRJ packing, which is the epitome of a glass.

The perfect-glass model corresponds to the ground state of a potential involving two-, three-, and four-body soft interactions corresponding to an amorphous structure factor.

The disordered ground states are unique such that they can always be superposed onto each other or their mirror image.
Hyperuniformity of Disordered Two-Phase Materials


For typical disordered media, volume-fraction variance $\sigma^2_V(R)$ for large $R$ goes to zero like $R^{-d}$.

For hyperuniform disordered two-phase media, $\sigma^2_V(R)$ goes to zero faster than $R^{-d}$, equivalent to following condition on spectral density $\tilde{\chi}_V(k)$:

$$\lim_{|k| \to 0} \tilde{\chi}_V(k) = 0.$$
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Interfacial-area fluctuations play an important role in static and surface-area evolving structures. Here we define $\sigma^2_S(R)$ and hyperuniformity condition is (Torquato, PRE, 2016)

$$\lim_{|k| \to 0} \tilde{\chi}_S(k) = 0.$$
Disordered hyperuniform two-phase systems can be designed with targeted spectral functions (Chen and Torquato, Acta Materialia, 2018). This is the digitized analog of the Fourier-based collective-coordinate procedure.

For example, consider following hyperuniform functional forms in 2D and 3D:
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For example, consider following hyperuniform functional forms in 2D and 3D:

The following is a 2D realization:
Multifunctional Composites for Elastic and Electromagnetic Wave Propagation

Kim and Torquato, arXiv:1908.06662, 2019

- Derived accurate formulas for effective elastic and electromagnetic wave characteristics of composites, such as effective moduli, wave speeds and attenuation coefficients, that depend explicitly on the spectral density $\tilde{\chi}_V(k)$.

- By eliminating common microstructural quantity involving spectral density, found “cross-property relations” that link effective elastic and electromagnetic wave characteristics to one another, facilitating multifunctional design.
Derived accurate formulas for effective elastic and electromagnetic wave characteristics of composites, such as effective moduli, wave speeds and attenuation coefficients, that depend explicitly on the spectral density $\tilde{\chi}_\nu(k)$.

By eliminating common microstructural quantity involving spectral density, found “cross-property relations” that link effective elastic and electromagnetic wave characteristics to one another, facilitating multifunctional design.

Left panel: Both elastic and electromagnetic waves can be attenuated due to scattering, if this composite has a non-zero scattering intensity at $k_I$.

Right panel: Composite attenuates elastic waves but is transparent to electromagnetic waves.
Multifunctional Composites for Elastic and Electromagnetic Wave Propagation

Kim and Torquato, arXiv:1908.06662, 2019

- Explored the wave characteristics of a disordered composites, including exotic disordered “hyperuniform” varieties.

- Showed that composites with disordered hyperuniform microstructures exhibit novel elastic wave characteristics, e.g., low-pass or narrow-band-pass filters that transmit or absorb elastic waves “isotropically” for a range of wavenumbers.

- They have advantages over ordered ones, such as optimal, direction-independent properties and robustness against defects.

- By engineering disordered hyperuniform microstructures and phase properties, composites can exhibit “anomalous dispersion” with resonance-like attenuation.

- Our cross-property relations for effective wave characteristics can be used in design multifunctional composites via inverse techniques, e.g., components of aircraft/spacecraft or buildings & nondestructive evaluation of elastic moduli from dielectric response.
Other Generalizations of Hyperuniformity

Recently considered (Torquato, PRE 2016)

Random scalar fields: Concentration and temperature fields in random media and turbulent flows, laser speckle patterns, and temperature fluctuations associated with CMB. Spinodal decomposition patterns are hyperuniform: Ma & Torquato, PRE (2017)

Random vector fields: Random media (e.g., heat, current, electric, magnetic and velocity vector fields) and turbulence.

