Hyperuniform States of Matter

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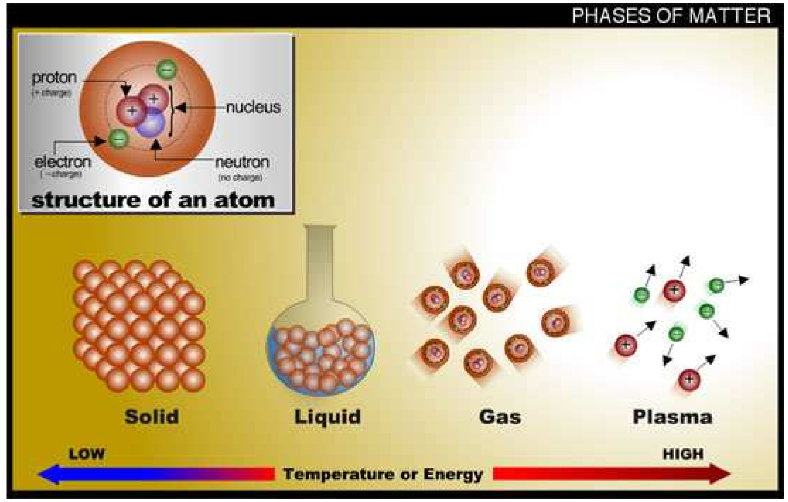
Review article: S. Torquato, "Hyperuniform States of Matter," Physics Reports, 745, 1 (2018).

States (Phases) of Matter



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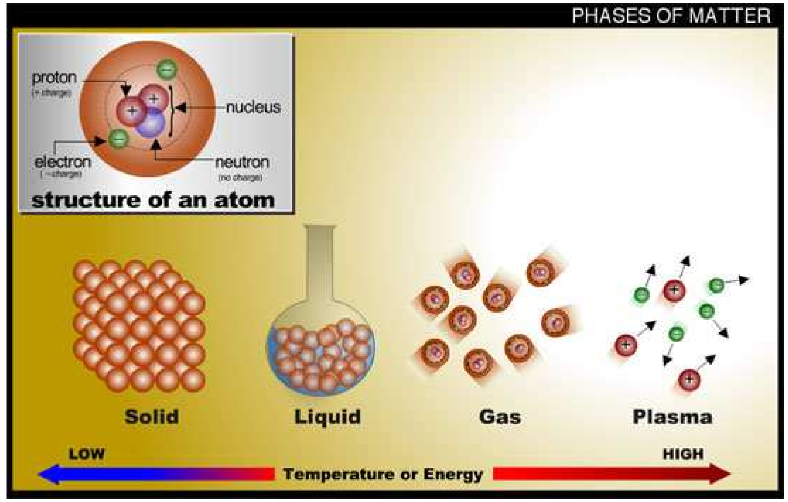

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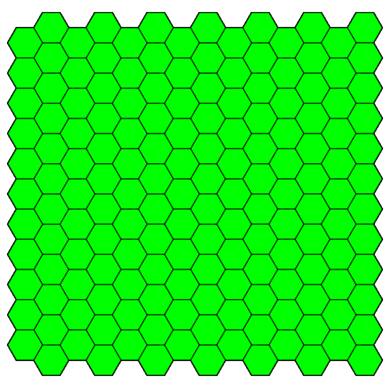
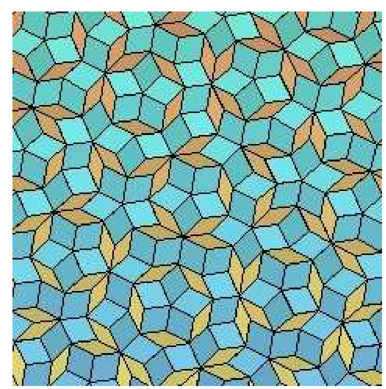
- **We now know there are a multitude of distinguishable states of matter, e.g., quasicrystals and liquid crystals, which break the continuous translational and rotational symmetries of a liquid differently from a solid crystal.**



Crystal tiling



Quasicrystalline tiling

- **Quasicrystals taught us how to generalize the concept of long-range order.**

Qualitatively, What is Hyperuniformity?

- **A hyperuniform many-particle system is one in which large-scale density fluctuations are greatly suppressed compared to those of typical disordered systems (e.g., liquids).**

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- Hyperuniformity provides a unified means of categorizing and characterizing crystals, quasicrystals and special disordered systems. Thus, hyperuniformity concept generalizes our traditional notions of long-range order.

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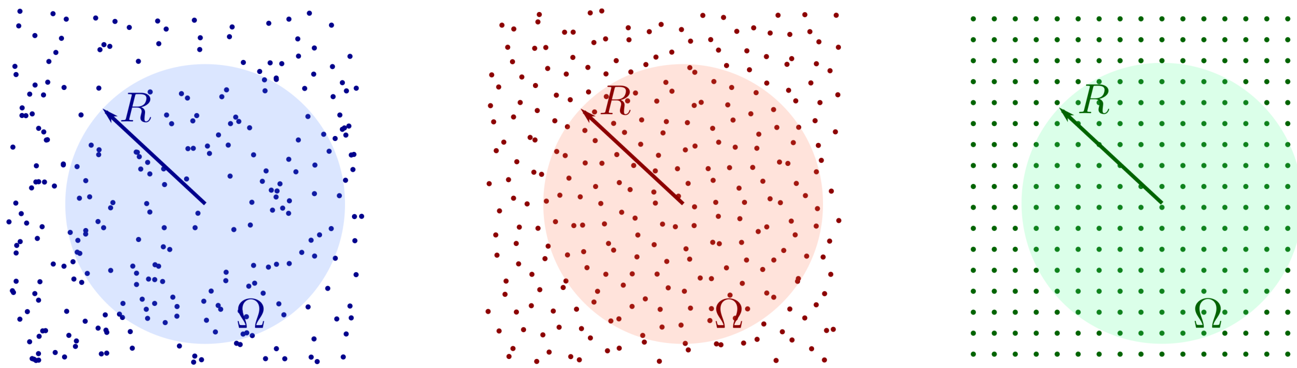
- All **perfect crystals** (periodic systems) and **quasicrystals** are hyperuniform.
- **Hyperuniformity** provides a **unified means of categorizing and characterizing** crystals, quasicrystals and **special disordered** systems. Thus, hyperuniformity concept generalizes our **traditional** notions of **long-range order**.
- **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 way 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.

Quantitatively, What is Hyperuniformity?

Torquato and Stillinger, Phys. Rev. E (2003)

- **Points can represent molecules of a material, stars in a galaxy, or trees in a forest. Let $\Omega \subset \mathbb{R}^d$ represent a 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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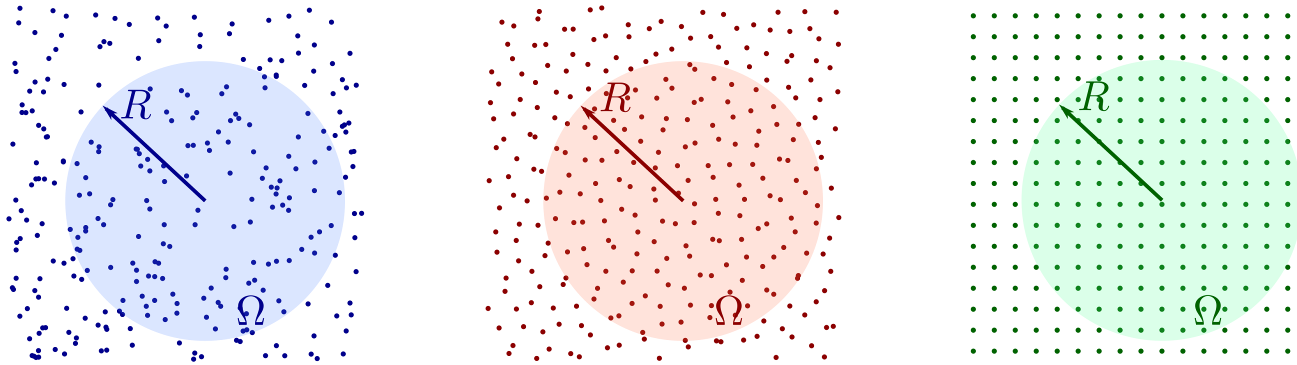

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- **For Poisson point patterns and many disordered point patterns, $\sigma^2(R) \sim R^d$.**
- **We call point patterns whose variance grows more slowly than R^d (window volume) hyperuniform. Implies that scattering or structure factor vanishes in infinite-wavelength limit, i.e.,**

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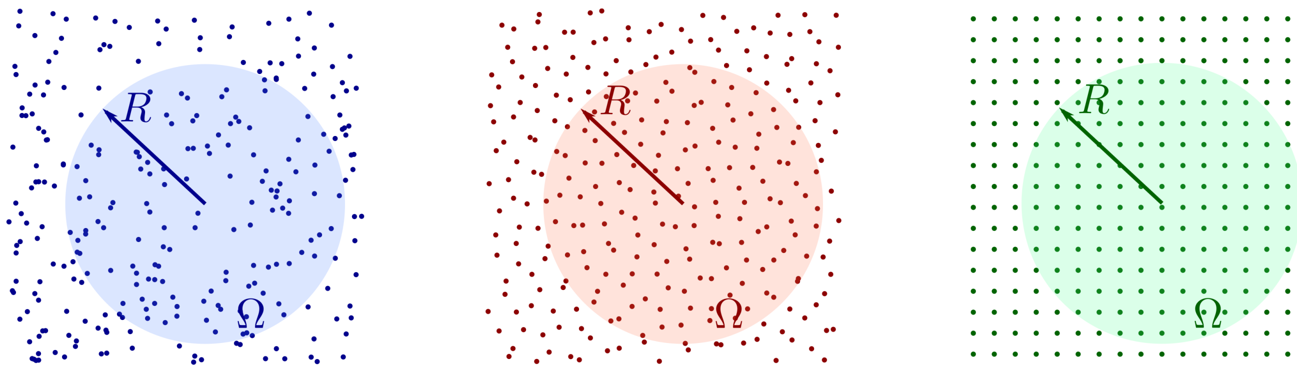

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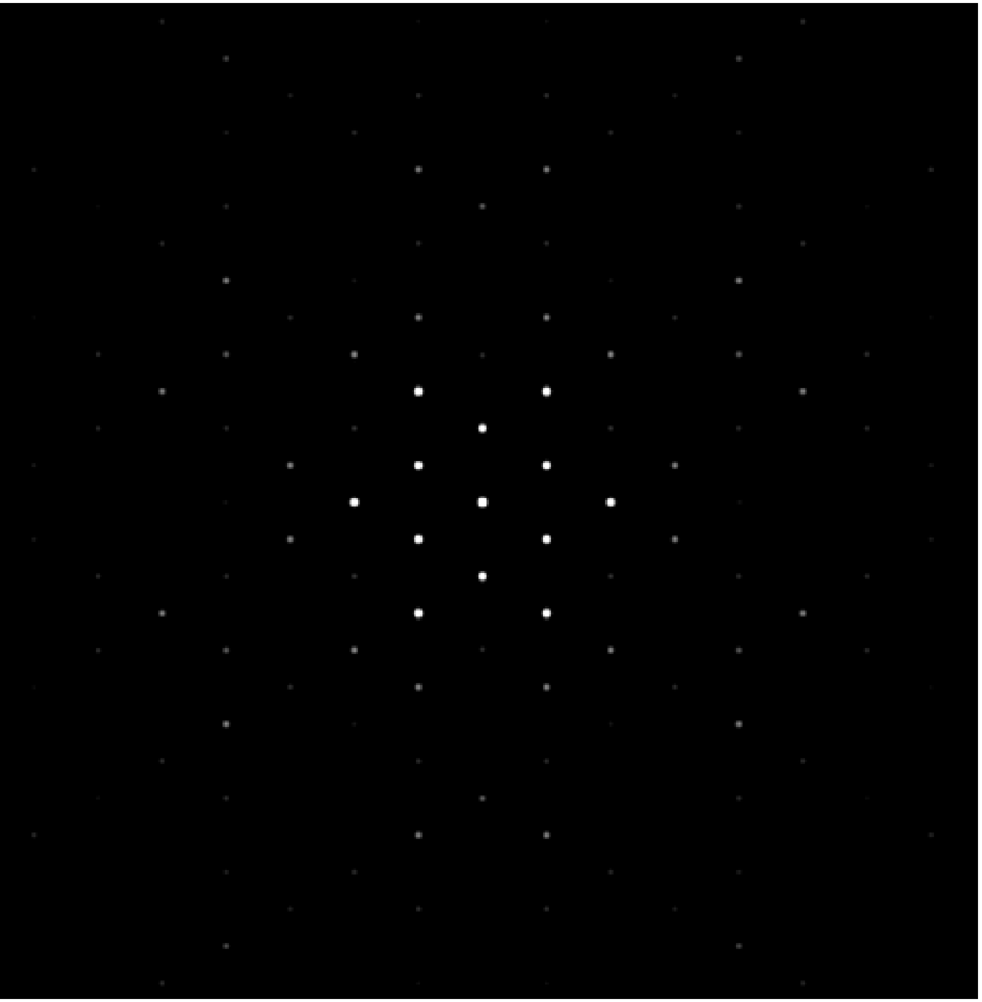
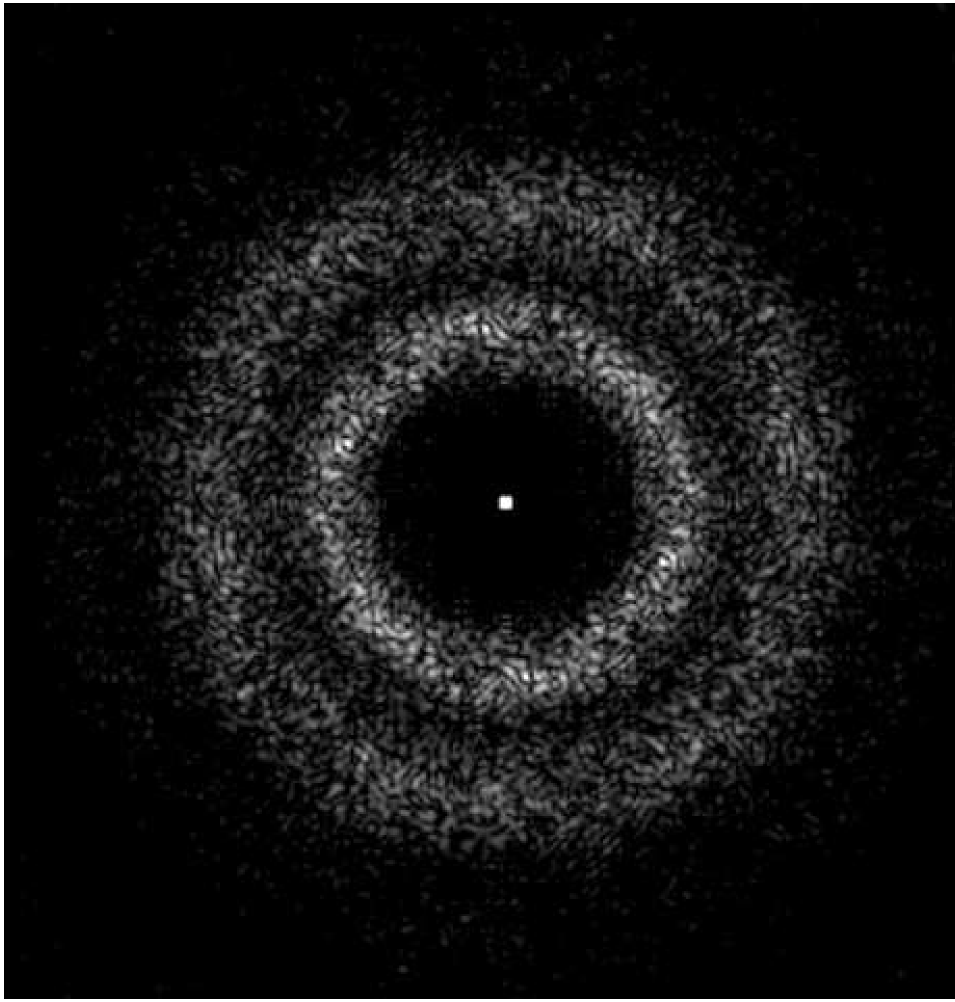
$$S(\mathbf{k}) \to 0 \text{ for } |\mathbf{k}| \to 0.$$

