Disordered Hyperuniform Materials and Their Novel Optical Properties

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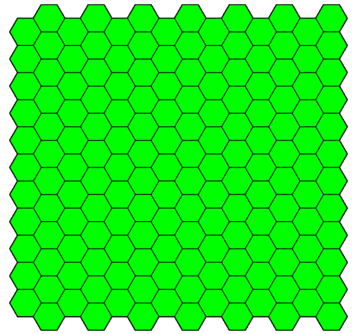
Review article: **S. Torquato, “Hyperuniform States of Matter,” Physics Reports, 745, 1 (2018).**

OUTLINE

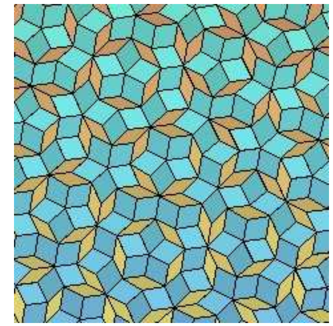
- 1. Brief Review of Hyperuniformity
- 2. Multihyperuniformity
- 3. Stealthy Hyperuniformity and Order Across Length Scales
- 4. Novel Optical Properties

Long-Range Order: Crystals and Quasicrystals

- **Multitude** of distinguishable states of matter that break **continuous translational/rotational symmetries of a liquid** differently from a solid **crystal**.



Crystal



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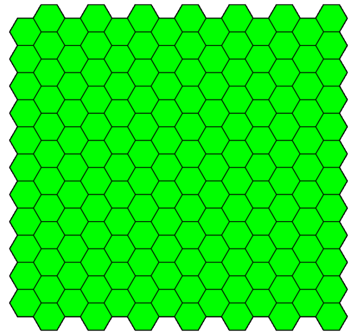

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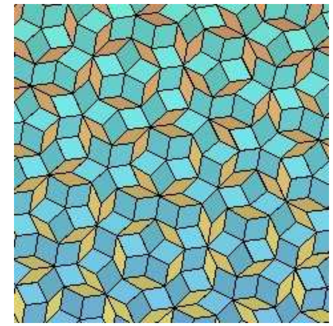
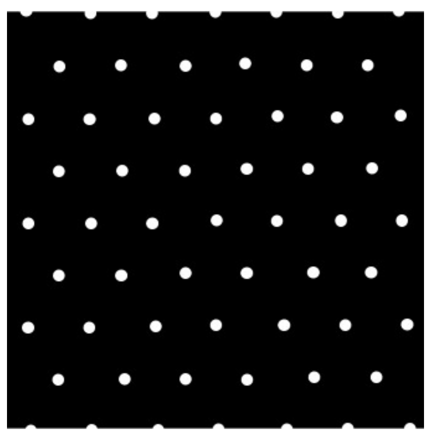
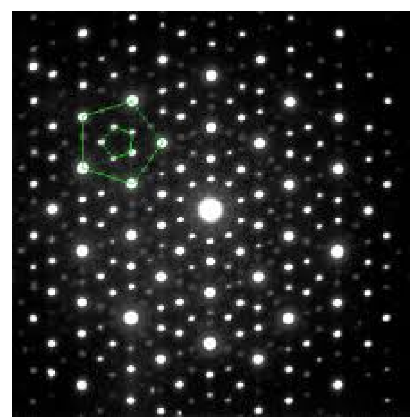


Quasicrystal

- Crystals have both **long-range periodic translational and orientational order**.
- **Quasicrystals** taught us how to generalize the concept of **long-range order**. They possess **long-range quasiperiodic translational** order and long-range **orientational order with prohibited crystallographic symmetries**. Shechtman et al. PRL (1984); Levine & Steinhardt, PRL (1984)





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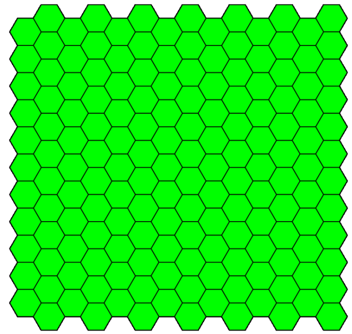