Structurally anisotropic materials: Many-particle systems and random media that are statistically anisotropic, requiring generalization to directional hyperuniformity.
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    ![Spinodal decomposition patterns](image)
    
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![S(k) diagram](image)
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  - Random scalar fields: Concentration and temperature fields in random media and turbulent flows, laser speckle patterns, and temperature fluctuations associated with CMB.
    
    ![Image of spinodal decomposition patterns](spinodal.png)
    
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- Is there a many-particle system with following anisotropic scattering pattern?

![Image of scattering pattern](scattering.png)

- Treatment of spin systems, both classical [Chertkov et al., PRB (2016)] and quantum-mechanical [Crowley, Laumann & Gopalakrishnan, arXiv:1809.04595]
Challenge: Creation of Very Large Hyperuniform Samples Across Length Scales

- It is a numerical/experimental challenge to generate very large samples that are hyperuniform with high fidelity across length scales, e.g. nanometers to millimeters.

- Collective coordinates enable essentially perfectly hyperuniform systems, but current methods are sample-size limited.

- Particle or colloidal systems in equilibrium require long-range interactions.
Challenge: Creation of Very Large Hyperuniform Samples Across Length Scales

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- Collective coordinates enable essentially perfectly hyperuniform systems, but current methods are sample-size limited.

- Particle or colloidal systems in equilibrium require long-range interactions.

- It is reasonable to try charged colloids and tune temperature and screening length to manipulate the spectral density at small $k$. We recently carried out such a computational study using a mixture of binary charged colloids that interact via a repulsive hard-core Yukawa potential: Chen, Lomba & Torquato, PCCP (2018).

- We found that at dimensionless temperatures below 0.05 and dimensionless inverse screening lengths below 1.0, which are experimentally accessible, the disordered systems become effectively hyperuniform.

- As expected, as the temperature and inverse screening length decrease, the level of hyperuniformity was shown to increase.
Tessellation-Based Procedure to Create Very Large Hyperuniform Packings

Kim and Torquato, Acta Mater., 2019
Kim and Torquato, arXiv:1901.10006, 2019

- Introduced a new and simple construction procedure that ensure perfect hyperuniformity for very large sample sizes.

- Beginning with a tessellation of space (e.g., Voronoi, Delaunay, Laguerre, sphere, ...), insert a particle into each cell such that local-cell packing fractions are identical to global packing fraction.

- We prove that this results in packings of particles with a size distribution that are guaranteed to be perfectly hyperuniform in the infinite-sample-size limit.

- This enables an algorithm that converts a very large nonhyperuniform disordered packing into a hyperuniform one.

- Also establish hyperuniformity of the famous Hashin-Shtrikman multiscale packings, which possess optimal transport and elastic properties.

- Our hyperuniform designs can be readily fabricated using modern photolithographic and 3D printing technologies.
We proved that for initial tessellations satisfying the **bounded-cell condition**, the resulting polydisperse packings with packing fraction $\phi$ are hyperuniform of class I:

$$\tilde{\chi}_V(k) \sim \phi^2 |k|^4$$

Since fluctuations only arise **near the window boundary**, we can prove

$$\sigma^2_V(R) \sim R^{-(d+1)}$$
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Since fluctuations only arise near the window boundary, we can prove

$$\sigma_V^2(R) \sim R^{-(d+1)}$$

When sphere tessellations are employed, the resulting hyperuniform structures are the optimal Hashin-Shtrikman coated-spheres model.

We proved that for disordered coated-spheres model, $\tilde{\chi}_V(k) \sim |k|^4$. 

Algorithmic Implementation: Voronoi and Sphere Tessellations

Our algorithm can convert extremely large nonhyperuniform packings (up to $N = 10^8$), such as equilibrium or RSA packings, into hyperuniform ones via Voronoi tessellations in 2D or 3D. For example, for 2D equilibrium hard-disk packings at $\phi_{\text{init}} = 0.65$, the final packing fraction can be adjusted up to $\phi = 0.437$. 