- **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

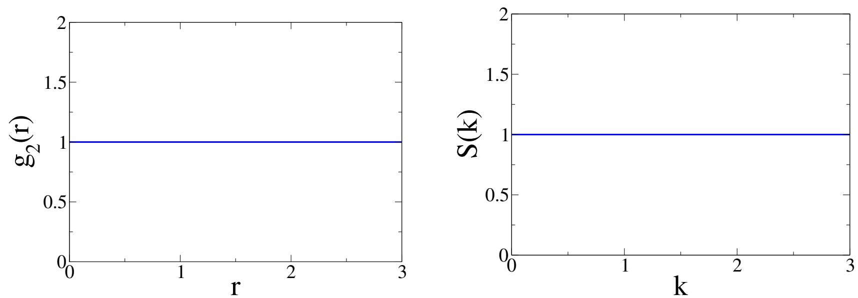




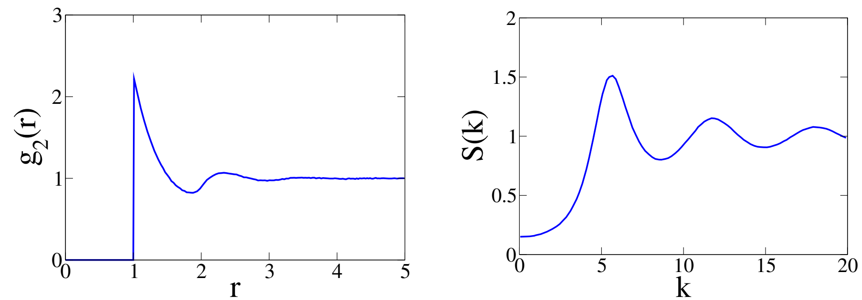
Pair Statistics in Direct and Fourier Spaces

- For particle systems in $\mathbb{R}^d$ at **number density** ρ , $g_2(r)$ is a **nonnegative radial function** that is proportional to the **probability density of pair distances** r.
- The nonnegative **structure factor** $S(k) \equiv 1 + \rho\tilde{h}(k)$ is obtained from the Fourier transform of $h(r) = g_2(r) - 1$, which we denote by $\tilde{h}(k)$.

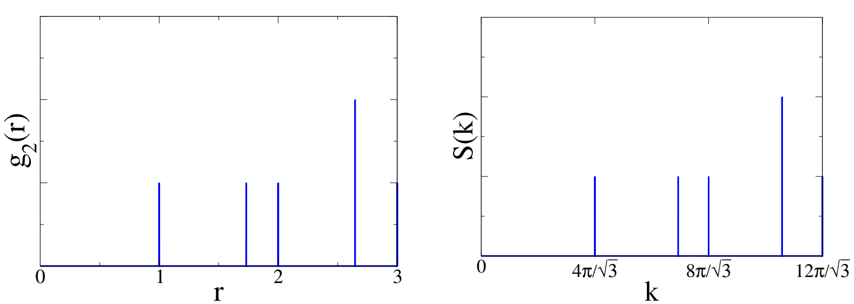
Poisson Distribution (Ideal Gas)



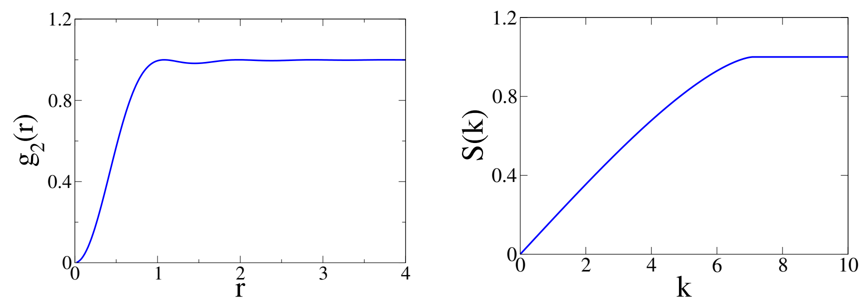
Liquid



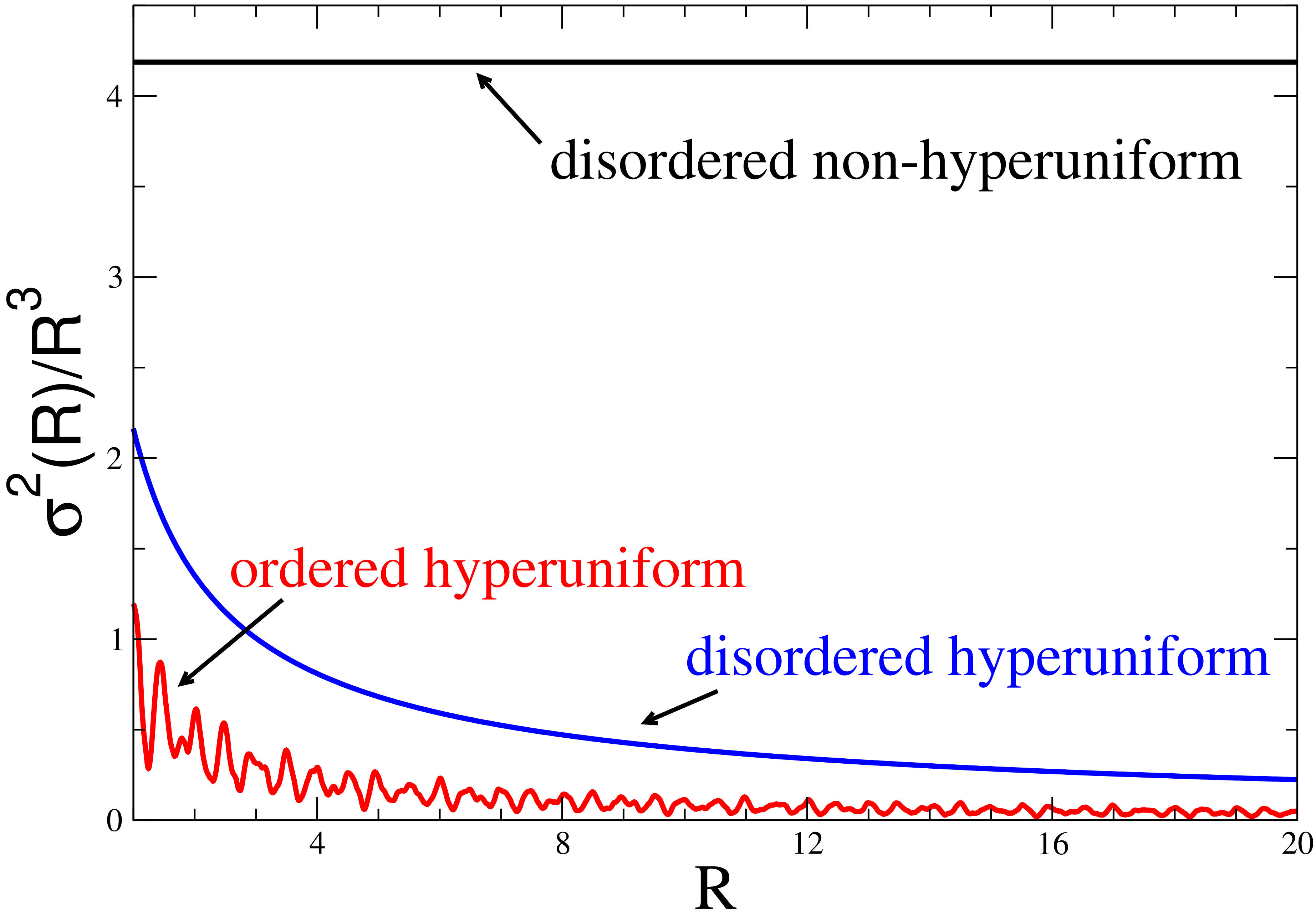
Lattice



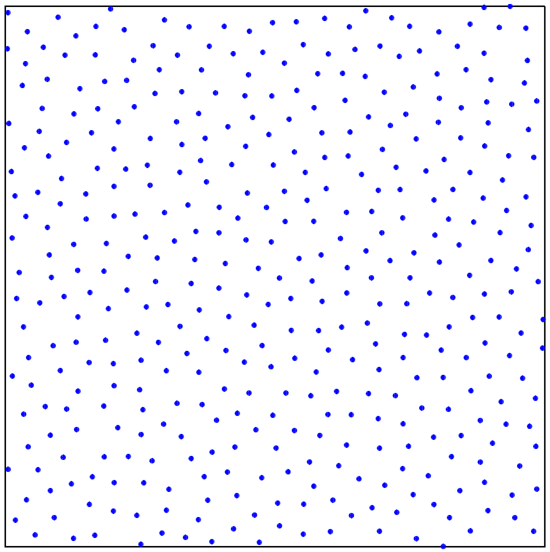
Disordered Hyperuniform System

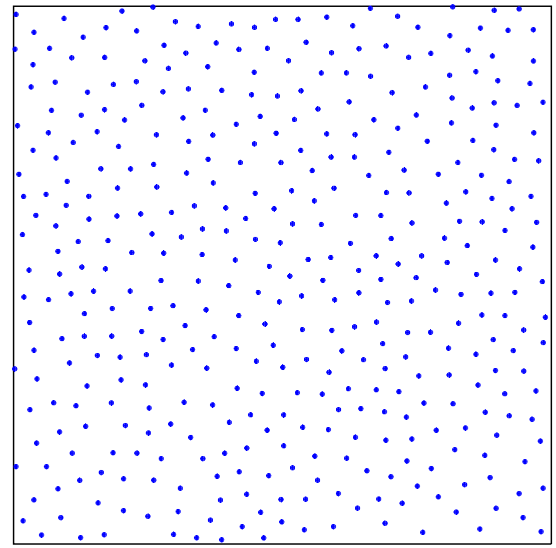


Scaled Number Variance for 3D Systems at Unit Density



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 ρ,

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

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

Some observations:

  - Any ground state $(T = 0)$ in which the isothermal compressibility κ_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.

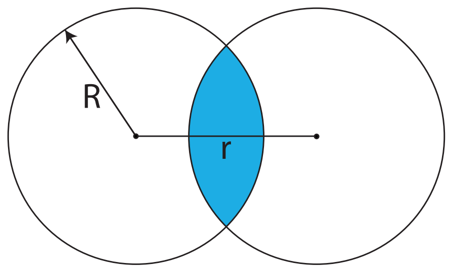
- Note that a system at a thermal critical point is anti-hyperuniform in the sense that $\lim_{k \to 0} S(k) = +\infty$.