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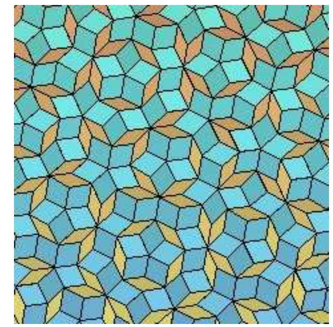

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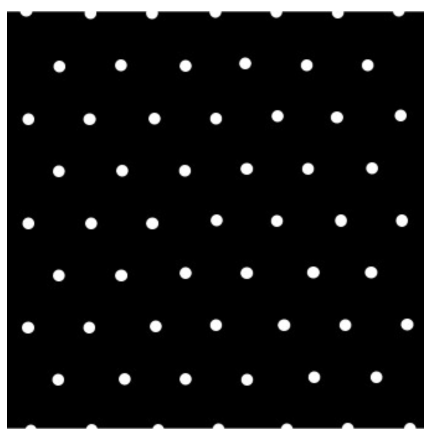

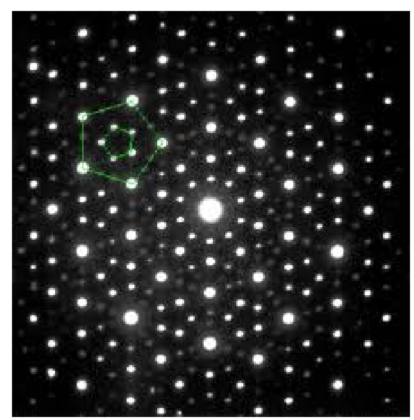

- **Hyperuniformity generalizes** these established notions of long-range order.
- Hyperuniformity also **forces us to re-think what we mean by "disorder."**

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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- **Disordered hyperuniform** many-particle systems can be regarded to be **new ideal states of matter** in that they

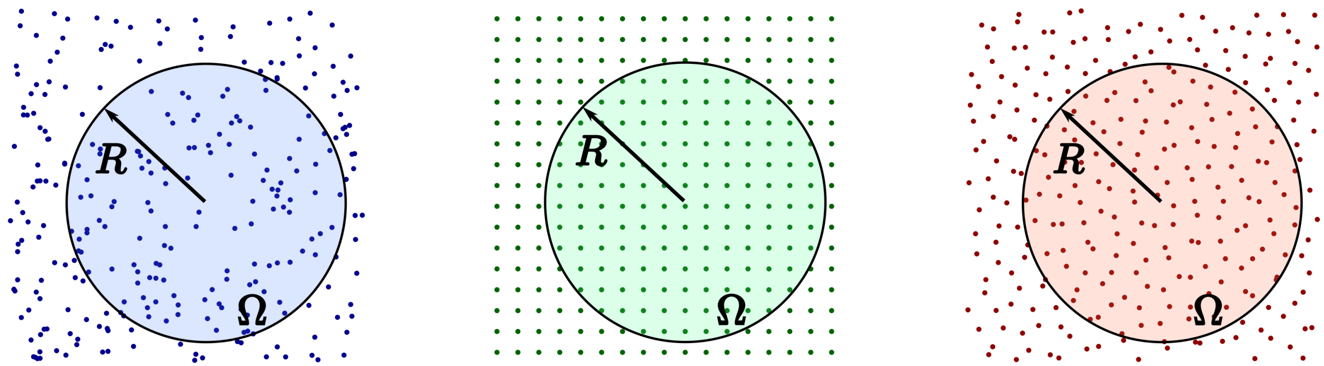
  1. *behave more like **crystals or quasicrystals** in the way they **suppress large-scale density fluctuations**, and yet are also like **liquids and glasses**, since they are statistically **isotropic structures with no Bragg peaks***;
  2. *can exist as both as **equilibrium** and **nonequilibrium** phases*;
  3. *come in **quantum-mechanical** and **classical** varieties*;
  4. *and, are endowed with **unique bulk physical properties***.

Understanding such disordered states of matter requires new theoretical tools and present experimental challenges.