![Graph showing the packing fraction distribution](image)

- 2D equilibrium progenitor packings
- Constructed packings

$\tilde{\chi}_V(k) \times 10^4$ vs. $k \rho^{-1/2}$
Algorithmic Implementation: Voronoi and Sphere Tessellations

- Our algorithm can convert extremely large nonhyperuniform packings \((\text{up to } N = 10^8)\), such as equilibrium or RSA packings, into hyperuniform ones via Voronoi tessellations in 2D or 3D.
- For example, for 2D equilibrium hard-disk packings at \(\phi_{\text{init}} = 0.65\), the final packing fraction can be adjusted up to \(\phi = 0.437\).

![Graph showing packing fraction vs. \(k\rho^{-1/2}\)]

- We numerically verified our results for the coated-spheres model via a multi-stage version of RSA with decreasing particle diameter \(D_m = D_1 m^{-p/d}\) at stage \(m\). With \(p = 1.8\) and \(\phi = 0.5\):

![Graph showing packing fraction vs. \(kD_1\)]
Universal hidden order in amorphous cellular geometries


Inherent structures of the Quantizer problem:

In each Voronoi cell, iteratively replace the generator by the center of mass (Lloyd 1957).

Quick convergence even for large system sizes
Universal hidden order in amorphous cellular geometries

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- Quick convergence even for large system sizes

![Graph showing structure factor](image)

Even though the initial configurations differ vastly from stealthy to anti-hyperuniform, …
Universal hidden order in amorphous cellular geometries

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Quantizer problem:

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iteratively replace the generator
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Quick convergence
even for large system sizes

Even though the initial configurations
differ vastly from stealthy to
anti-hyperuniform, …

their final configurations are universal
with the same
– two-point statistics,
– energy distributions,
– Minkowski tensors, etc.
within error bars.

They are fully amorphous and
effectively hyperuniform.
Hyperuniformity provides a unified means of categorizing and characterizing crystals, quasicrystals and special correlated disordered systems.

Hyperuniformity concept brings to the fore the importance of long-wavelength correlations in non-hyperuniform systems (liquids and glasses). The degree of hyperuniformity provides an order metric for the extent to which large-scale density fluctuations are suppressed in such systems.

Disordered hyperuniform materials are ideal states of amorphous matter that often are endowed with novel bulk properties that we are only beginning to discover.

We can now produce disordered hyperuniform materials with designed spectra.

Hyperuniform scalar and vector fields as well as directional hyperuniform materials represent exciting new extensions.

Hyperuniformity has become a powerful concept that connects a variety of seemingly unrelated systems that arise in physics, chemistry, materials science, mathematics, and biology.
CONCLUSIONS

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Collaborators

Roberto Car (Princeton)        Enrique Lomba (Madrid)
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Amorphous Silicon is Nearly Hyperuniform

Highly sensitive transmission X-ray scattering measurements performed at Argonne on amorphous-silicon (a-Si) samples reveals that they are nearly hyperuniform with $S(0) = 0.0075$.

Long, Roorda, Hejna, Torquato, and Steinhardt (2013)

This is significantly below the putative lower bound recently suggested by de Graff and Thorpe (2009) but consistent with the recently proposed nearly hyperuniform network picture of a-Si (Hejna, Steinhardt and Torquato, 2013).
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Increasing the degree of hyperuniformity of a-Si appears to be correlated with a larger electronic band gap (Hejna, Steinhardt and Torquato, 2013).
Structural Glasses and Growing Length Scales

Important question in glass physics: Do growing relaxation times under supercooling have accompanying growing structural length scales? Lubchenko & Wolynes (2006); Berthier et al. (2007); Karmakar, Dasgupta & Sastry (2009); Chandler & Garrahan (2010); Hocky, Markland & Reichman (2012)
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We studied glass-forming liquid models that support an alternative view: existence of growing static length scales (due to increase of the degree of hyperuniformity) as the temperature $T$ of the supercooled liquid is decreased to and below $T_g$ that is intrinsically nonequilibrium in nature.
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We studied glass-forming liquid models that support an alternative view: existence of growing static length scales (due to increase of the degree of hyperuniformity) as the temperature $T$ of the supercooled liquid is decreased to and below $T_g$ that is intrinsically nonequilibrium in nature.