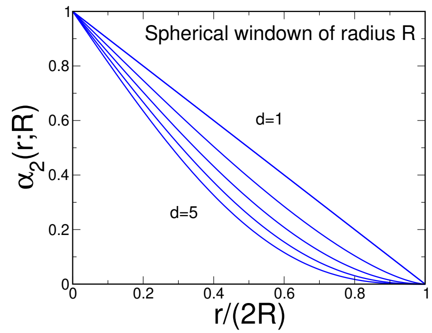
ENSEMBLE-AVERAGE FORMULATION

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

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

$\alpha_2(\mathbf{r};R)$**- scaled intersection volume of 2 windows of radius R separated by $\mathbf{r}$**





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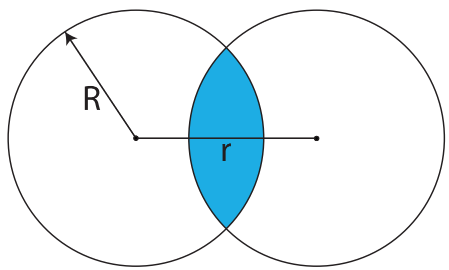

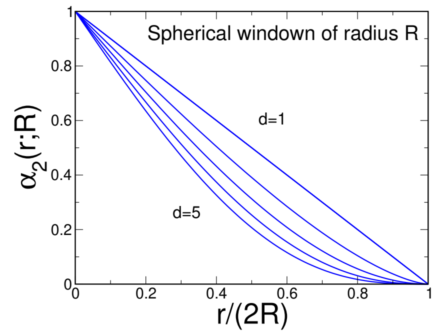

- **For a certain class of systems and large R, we can show**

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

where A and B are the "volume" and "surface-area" coefficients:

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

  - **Hyperuniform**: $A = 0$, $B > 0$ $\Longrightarrow$ **Sum rule:** $\rho\int_{\mathbb{R}^d} h(\mathbf{r})d\mathbf{r} = -1$
  - **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

- $h(\mathbf{r})$ **can be divided into direct correlations, via function** $c(\mathbf{r})$**, and indirect correlations:**

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

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

- This is the inverse of the behavior at liquid-gas (or magnetic) critical points, where $h(\mathbf{r})$ is long-ranged (compressibility or susceptibility diverges) and $c(\mathbf{r})$ 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,

$$\begin{aligned} c(\mathbf{r}) &\sim -\frac{1}{r^{d-2+\eta}} \quad (r\to\infty), \qquad \tilde{c}(\mathbf{k}) \sim -\frac{1}{k^{2-\eta}} \quad (k\to 0), \\ h(\mathbf{r}) &\sim -\frac{1}{r^{d+2-\eta}} \quad (r\to\infty), \qquad S(\mathbf{k}) \sim k^{2-\eta} \quad (k\to 0), \end{aligned}$$

  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

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

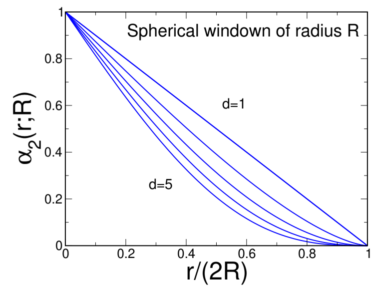
- However, long-range interactions are not required to drive a nonequilibrium system to a disordered hyperuniform state.

SINGLE-CONFIGURATION FORMULATION & GROUND STATES

- **We showed**

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

where $\alpha_2(r;R)$ can be viewed as a repulsive pair potential:



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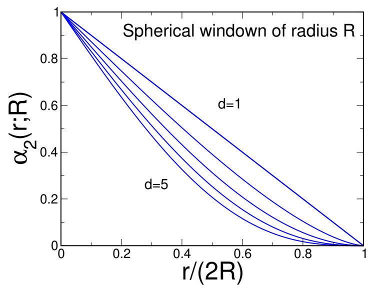

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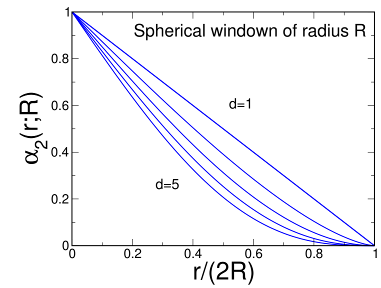
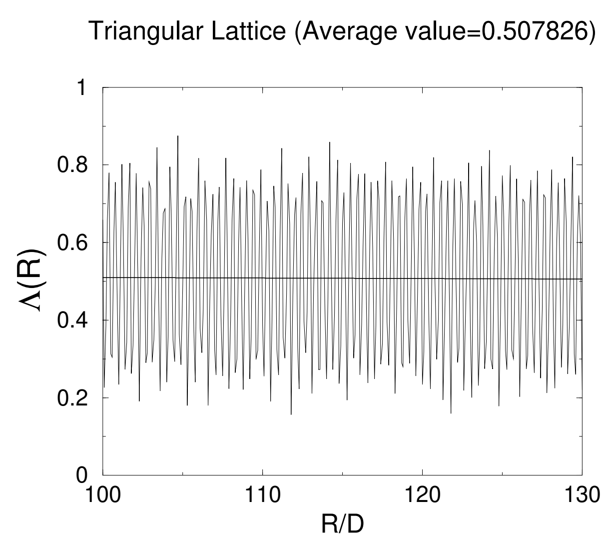


- **Finding global minimum of $\sigma^2(R)$ equivalent to finding ground state.**
- **For large R, in the special case of hyperuniform systems,**

$$\sigma^2(R) = \Lambda(R)\left(\frac{R}{D}\right)^{d-1} + \mathcal{O}\left(\frac{R}{D}\right)^{d-3}$$



Hyperuniformity, Number Theory and Sphere Packings

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- **We showed that for a lattice**

$$\sigma^2(R) = \sum_{\mathbf{q}\neq\mathbf{0}} \left(\frac{2\pi R}{q}\right)^d [J_{d/2}(qR)]^2, \qquad \overline{\Lambda} = 2^d \pi^{d-1} \sum_{\mathbf{q}\neq\mathbf{0}} \frac{1}{|\mathbf{q}|^{d+1}}.$$

- **Epstein zeta function for a lattice is defined by**

$$Z(s) = \sum_{\mathbf{q}\neq\mathbf{0}} \frac{1}{|\mathbf{q}|^{2s}}, \qquad \text{Re } s > d/2.$$

Summand can be viewed as an inverse power-law potential. For lattices, minimizer of $Z(d+1)$ is the lattice dual to the minimizer of $\overline{\Lambda}$.

Sarnak and Strömbergsson (2006)

- **Surface-area coefficient $\overline{\Lambda}$ provides useful way to rank order crystals, quasicrystals and special correlated disordered point patterns.**

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Sarnak and Strömbergsson (2006)

- **Surface-area coefficient $\overline{\Lambda}$ provides useful way to rank order crystals, quasicrystals and special correlated disordered point patterns.**
- **For certain d, minimizer of Epstein zeta function is related to the optimal sphere packings.**

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.**

Pattern	$\overline{\Lambda}$
Integer Lattice	$1/6 \approx 0.166667$
Step+Delta-Function g_2	3/16 =0.1875
Fibonacci Chain*	0.2011
Step-Function g_2	$1/4 = 0.25$
Randomized Lattice	$1/3 \approx 0.333333$

*Zachary & Torquato (2009)

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- More recent work on hyperuniformity of quasicrystals: Ogŭz, Socolar, Steinhardt and Torquato (2016).

Quantifying Suppression of Density Fluctuations at Large Scales: 2D

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

2D Pattern	$\overline{\Lambda}/\phi^{1/2}$
Triangular Lattice	0.508347
Square Lattice	0.516401
Honeycomb Lattice	0.567026
Kagomé Lattice	0.586990
Penrose Tiling*	0.597798
Step+Delta-Function g_2	0.600211
Step-Function g_2	0.848826
One-Component Plasma	1.12838

*Zachary & Torquato (2009)

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.**

Pattern	$\overline{\Lambda}/\phi^{2/3}$
BCC Lattice	1.24476
FCC Lattice	1.24552
HCP Lattice	1.24569
SC Lattice	1.28920
Diamond Lattice	1.41892
Wurtzite Lattice	1.42184
Damped-Oscillating g_2	1.44837
Step+Delta-Function g_2	1.52686
Step-Function g_2	2.25

- **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**

$$S(k) \sim |\mathbf{k}|^{\alpha}, \qquad (|\mathbf{k}| \to \mathbf{0})$$

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)

$$\sigma^2(R) \sim \begin{cases} R^{d-1}, & \alpha > 1 \qquad (\textbf{CLASS I}) \\ R^{d-1}\ln R, & \alpha = 1 \qquad (\textbf{CLASS II}) \\ R^{d-\alpha}, & 0 < \alpha < 1 \qquad (\textbf{CLASS III}) \end{cases}$$

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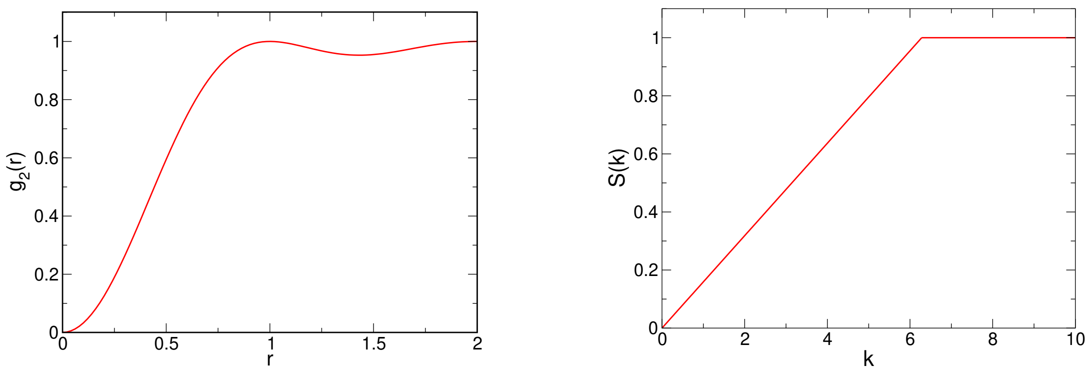
- **Class I**: $\sigma^2(R) \sim R^{d-1}$: Crystals, quasicrystals, **stealthy disordered ground states**, charged systems, g_2-invariant disordered point processes.
- **Class II**: $\sigma^2(R) \sim R^{d-1}\ln(R)$: Quasicrystals, classical disordered ground states, **zeros of the Riemann zeta function, 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.

1D Disordered Hyperuniform Systems

- 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;
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- 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

$$\Phi_N(\mathbf{r}^N) = \frac{N}{2}\sum_{i=1}^{N} |\mathbf{r}_i|^2 - \sum_{i\leq j}^{N} \ln(|\mathbf{r}_i - \mathbf{r}_j|)\,.$$

Sandier and Serfaty, Prob. Theory & Related Fields (2015)

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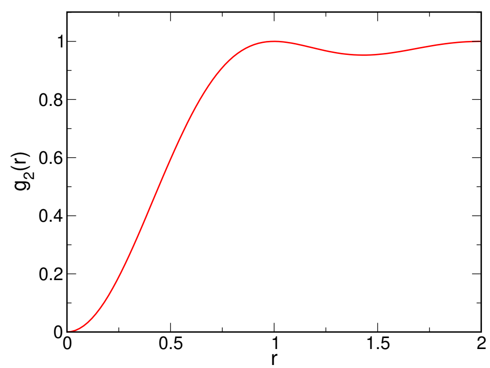

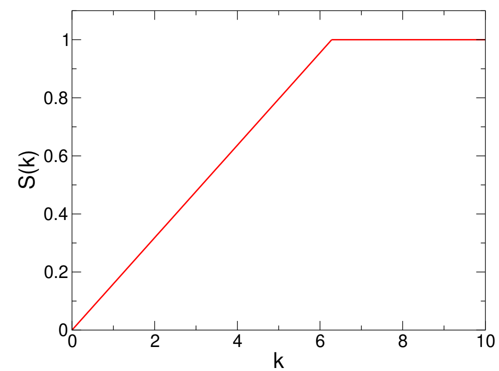

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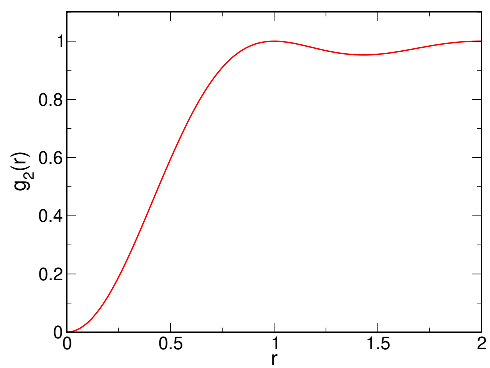

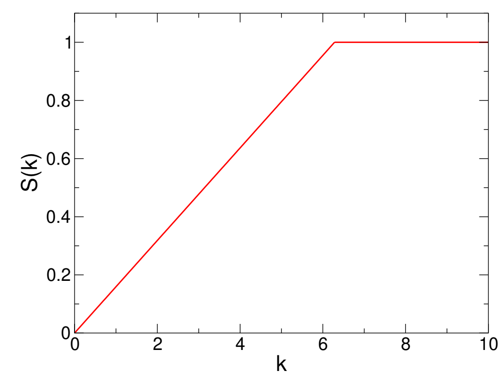

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- **Recently showed that prime numbers in a distinguished limit are hyperuniform**

Recent 2D and 3D Examples of Disordered Hyperuniform Systems

Physical Examples

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Nearly Hyperuniform Disordered Systems

- Amorphous Silicon (nonequilibrium states): Henja et al. PRB (2013)
- Polymers (equilibrium states): Xu et al. Macromolecules (2016); Chremos et al. Ann. Phys. (2017)
- Amorphous Ices (nonequilibrium states): Martelli et al. PRL (2017)

Hyperuniformity and Jammed Packings

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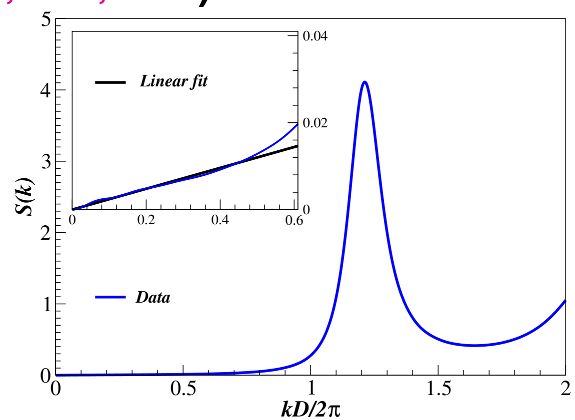
- **Such packings of identical spheres have been shown to be hyperuniform with quasi-long-range (QLR) pair correlations in which $h(r)$ decays as $-1/r^4$ (Donev, Stillinger & Torquato, PRL, 2005).**



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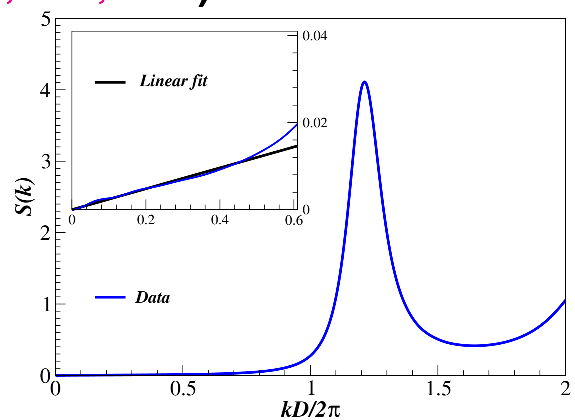