Large-Scale Density Fluctuations and Hyperuniformity

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

- **Points in $\mathbb{R}^d$ 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.**



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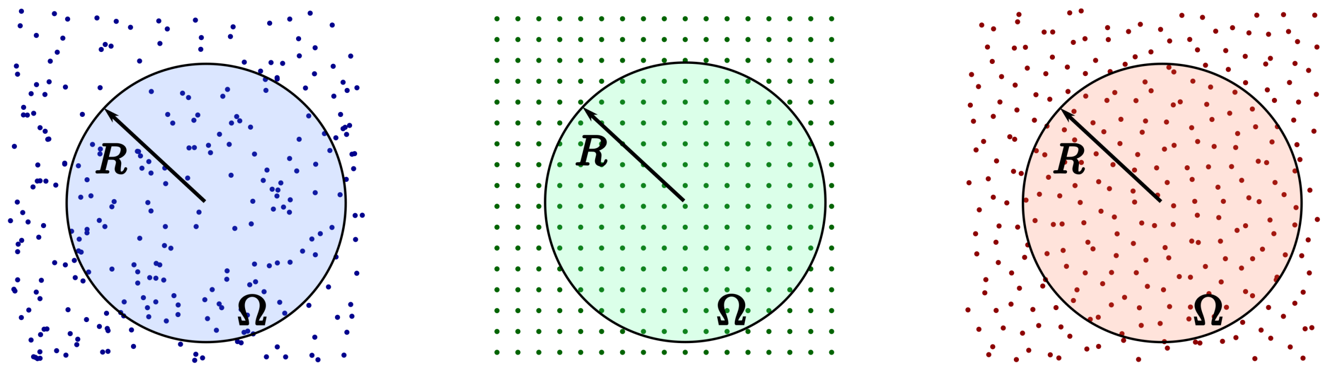

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- **We call point patterns whose variance $\sigma^2(R)$ grows more slowly than R^d (window volume) hyperuniform, implying that structure factor vanishes in infinite-wavelength limit, i.e.,**

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

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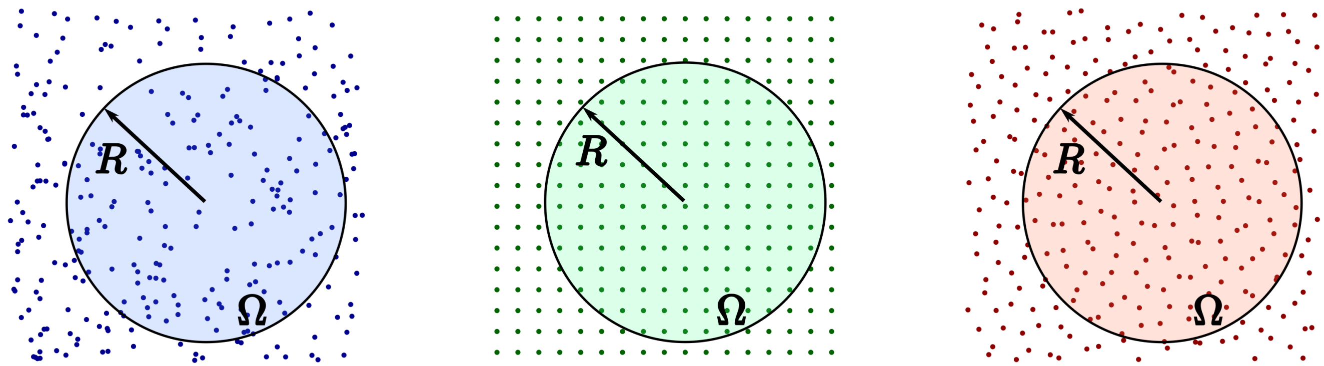