The degree of deviation from thermal equilibrium is determined from a nonequilibrium index

$$X = \frac{S(k = 0)}{\rho k_B T \kappa_T} - 1,$$

which increases upon supercooling. Marcotte, Stillinger & Torquato (2013)
By many measures, the prime numbers can be regarded to be pseudo-random numbers.

We treated the primes in some interval \([M, M + L]\) to be a special lattice-gas model: primes are “occupied” sites on a integer lattice of spacing 2 that contains all of the positive odd integers and the unoccupied sites are the odd composite integers.

We numerically studied intervals with \(M\) large, and \(L/M\) smaller than unity and found unexpected structure on all length scales!

Figure 2: \(S(k)\) for prime numbers for \(M = 10^{10} + 1\) and \(L = 10^5\) contains many peaks of various heights, creating a type of self-similarity.
Our main results are obtained for the interval $M \leq p \leq M + L$ with $M$ very large and the ratio $L/M$ held constant. This enables us to treat the primes as a homogeneous point pattern.

In the infinite-size limit, we showed that the primes are hyperuniform and that $S(k)$ is determined entirely by a set of dense Bragg peaks, i.e.,

$$\lim_{M \to \infty} \frac{S(k)}{2\pi \rho} = \sum_{n} \sum_{m} \frac{1}{\phi(n)^2} \delta \left( k - \frac{m\pi}{n} \right),$$

where the symbol $\flat$ is meant to indicate that the sum over $n$ only involves odd, square-free values of $n$ and the symbol $\times$ indicates that $m$ and $n$ have no common factor.

Unlike quasicrystals, the prime peaks occur at certain rational multiples of $\pi$, which is similar to limit-periodic systems.

Limit-periodic points sets $\equiv$ Aperiodic structures with dense set of Bragg peaks generated from a union of periodic structures with ever increasing periodicities.

But the primes show an erratic pattern of occupied and unoccupied sites, very different from the predictable and orderly patterns of standard limit–periodic systems. Hence, the primes are the first example of a point pattern that is effectively limit-periodic.

We identified a transition between ordered and disordered prime regimes that depends on the intervals studied.
Creation of Disordered Stealthy Ground States
Creation of Disordered Stealthy Ground States

One class of stealthy potentials involves the following power-law form:

\[ \tilde{v}(k) = v_0 \left(1 - \frac{k}{K}\right)^m \Theta(K - k), \]

where \( n \) is any whole number. The special case \( n = 0 \) is just the simple step function.

In the large-system (thermodynamic) limit with \( m = 0 \) and \( m = 4 \), we have the following large-\( r \) asymptotic behavior, respectively:

\[ v(r) \sim \frac{\cos(r)}{r^2} \quad (m = 0) \]
\[ v(r) \sim \frac{1}{r^4} \quad (m = 4) \]

While the specific forms of these stealthy potentials lead to the same convergent ground-state energies, this will not be the case for the pressure and other thermodynamic quantities.
Nontrivial: Dimensionality of the configuration space depends on the number density $\rho$ (or $\chi$) and there is a multitude of ways of sampling the ground-state manifold, each with its own probability measure. *Which ensemble? How are entropically favored states determined?*

Derived general exact relations for thermodynamic properties that apply to any ground-state ensemble as a function of $\rho$ in any $d$ and showed how disordered degenerate ground states arise.
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Derived general exact relations for thermodynamic properties that apply to any ground-state ensemble as a function of $\rho$ in any $d$ and showed how disordered degenerate ground states arise.

From previous considerations, we that an important contribution to $S(k)$ is a simple hard-core step function $\Theta(k - K)$, which can be viewed as a disordered hard-sphere system in Fourier space in the limit that $\chi \sim 1/\rho$ tends to zero, i.e., as the number density $\rho$ tends to infinity.