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- **Apparently, hyperuniform QLR correlations with decay $-1/r^{d+1}$ are a universal feature of general MRJ packings in $\mathbb{R}^d$.**

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

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Hyperuniformity and Spin-Polarized Free Fermions

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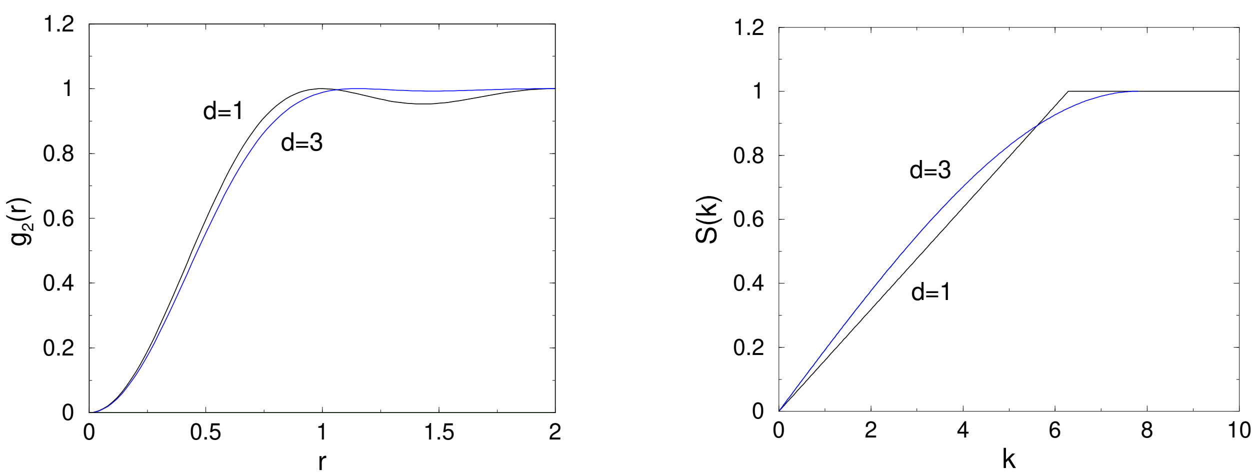
- **We provide exact generalizations of such a point process in d-dimensional Euclidean space $\mathbb{R}^d$ and the corresponding n-particle correlation functions, which correspond to those of spin-polarized free fermionic systems in $\mathbb{R}^d$.**



$$g_2(r) = 1 - \frac{2\Gamma(1+d/2)\cos^2\left(rK - \pi(d+1)/4\right)}{K\,\pi^{d/2+1}\,r^{d+1}} \qquad (r \to \infty)$$

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

Torquato, Zachary & Scardicchio, J. Stat. Mech., 2008

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).**
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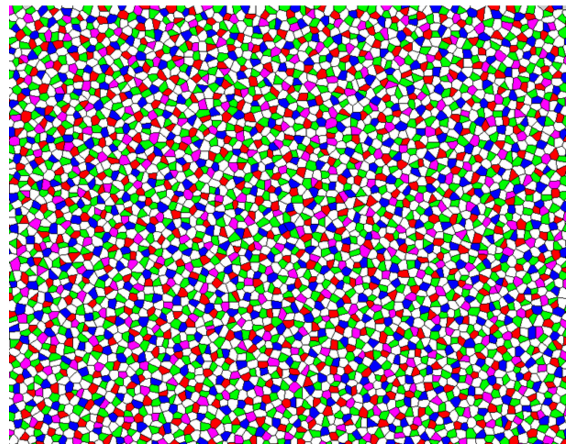
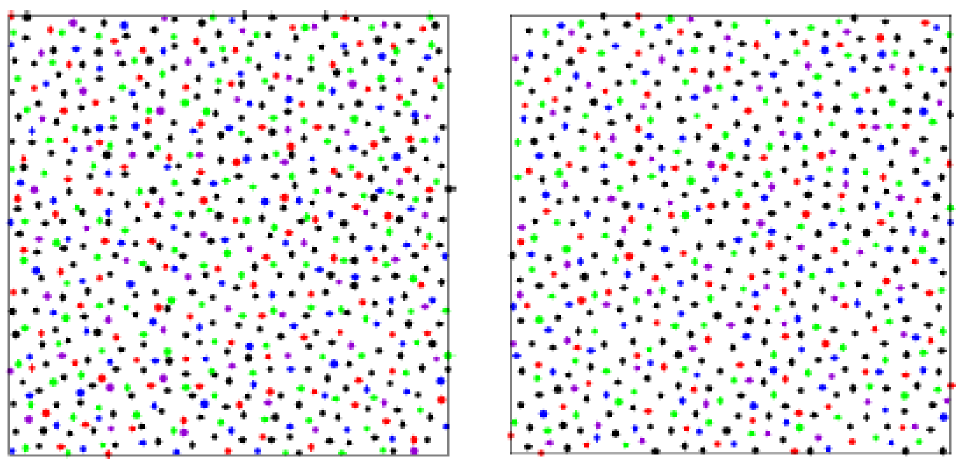


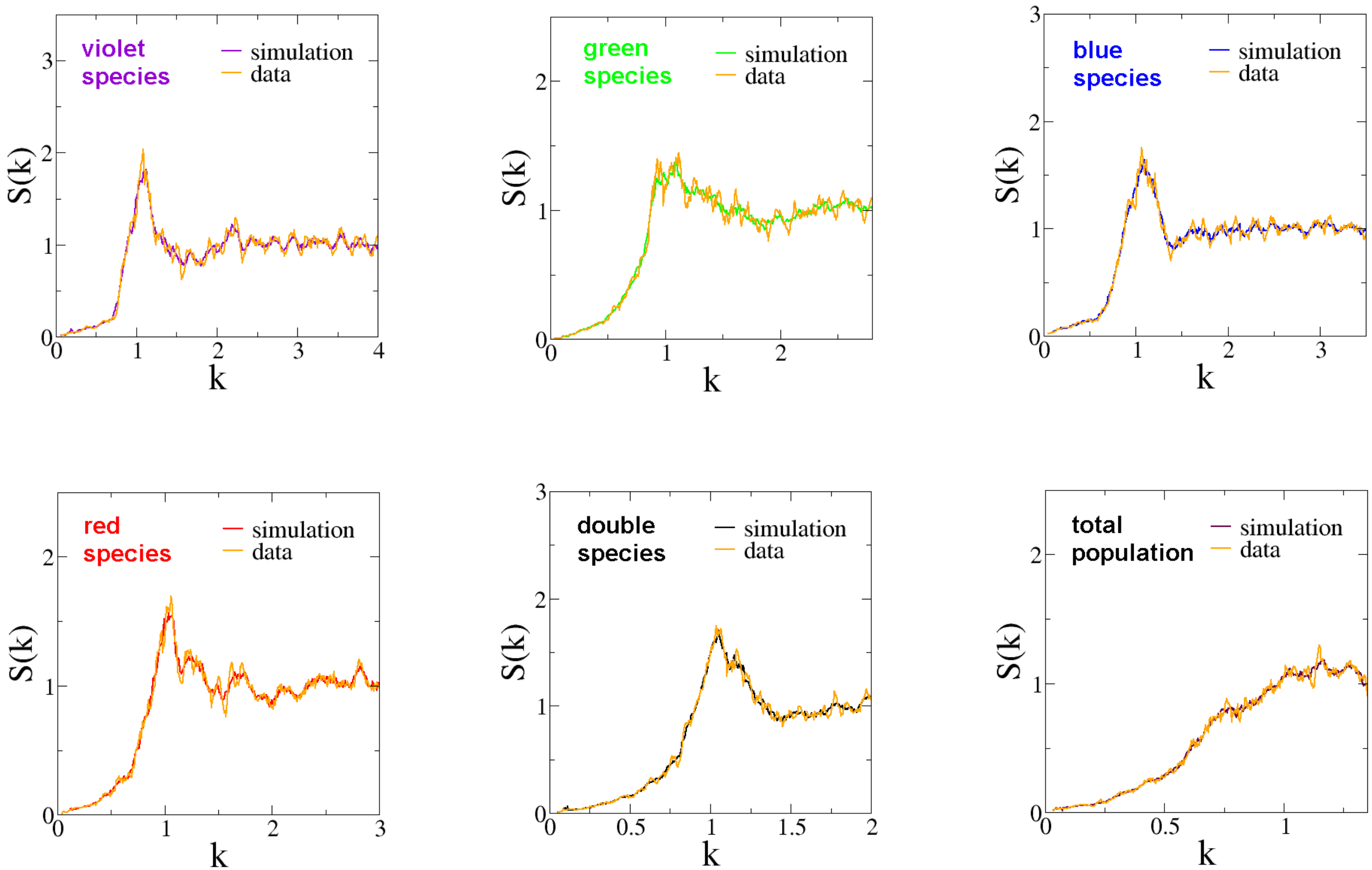
5 Cone Types



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).**



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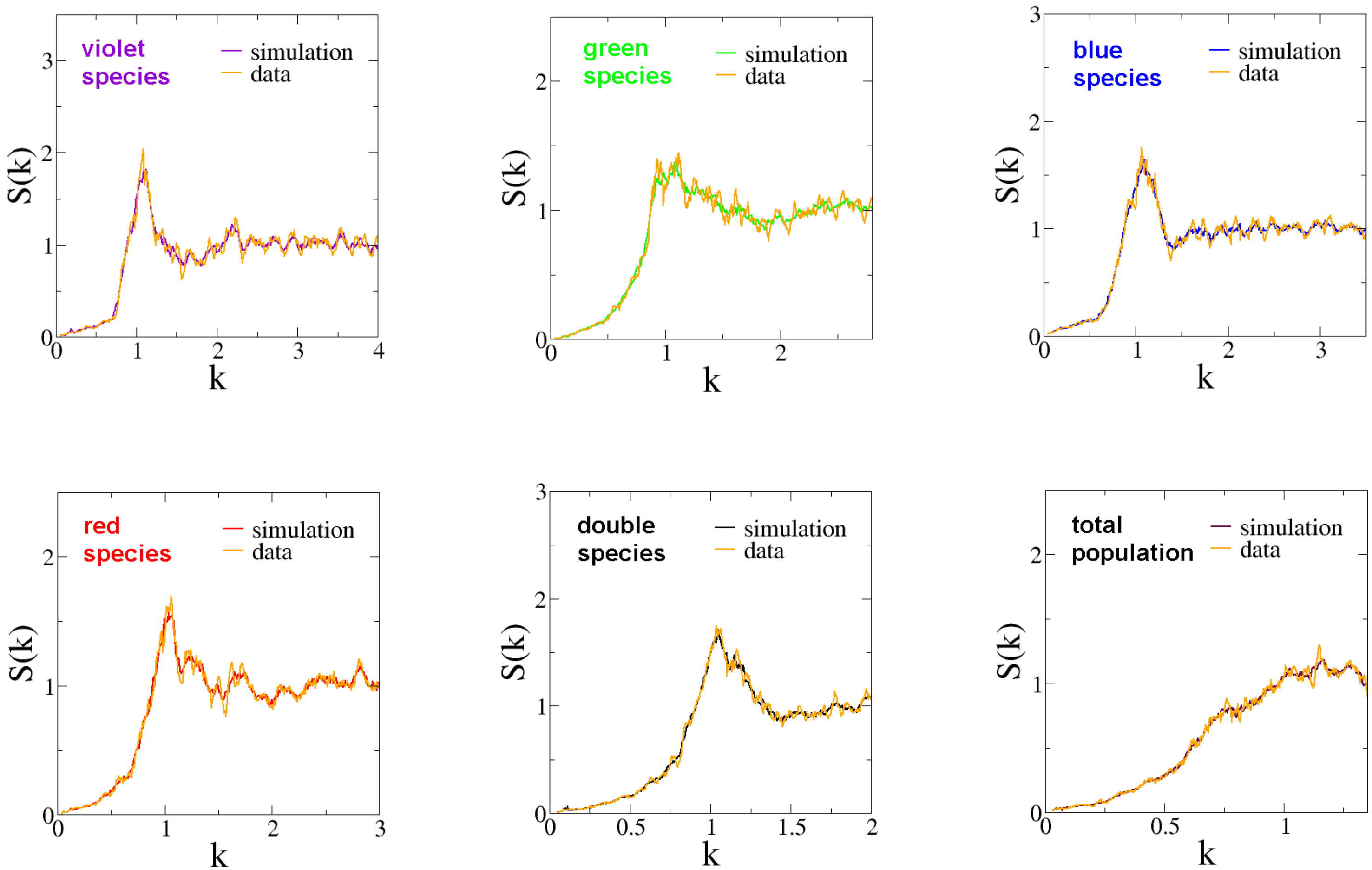

- **Recently showed that multihyperuniformity can be rigorously achieved via hard-disk plasmas (Lomba, Weis and Torquato, PRE 2018).**

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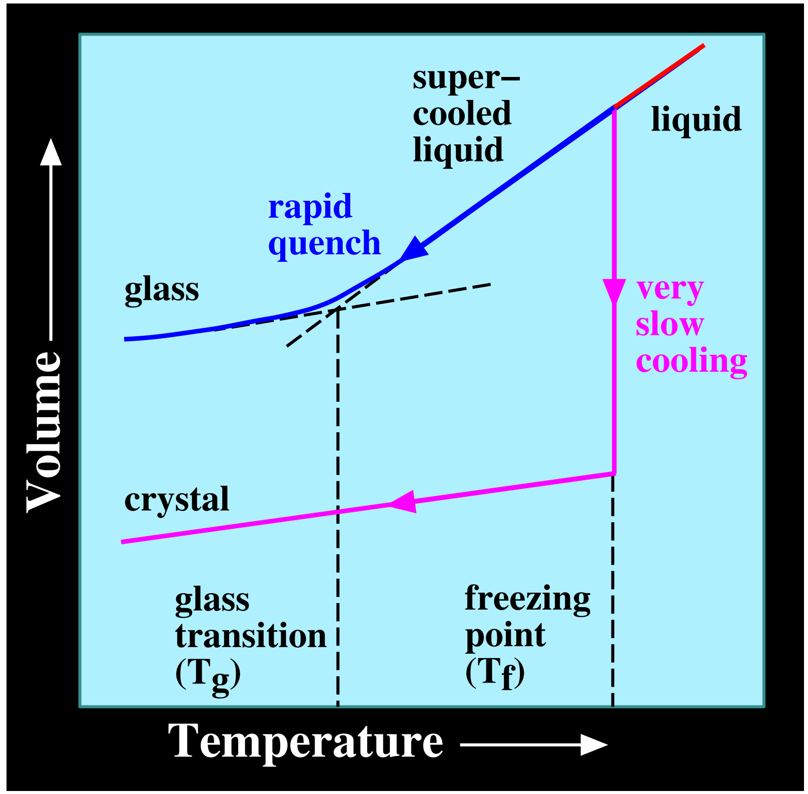