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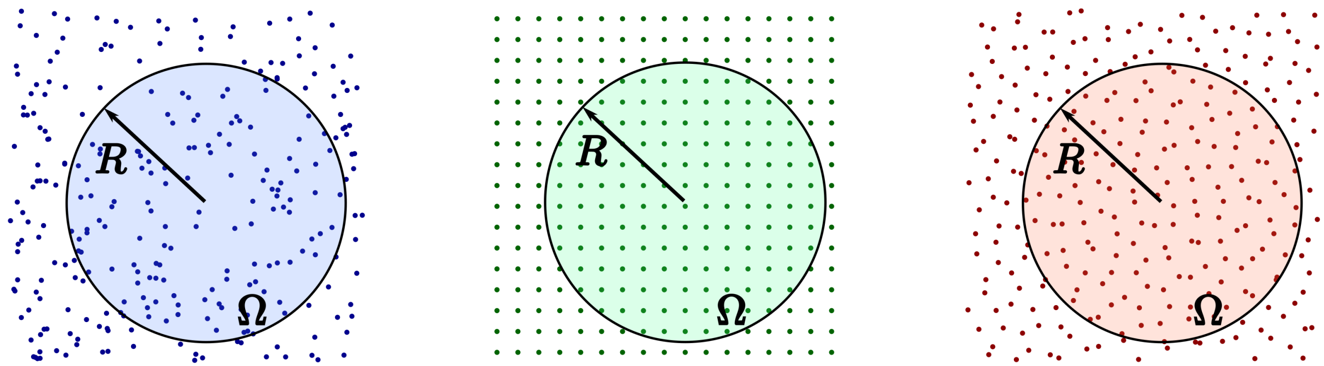

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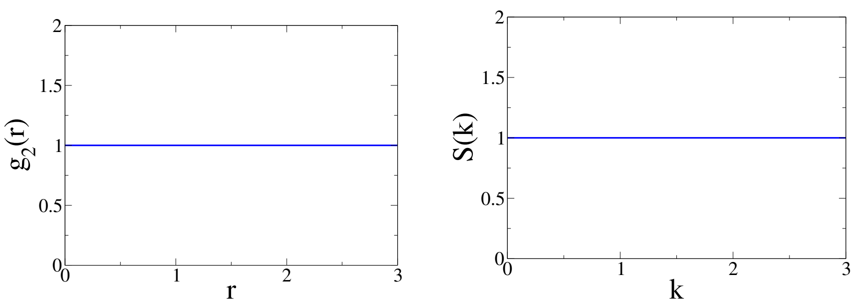
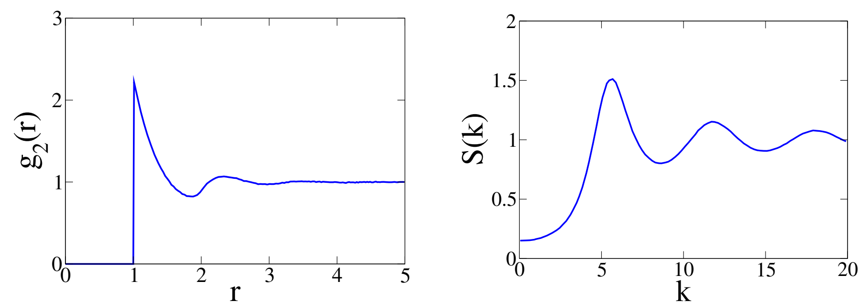
Pair Statistics in Direct and Fourier Spaces

- 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 $\tilde{h}(k)$.

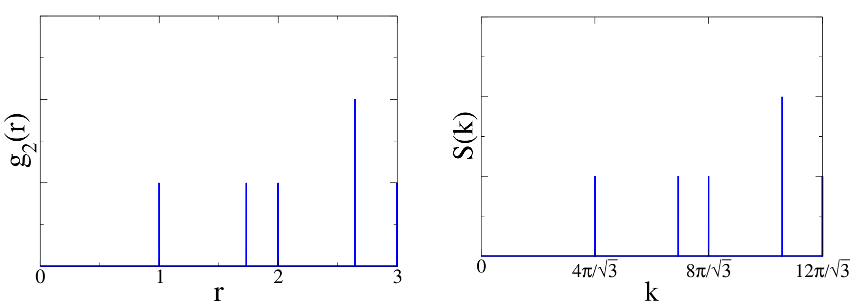
Poisson Distribution (Ideal Gas)