That the structure factor must have the behavior

$$S(k) \rightarrow \Theta(k - K), \quad \chi \rightarrow 0$$

is perfectly reasonable; it is a perturbation about the ideal-gas limit in which $S(k) = 1$ for all $k$. 
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We make the ansatz that for sufficiently small $\chi$, $S(k)$ in the canonical ensemble for a stealthy potential can be mapped to $g_2(r)$ for an effective disordered hard-sphere system for sufficiently small density.
Pseudo-Hard Spheres in Fourier Space

Let us define

\[ \tilde{H}(k) \equiv \rho \tilde{h}(k) = h_{HS}(r = k) \]

There is an Ornstein-Zernike integral equation that defines FT of appropriate direct correlation function, \( \tilde{C}(k) \):

\[ \tilde{H}(k) = \tilde{C}(k) + \eta \tilde{H}(k) \otimes \tilde{C}(k), \]

where \( \eta \) is an effective packing fraction. Therefore,

\[ H(r) = \frac{C(r)}{1 - (2\pi)^d \eta C(r)}. \]

This mapping enables us to exploit the well-developed accurate theories of standard Gibbsian disordered hard spheres in direct space.
Hole Probability $P(r)$ in Disordered Many-Particle Configurations

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- What about disordered hyperuniform systems? We know **not all disordered hyperuniform systems prohibit arbitrarily large holes** (e.g., $P(r) = \exp[-\kappa(d)r^{d+1}]$ for fermionic gases.).
Stealthy Systems Cannot Tolerate Arbitrarily Large Holes

We have shown that disordered stealthy hyperuniform configurations cannot tolerate arbitrarily large holes in the infinite-system-size. Indeed, the maximum hole size $R_{max}$ is inversely proportional to $K$ for any dimension:

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Disordered stealthy materials with the largest value of $\chi$ lead to the best optical, transport and mechanical properties.
Hyperuniformity and Jammed Packings

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A 3D maximally random jammed (MRJ) packing is a prototypical glass in that it is maximally disordered but perfectly rigid (infinite elastic moduli).

Such packings of identical spheres are hyperuniform ($S(k) \sim k$ for small $k$) with quasi-long-range (QLR) pair correlations in which $h(r)$ decays as $-1/r^4$ (Donev, Stillinger & Torquato, PRL, 2005).

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Apparently, hyperuniform QLR correlations with decay $-1/r^{d+1}$ are a universal feature of general MRJ packings in $\mathbb{R}^d$.

Zachary, Jiao and Torquato, PRL (2011): ellipsoids, superballs, sphere mixtures
Berthier et al., PRL (2011); Kurita and Weeks, PRE (2011); Dreyfus et al., PRE (2015): sphere mixtures
Jiao and Torquato, PRE (2011): polyhedra
Remark About Excited States

For a system in equilibrium, the compressibility relation relates the isothermal compressibility $\kappa_T$ to the structure factor at $k = 0$:

$$S(0) = \rho k_B T \kappa_T,$$

where $k_B$ is Boltzmann's constant. Because $\kappa_T$ is bounded according to $\kappa_T = \rho^{-2}$ for finite $\rho$, $S(0) = 0$ because $T = 0$, which clearly must be the case for stealthy ground states.

Now consider excited states infinitesimally close to the stealthy ground states, i.e., when temperature $T$ is positive and infinitesimaly small.

Under the assumption that the structure of such excited states will be infinitesimally near the ground-state configurations, we can estimate to an excellent approximation how $S(0)$ varies with $T$ for such excited states:

$$S(0) \sim c(d) \chi T$$

in units $k_B = v_0 = 1$. Moreover, it is expected that this positive value of $S(0)$ will be the uniform value of $S(k)$ for $0 \leq k \leq K$.

This behavior of $S(k)$ has indeed been verified by molecular dynamics simulations in the canonical ensemble:

![Graph showing the variation of $S(0)$ with $T$ with simulation and theory data points.](image)