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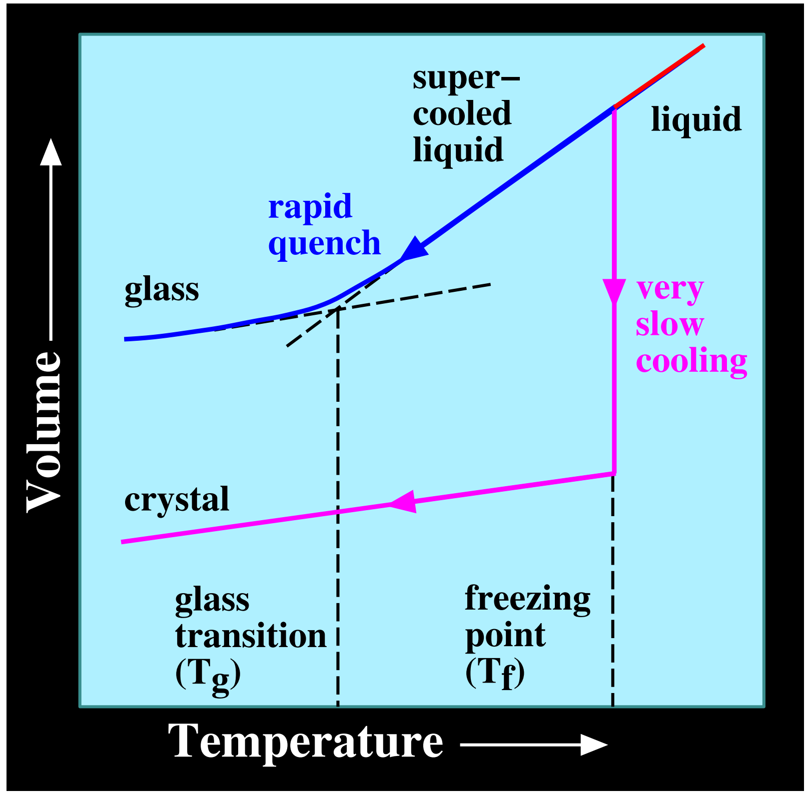

- Typically, ground states are **periodic with high crystallographic symmetries**.
- Can classical ground states derived from **nontrivial** interactions ever be **disordered** and hence **hyperuniform**?

Disordered Hyperuniform Ground State 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 Ω under periodic boundary conditions) with a pair potential $v(\mathbf{r})$ that is bounded with Fourier transform $\tilde{v}(\mathbf{k})$.

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  The total energy is

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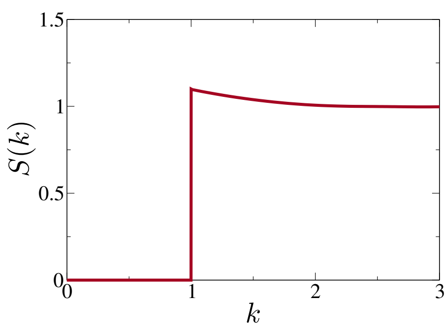
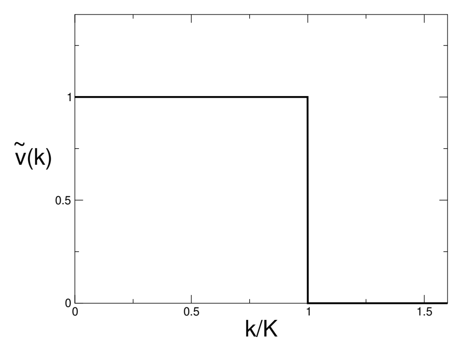
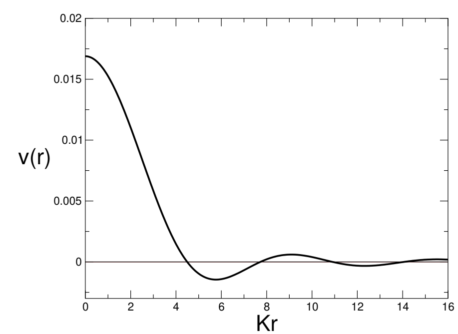
- **For $\tilde{v}(\mathbf{k})$ positive $\forall\ 0 \leq |\mathbf{k}| \leq K$ and zero otherwise, finding configurations in which $S(\mathbf{k})$ is constrained to be zero where $\tilde{v}(\mathbf{k})$ has support is equivalent to globally minimizing $\Phi(\mathbf{r}^N)$.**







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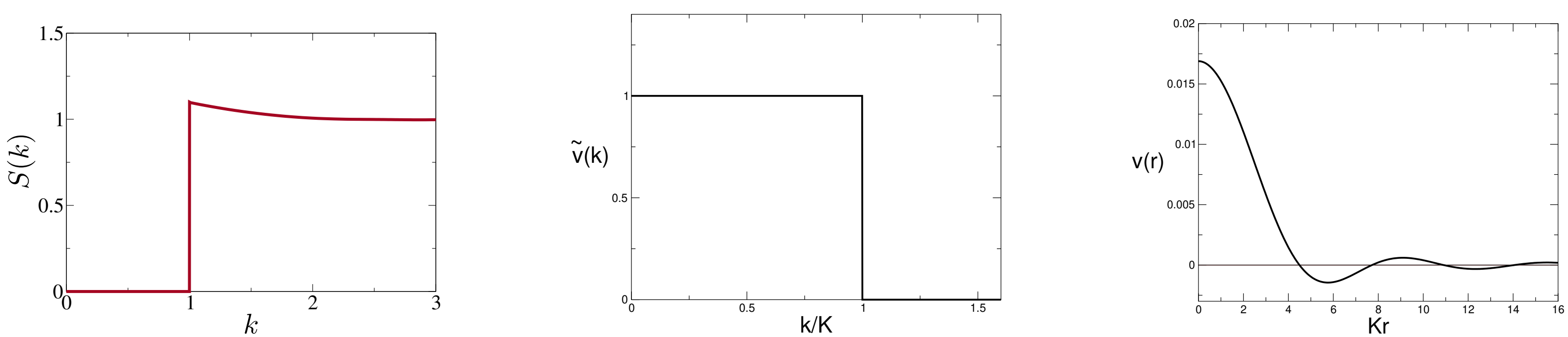

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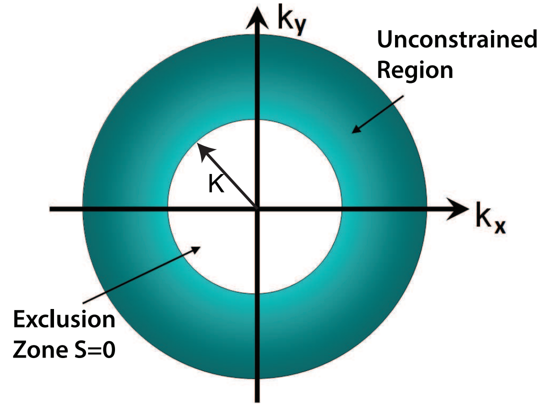
- **Stealthy patterns can be tuned by varying the parameter χ: ratio of number of constrained degrees of freedom to the total number of degrees of freedom, $d(N-1)$.**

Creation of Disordered Stealthy Ground States



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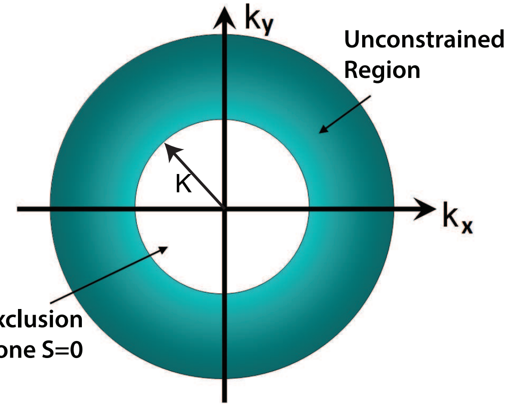
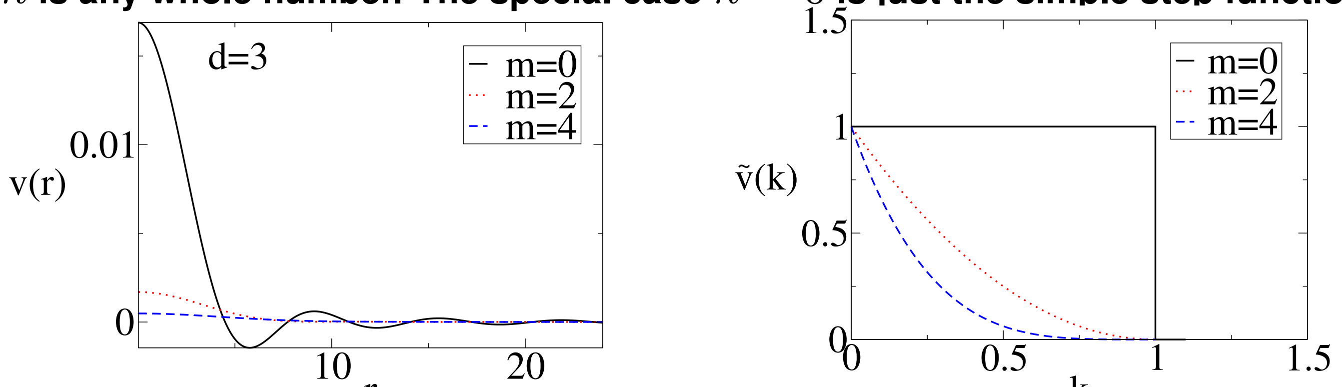


- One class of stealthy potentials involves the following **power-law** form:

$$\tilde{v}(k) = v_0(1 - k/K)^m \, \Theta(K - k),$$

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:

$$v(r) \sim \frac{\cos(r)}{r^2} \qquad (m = 0)$$

$$v(r) \sim \frac{1}{r^4} \qquad (m = 4)$$

- 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.

Creation of Disordered Stealthy Ground States via Collective Coordinat

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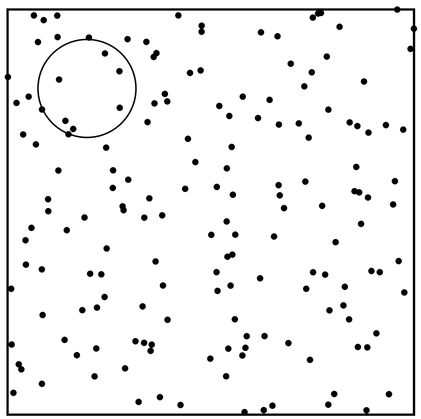
- For $0 \leq \chi < 0.5$, stealthy ground states are highly degenerate, disordered and isotropic. Success rate to achieve disordered ground states is 100%.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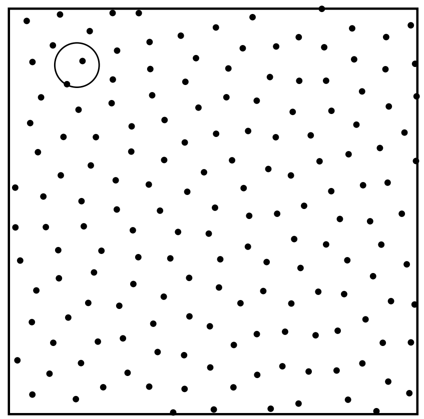
(b) $\chi = 0.41071$

As χ increases, short-range order increases. This suggests new order metric:

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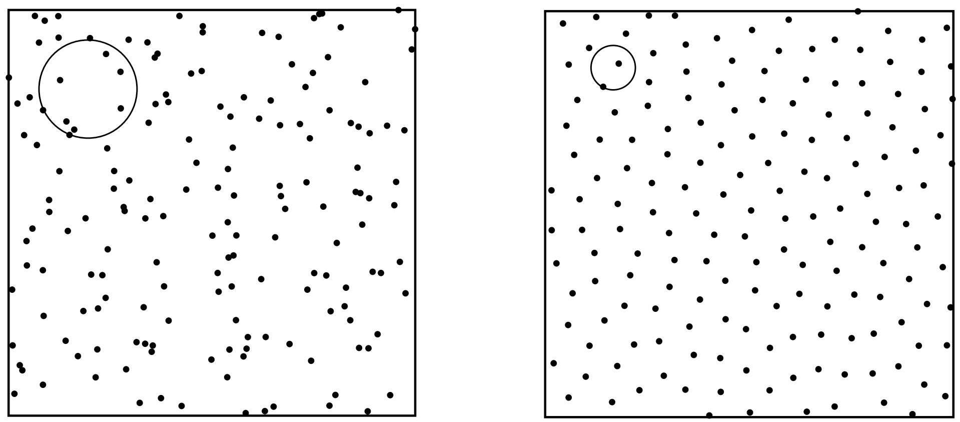

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- For $\chi > 1/2$ (no degrees of freedom), the system undergoes a transition to a crystal phase and the energy landscape becomes considerably more complex.