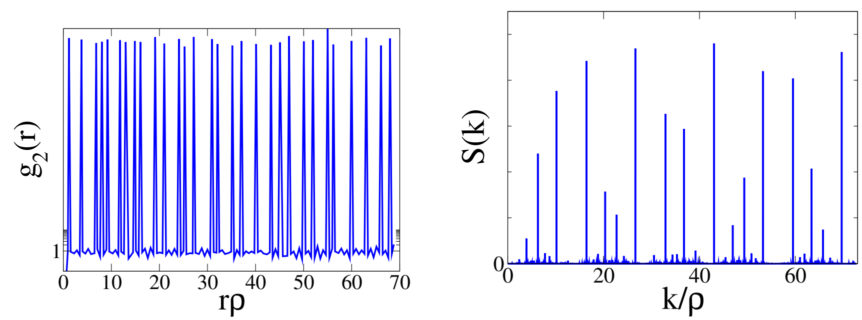
Liquid



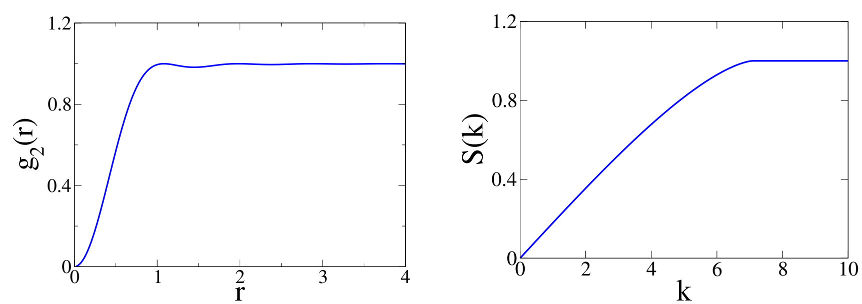
Lattice



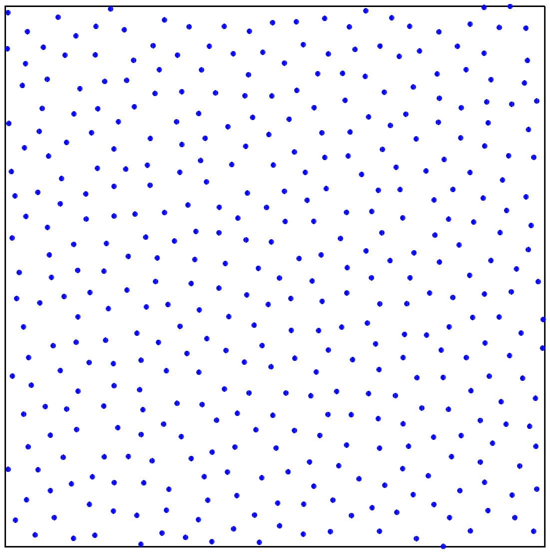
Quasicrystal

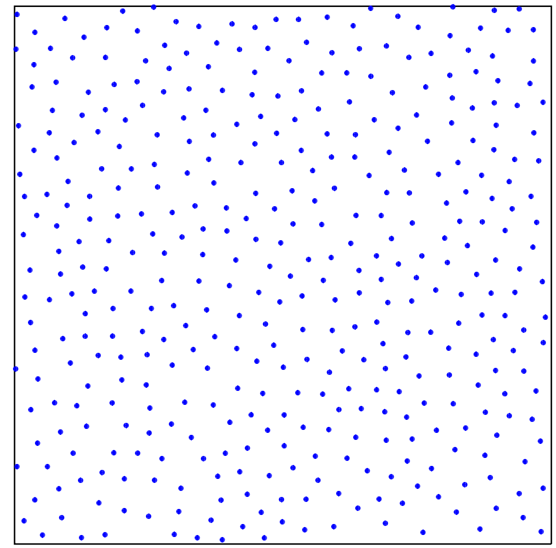


Disordered Hyperuniform System



Hidden Order on Large Length Scales





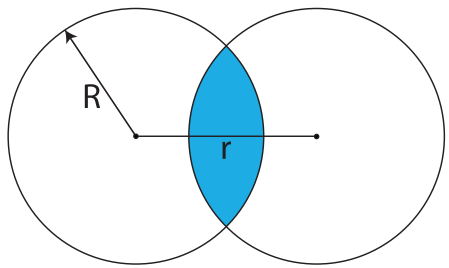
Which is the hyperuniform pattern?