Animations

Ensemble Theory of Disordered Ground States

Torquato, Zhang & Stillinger, Phys. Rev. X, 2015

- **Nontrivial: Dimensionality of the configuration space depends on the number density ρ (or χ) 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 ρ in any d and showed how disordered degenerate ground states arise.**

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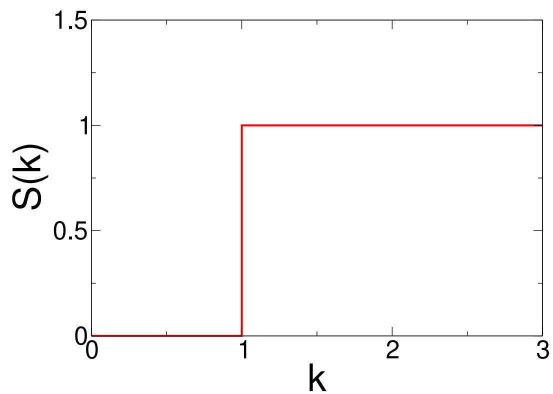
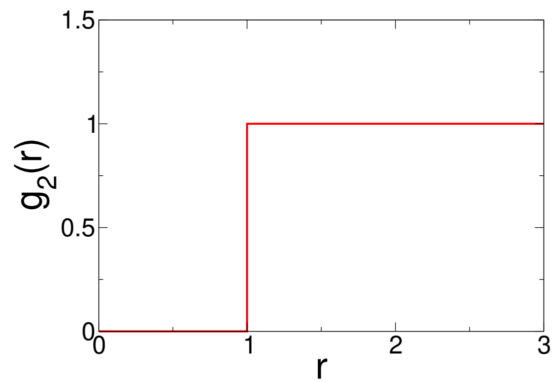
- From previous considerations, we that an important contribution to $S(k)$ is a simple hard-core step function $\Theta(k-K)$, which can be viewed as a **disordered hard-sphere system in Fourier space** in the limit that $\chi \sim 1/\rho$ tends to zero, i.e., as the number density ρ tends to infinity.





- That the structure factor must have the behavior

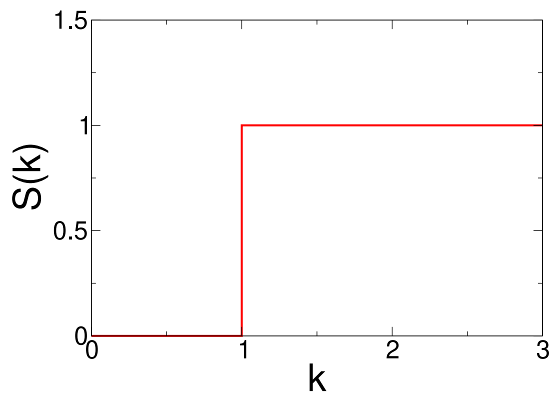
$$S(k) \rightarrow \Theta(k-K), \qquad \chi \rightarrow 0$$

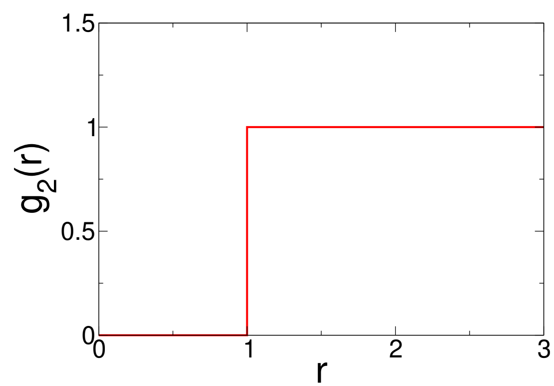
is perfectly reasonable; it is a **perturbation about the ideal-gas limit in which $S(k)=1$ for all k**.

Ensemble Theory of Disordered Ground States

Torquato, Zhang & Stillinger, Phys. Rev. X, 2015

- **Nontrivial**: Dimensionality of the configuration space depends on the number density ρ (or χ) 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 ρ 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 ρ tends to infinity.





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is perfectly reasonable; it is a **perturbation about the ideal-gas limit in which $S(k)=1$ for all k**.

- We make the **ansatz** that for sufficiently small χ, $S(k)$ 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

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

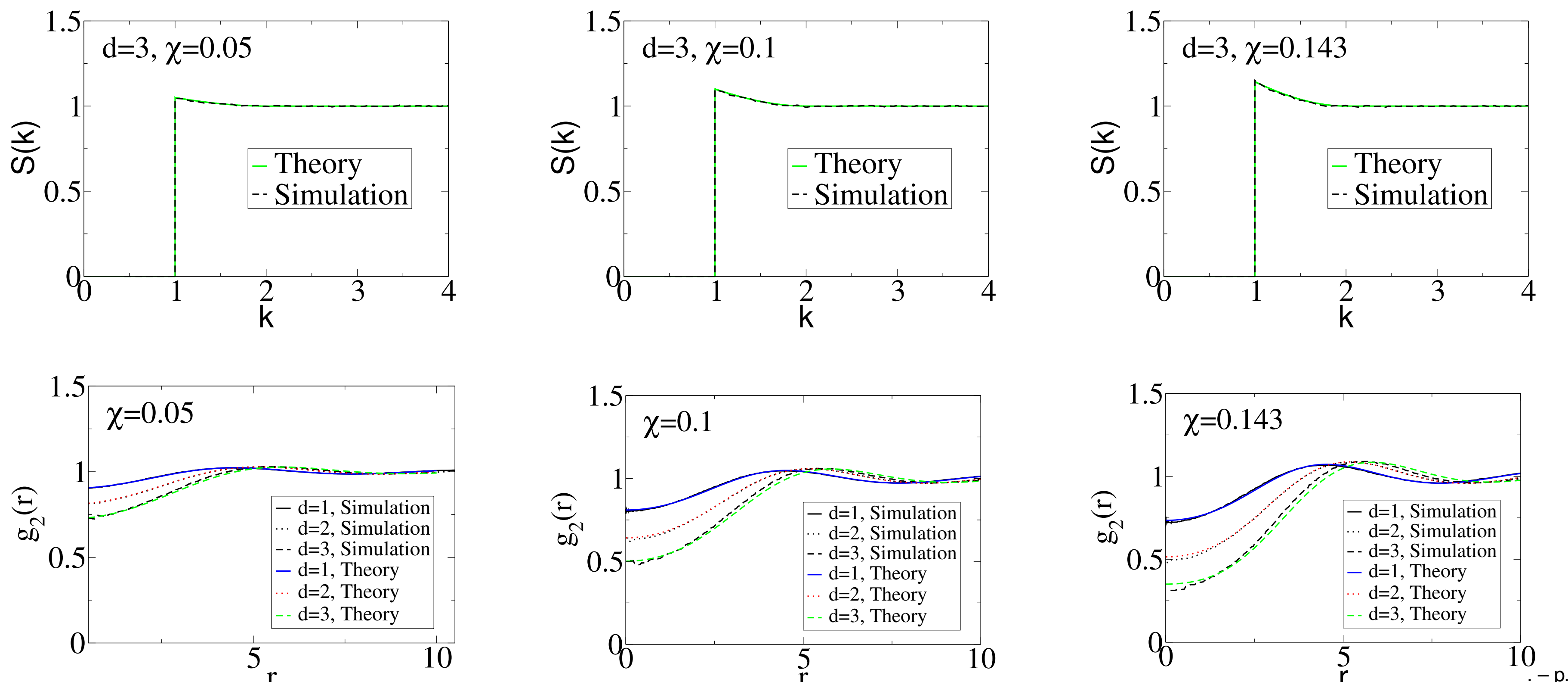
There is an Ornstein-Zernike integral eq. that defines FT of appropriate direct correlation function, $\tilde{C}(k)$:

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

where η 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.



Stealthy Disordered Ground States and Novel Materials

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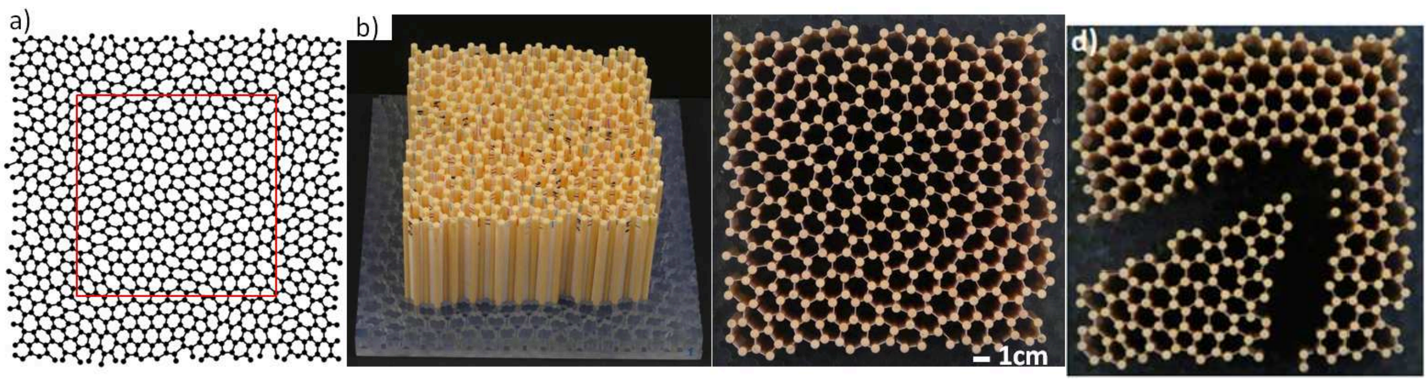

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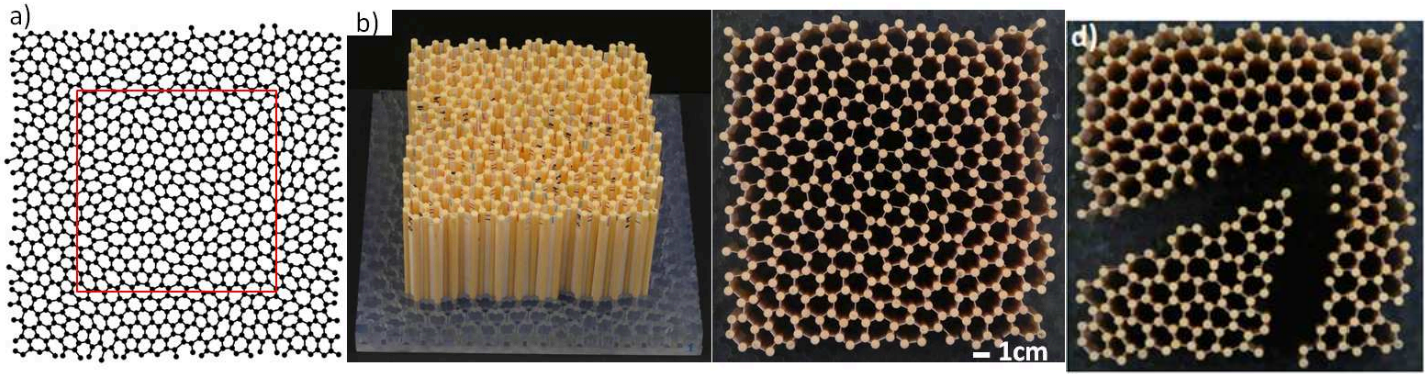

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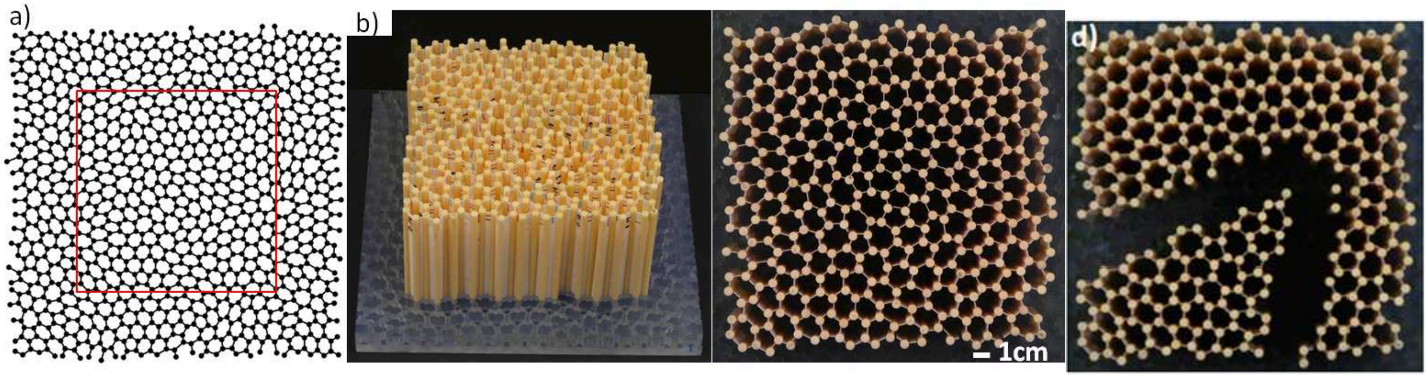

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- Nearly optimal transport and mechanical properties: Zhang et al. (2016); Torquato et al (2018).

WHY DO VERY LARGE DISORDERED STEALTHY HYPERUNIFORM

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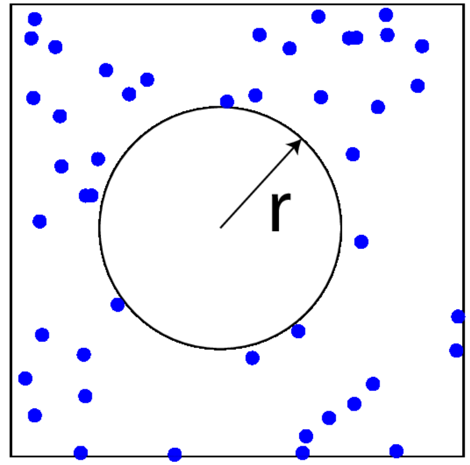
ANSWER: **Partly because they are disordered materials with some characteristics of crystals, including unusual "hole" statistics, i.e., holes of arbitrarily large size are prohibited in thermodynamic limit.**

Zhang, Stillinger and Torquato, Soft Matter (2017)
Ghosh and Lebowitz, Comm. Math. Phys. (2018)

Hole Probability $P(r)$ in Disordered Many-Particle Configurations



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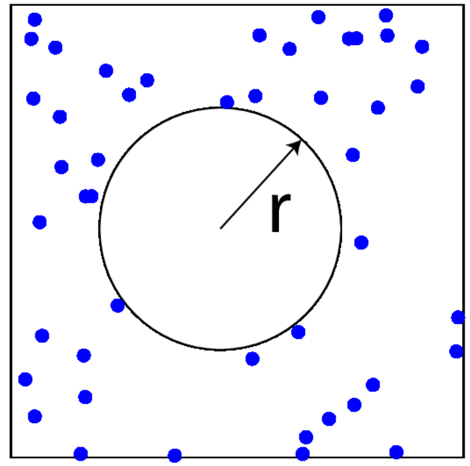

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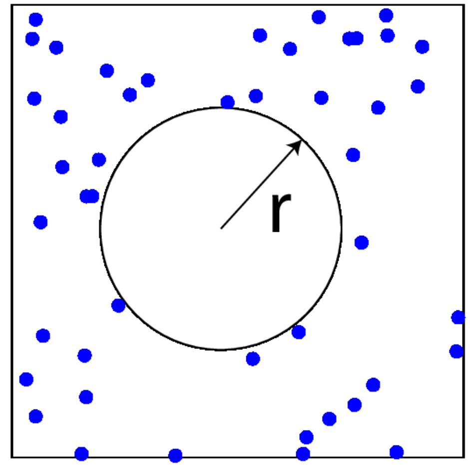

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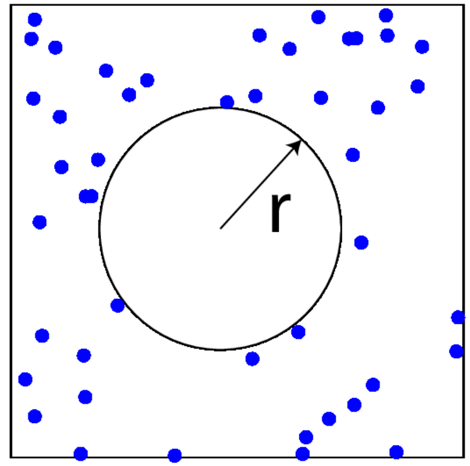

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- What about disordered hyperuniform systems? We know **not all disordered hyperuniform systems prohibit arbitrarily large holes** (e.g., $P(r) = \exp[-\kappa(d)r^{d+1}]$ for fermionic gases.).

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:

$$R_{max} \leq \frac{(d+1)\pi}{2K}$$

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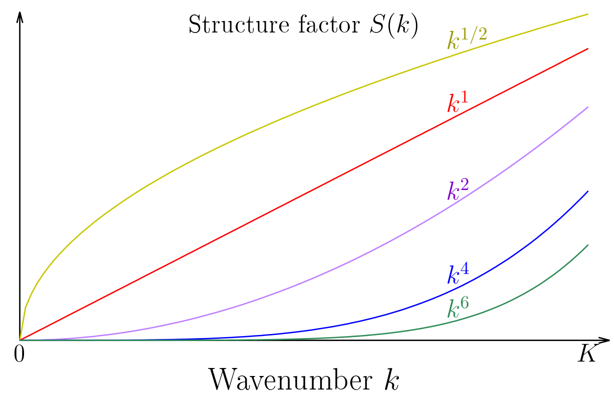
- Disordered stealthy materials with the largest value of χ lead to the best optical, transport and mechanical properties.

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)**

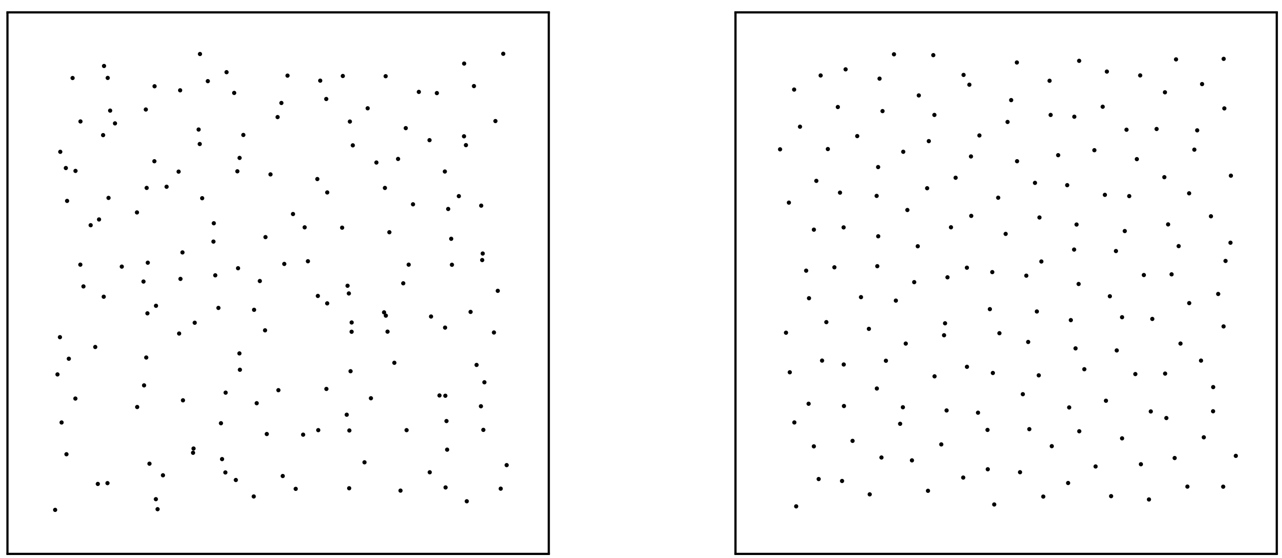
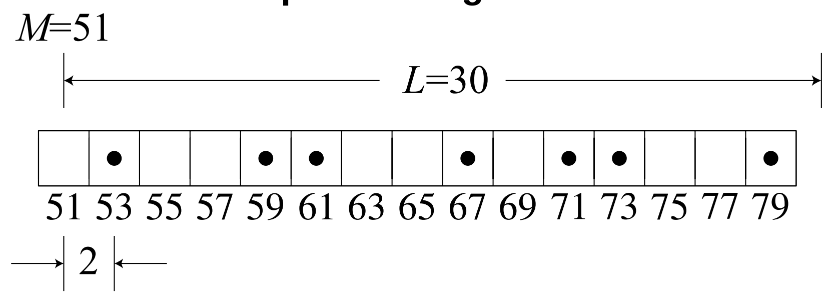


Figure 1: One of them is for $S(k) \sim k^6$ and other for $S(k) \sim k$.

Primes Teach Us About an Exotic Hyperuniform State

Zhang, Martelli and Torquato, J. Phys. A: Math. Theory (2018)

- 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!

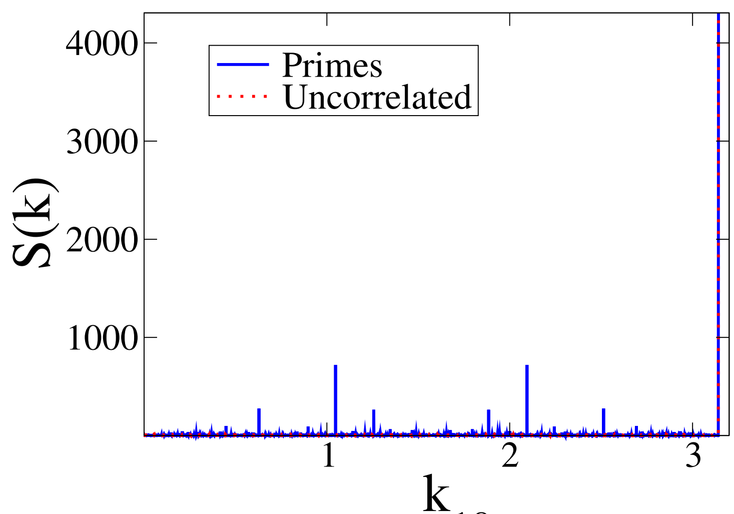


Figure 2: $S(k)$ for prime numbers for $M = 10^{10} + 1$ and $L = 10^5$ contains many peaks of various heights, creating a type of self-similarity.

Theoretical Treatment

Torquato, Zhang & de Courcy-Ireland, J. Stat. Mech. (2018); J. Phys A (2019)

- 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.,

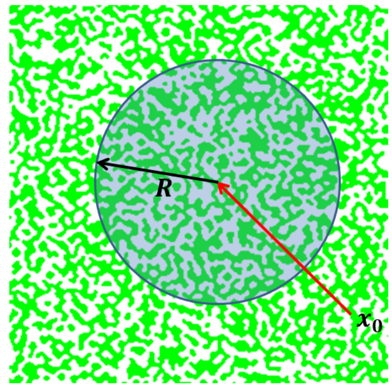
$$\lim_{M\to\infty} \frac{S(k)}{2\pi\rho} = \sum_{n}^{\flat} \sum_{m}^{\times} \frac{1}{\phi(n)^2} \delta\left(k - \frac{m\pi}{n}\right), \tag{-7}$$

  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 π, 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.

Hyperuniformity of Disordered Two-Phase Materials

- **Hyperuniformity concept was generalized to the case of heterogeneous materials: phase volume fraction fluctuates within a spherical window of radius R (Zachary and Torquato, J. Stat. Mech. 2009).**



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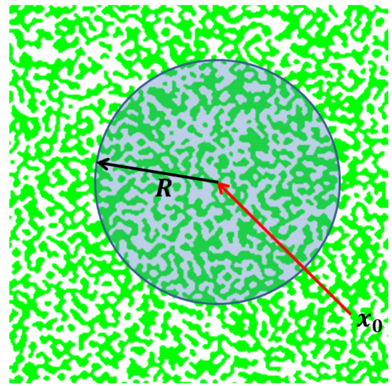

- **For typical disordered media, volume-fraction variance $\sigma_V^2(R)$ for large R goes to zero like R^{-d}.**
- **For hyperuniform disordered two-phase media, $\sigma_V^2(R)$ goes to zero faster than R^{-d}, equivalent to following condition on spectral density $\tilde{\chi}_V(\mathbf{k})$:**

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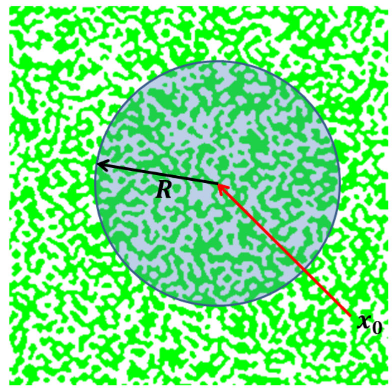

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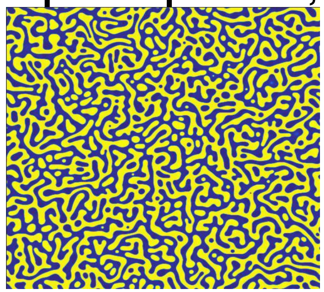
- **Interfacial-area fluctuations play an important role in static and surface-area evolving structures. Here we define $\sigma_S^2(R)$ and hyperuniformity condition is (Torquato, PRE, 2016)**

$$\lim_{|\mathbf{k}|\to 0} \tilde{\chi}_S(\mathbf{k}) = 0.$$

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.
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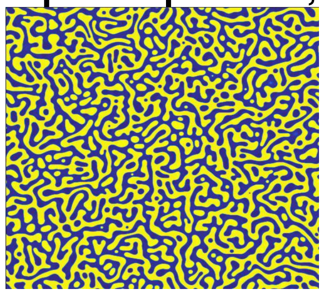

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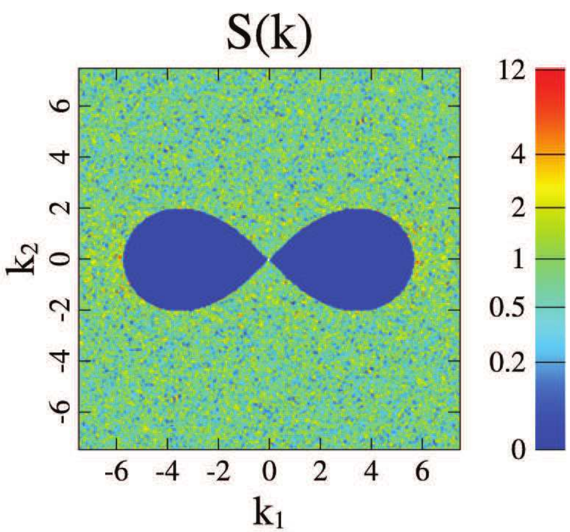

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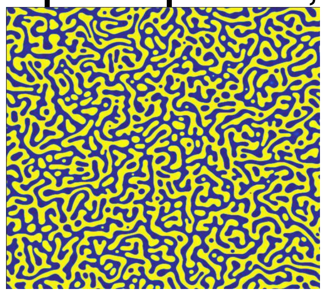

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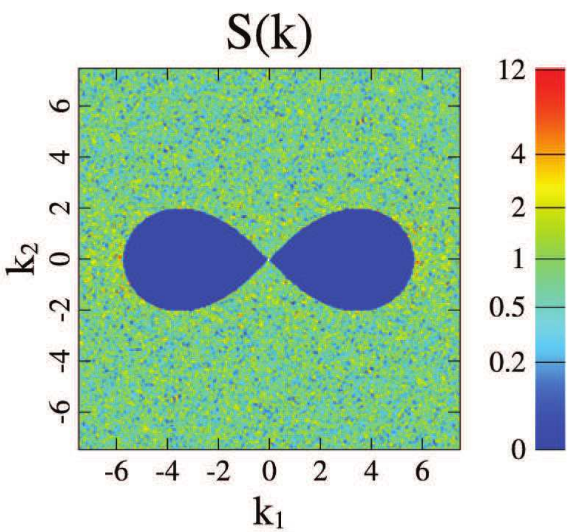

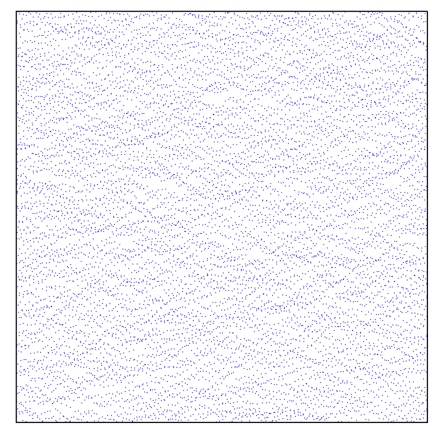

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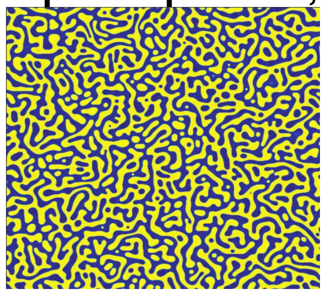