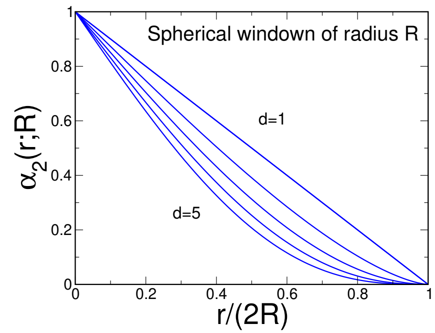
ENSEMBLE-AVERAGE FORMULATION

- **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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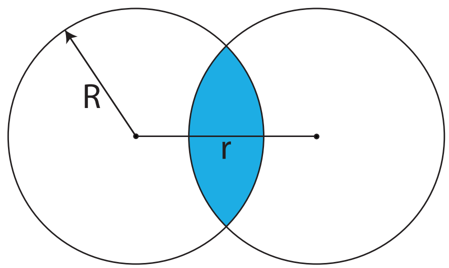
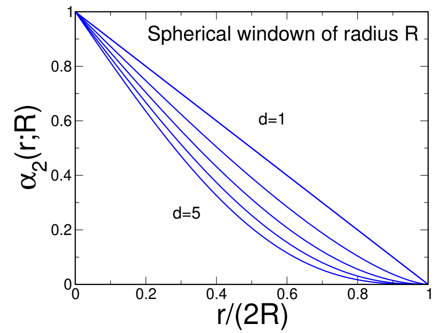




- **For a certain class of systems and large R, we can show**

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

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**: $A = 0, B > 0 \implies$ **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 B/A - Larger (smaller) is B/A, the larger (smaller) is the hyperuniformity scaling regime for $\sigma^2(R)$.**

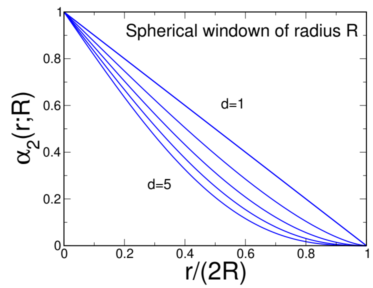
- **We'll see that you can have other variance scalings between R^{d-1} and R^d.**

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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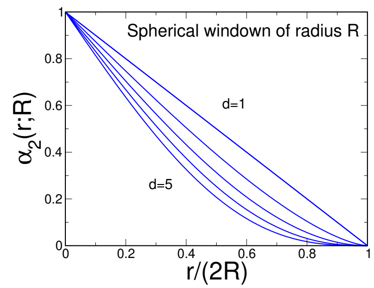

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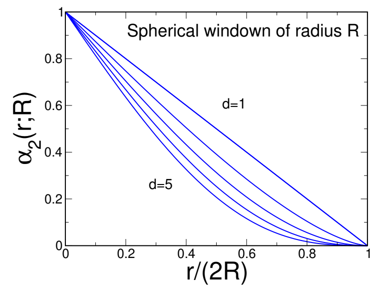
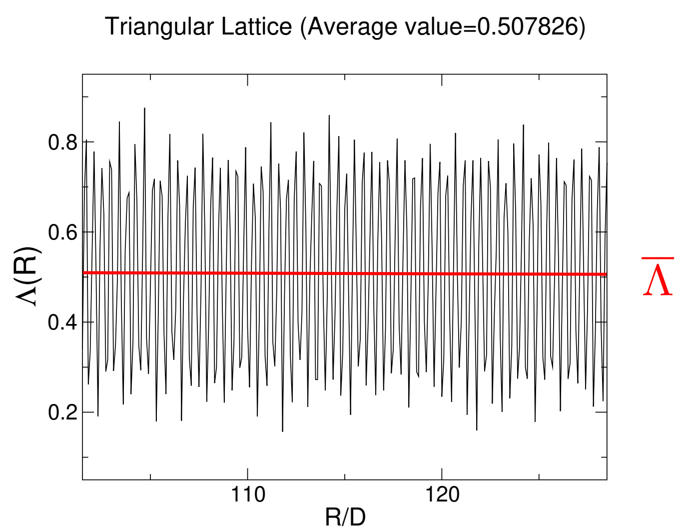


- **Finding global minimum of $\sigma^2(R)$ equivalent to finding ground state.**
- **For large R, in the special case of hyperuniform systems,**

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



Quantifying Suppression of Density Fluctuations at Large Scales: 1D

- **For any d, averaging fluctuating quantity $\Lambda(R)$ gives coefficient of interest:**

$$\overline{\Lambda} = \lim_{L\to\infty} \frac{1}{L}\int_0^L \Lambda(R)dR$$

Lower the surface-area coefficient $\overline{\Lambda}$, greater the suppression of large-scale fluctuations in a hyperuniform system.