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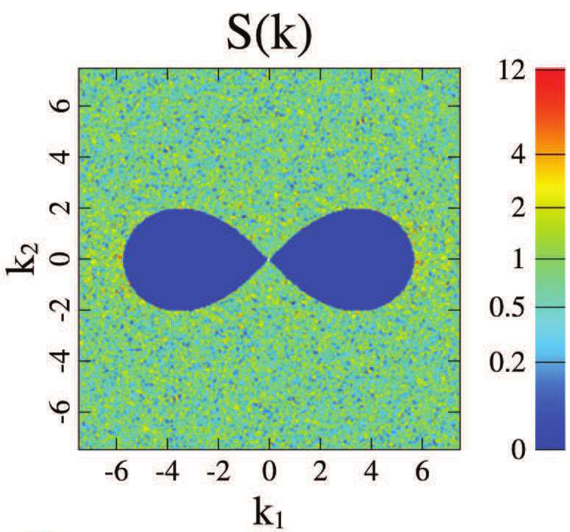

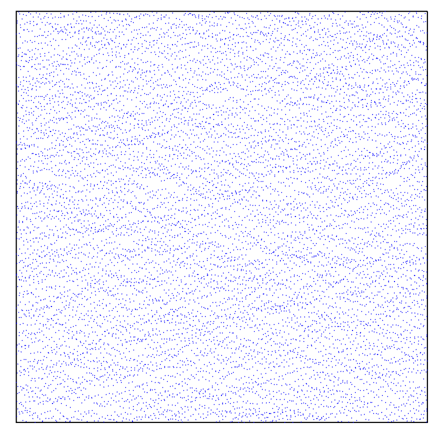

- Treatment of spin systems, both classical [Chertkov et al., PRB (2016)] and quantum-mechanical [Crowley, Laumann & Gopalakrishnan, PRB (2019)]

Challenge: Creation of Very Large Hyperuniform Samples Across Length Sca

- Numerical/experimental challenge to generate very large samples that are hyperuniform with high fidelity across length scales, e.g., from centimeters down to nanometers.

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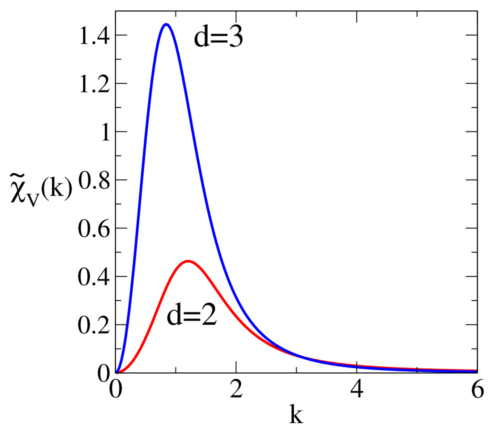
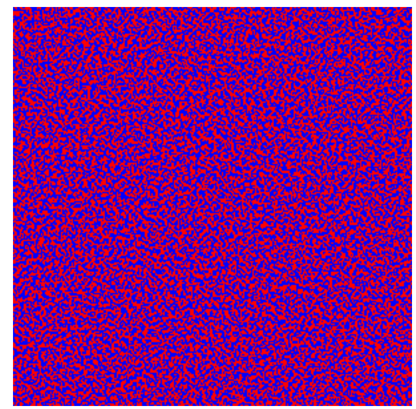
Designing Disordered Hyperuniform Composites

Chen and Torquato, Acta Materialia (2018)

- Can design disordered hyperuniform composites with targeted spectral densities $\tilde{\chi}_V(\mathbf{k})$.
- For example, consider following hyperuniform functional forms in 2D and 3D:





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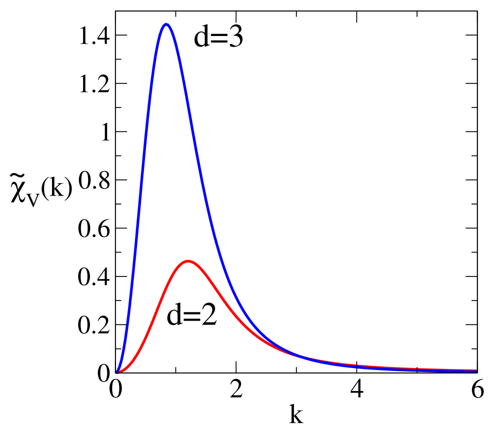

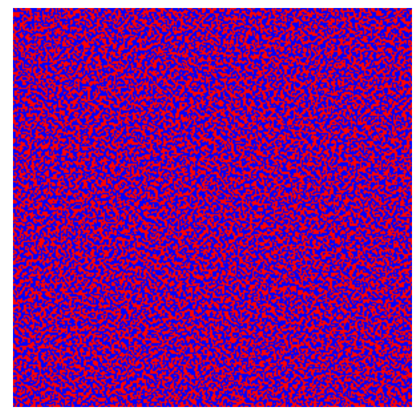

- Our **hyperuniform designs** can be readily **fabricated** using modern **photolithographic and 3D printing technologies** (100 micron resolution).

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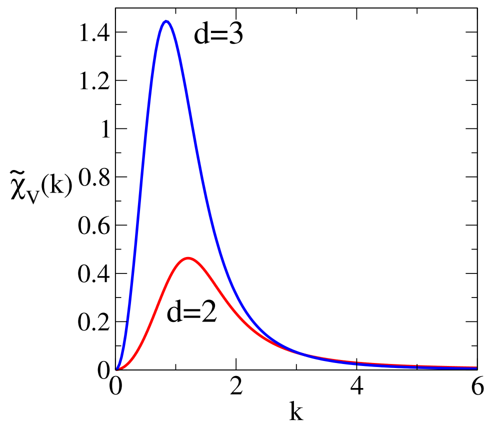
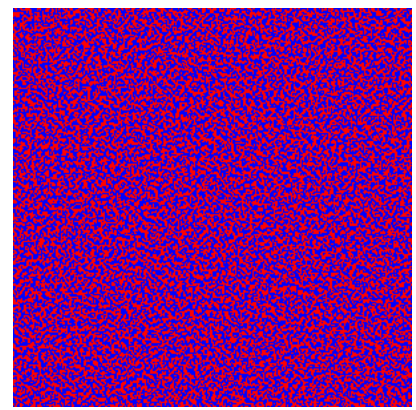




- Our hyperuniform designs can be readily fabricated using modern photolithographic and 3D printing technologies (100 micron resolution).

Charged Colloids

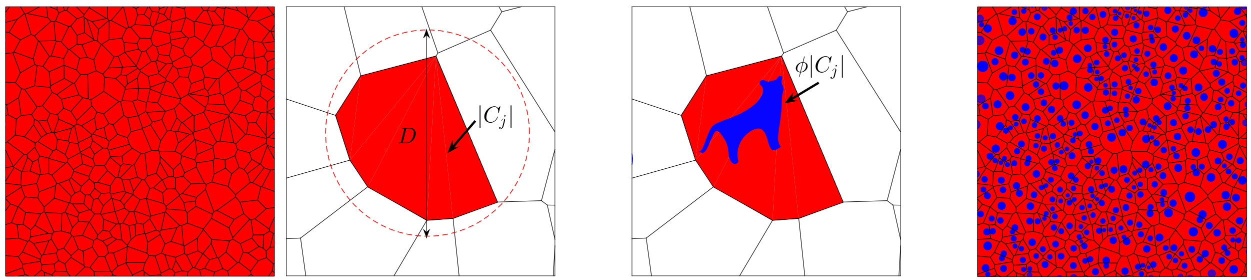
Chen, Lomba & Torquato, PCCP (2018)

- Particle or colloidal systems in equilibrium require long-range interactions.
- We carried out computer simulations to model charged colloids and tune temperature and screening length to manipulate the spectral density at small k: Chen, Lomba & Torquato, PCCP

Tessellation-Based Procedure to Create Very Large Hyperuniform Packings

Kim and Torquato, Acta Mater. (2019)

- Introduced a construction procedure that ensures perfect hyperuniformity for very large systems.
- 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 proved that this results in packings of particles with a size distribution that are guaranteed to be perfectly hyperuniform in the infinite-sample-size limit.
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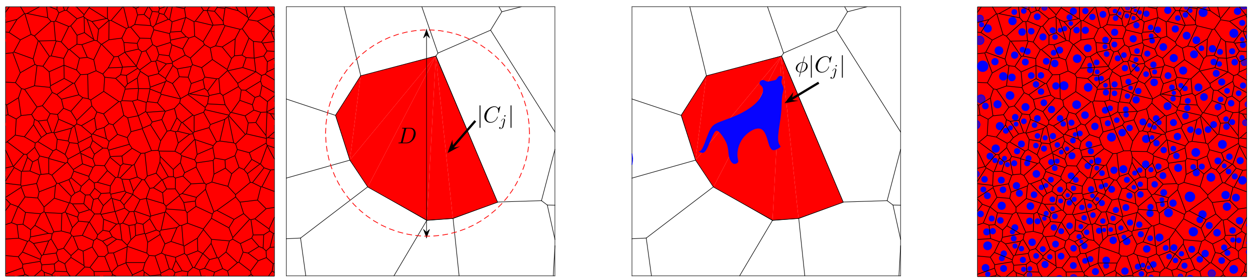
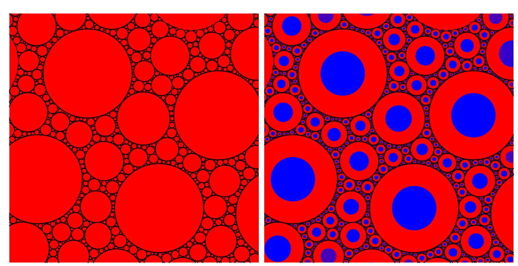


- We proved that this results in packings of particles with a size distribution that are **guaranteed** to be **perfectly hyperuniform in the infinite-sample-size limit**.
- Converts a **very large nonhyperuniform** disordered packing into a **hyperuniform** one!
- Also established hyperuniformity of the famous **Hashin-Shtrikman multiscale packings**, which possess **optimal transport and elastic properties**.



- Again, these **hyperuniform designs** can be readily **fabricated** using modern **photolithographic and 3D printing technologies**.

Optimized Large Hyperuniform Binary Colloids via Dipolar Interactions

Ma, Lomba and Torquato, Physical Review Letters (2020)

- The creation of disordered hyperuniform materials with extraordinary optical properties requires a capacity to synthesize large samples that are effectively hyperuniform down to the nanoscale.
- Proposed a feasible fabrication protocol using binary mixtures of paramagnetic colloidal particles confined in a 2D plane.
- The strong and long-ranged dipolar interaction induced by a tunable magnetic field is free from screening effects that attenuates long-ranged electrostatic interactions in charged colloidal systems.

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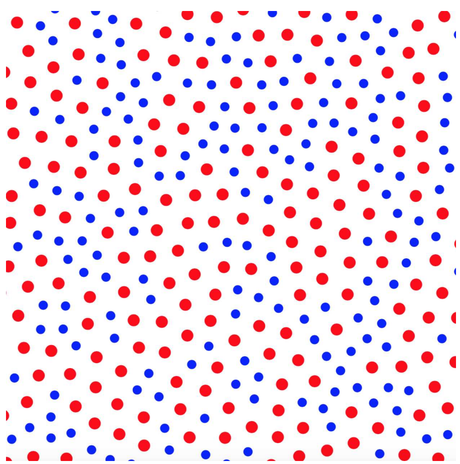
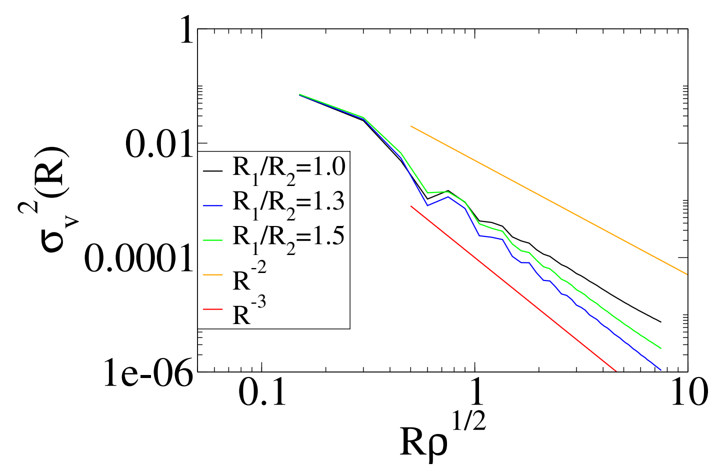
- We determined a family of optimal size ratios that makes the two-phase system effectively hyperuniform.





- Our methodology paves the way to self-assemble large disordered hyperuniform materials that function in the ultraviolet, visible and infrared regime and hence may accelerate the discovery of novel photonic materials.

eyond Number Variance: Higher-Order Cumulants and Probability Distributio

Torquato, Kim and Klatt, arXiv:2012.02358 (2020)

- We analyze the skewness $\gamma_1(R)$, excess kurtosis $\gamma_2(R)$ and the corresponding probability distribution function $P[N(R)]$ of a large family of models across the first three space dimensions, including both hyperuniform and nonhyperuniform models.
- We derive explicit integral expressions for $\gamma_1(R)$ and $\gamma_2(R)$ involving up to three- and four-body correlation functions, respectively.
- We also derive rigorous bounds on $\gamma_1(R)$, $\gamma_2(R)$ and $P[N(R)]$.
- The majority of the models obey a central limit theorem (CLT).
- Among all models, the convergence to a central limit theorem (CLT) is generally fastest for the disordered hyperuniform processes such that $\gamma_1(R) \sim l_2(R) \sim R^{-(d+1)/2}$ and $\gamma_2(R) \sim R^{-(d+1)}$ for large R.
- The convergence to a CLT is slower for standard nonhyperuniform models and slowest for the "antihyperuniform" model studied here.
- We prove that 1D hyperuniform systems of class I or any d-dimensional lattice cannot obey a CLT.

CONCLUSIONS

- 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.

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<u>Collaborators</u>

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