- **The surface-area coefficient $\overline{\Lambda}$ for some crystal, quasicrystal and disordered 1D hyperuniform point patterns (Torquato & Stillinger 2003).**

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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 1D quasicrystals: Oğuz, 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** 2D hyperuniform point patterns (Torquato & Stillinger 2003).

2D Pattern	$\overline{\Lambda}$
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: 2D

- The surface-area coefficient $\overline{\Lambda}$ for some crystal, quasicrystal and disordered 2D hyperuniform point patterns (Torquato & Stillinger 2003).

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*Zachary & Torquato (2009)

- More recent work on hyperuniformity of 2D quasicrystals: Lin, Steinhardt and Torquato (2017).

Quantifying Suppression of Density Fluctuations at Large Scales: 3D

- The surface-area coefficient $\overline{\Lambda}$ for some crystal and disordered 3D hyperuniform point patterns (Torquato & Stillinger 2003).

Pattern	$\overline{\Lambda}$
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).
- Minimizers of $\overline{\Lambda}$ and Epstein zeta function are directly related.

General Hyperuniform Scaling Behaviors

- Consider systems characterized by a power-law structure factor

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

- For hyperuniform systems, $\alpha > 0$, can prove number variance $\sigma^2(R)$ has following large-R scalings (Zachary and Torquato, 2011):

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

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- Class I: $\sigma^2(R) \sim R^{d-1}$: Crystals, quasicrystals, stealthy disordered ground states, one-component plasmas, Laughlin's "incompressible" quantum fluid, g_2-invariant disordered point processes, vortex structures in type-II superconductors.
- Class II: $\sigma^2(R) \sim R^{d-1}\ln(R)$: Quasicrystals, classical disordered ground states, nontrivial Riemann zeros, eigenvalues of random matrices, fermionic systems, superfluid helium, maximally random jammed packings, prime numbers.
- Class III: $\sigma^2(R) \sim R^{d-\alpha}$ $(0 < \alpha < 1)$: Classical disordered ground states, nonequilibrium phase transitions/random organization models.

General Hyperuniform Scaling Behaviors

- Consider systems characterized by a **power-law structure factor**

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

- For **hyperuniform systems**, $\alpha > 0$, can prove number variance $\sigma^2(R)$ has following large-R scalings (**Zachary and Torquato, 2011**):

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

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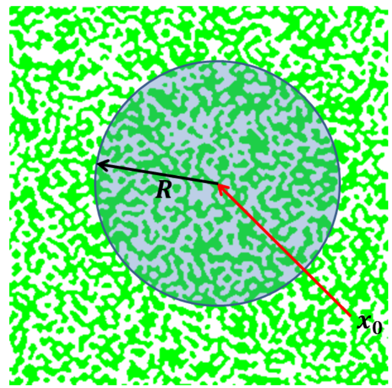
General Nonhyperuniform Scaling Behaviors

$$\sigma^2(R) \sim \begin{cases} R^{d}, & \alpha = 0 \quad (\textbf{typical nonhyperuniform}) \\ R^{d-\alpha}, & -d < \alpha < 0 \quad (\textbf{anti-hyperuniform}). \end{cases}$$

- Thus, can classify **all translationally invariant** states of matter according to their **large-scale density fluctuations**.

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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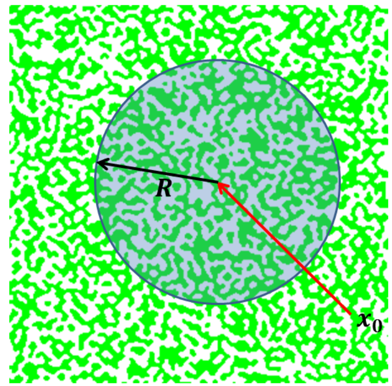

- For typical disordered media, volume-fraction variance $\sigma_V^2(R)$ for large R goes to zero like R^{-d}.
- For hyperuniform disordered two-phase media, $\sigma_V^2(R)$ goes to zero faster than R^{-d}, equivalent to following condition on spectral density $\tilde{\chi}_V(\mathbf{k})$:

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

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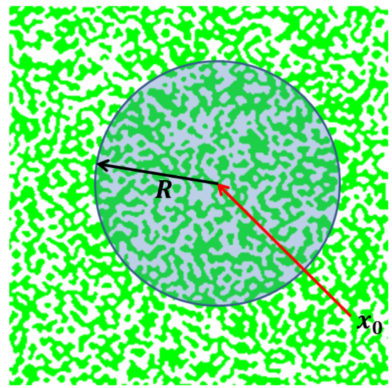

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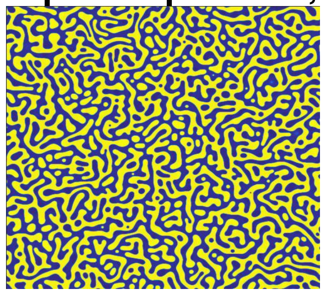
- Interfacial-area fluctuations play an important role in static and surface-area evolving structures. Here we define $\sigma_S^2(R)$ and hyperuniformity condition is (Torquato, PRE, 2016)

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

Other Generalizations of Hyperuniformity

- Some generalizations (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/tensor fields: Random media (e.g., heat, current, electric, magnetic, velocity and stress fields), turbulence, etc.
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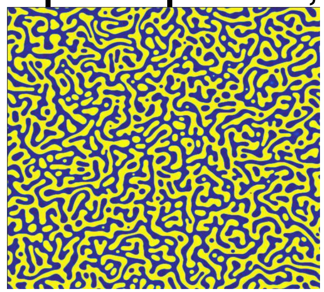

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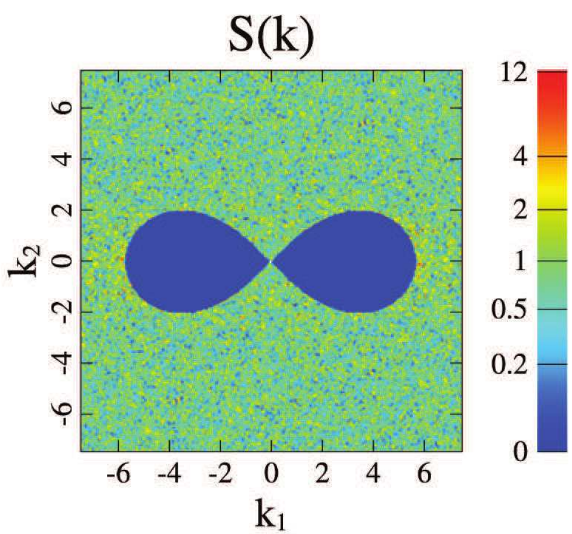

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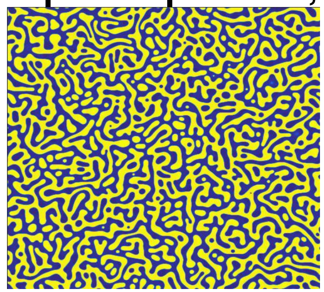

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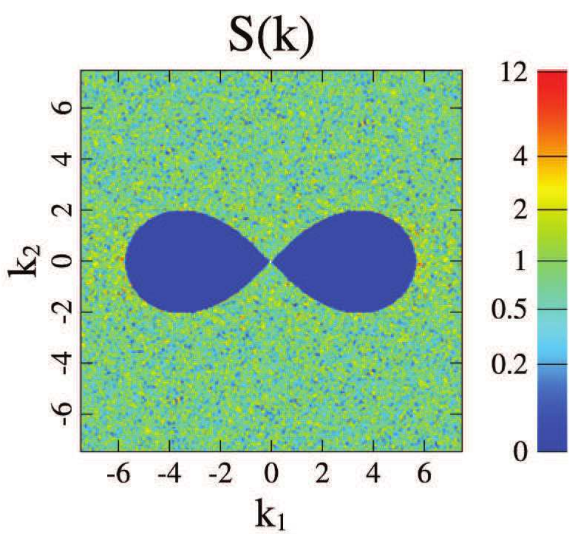

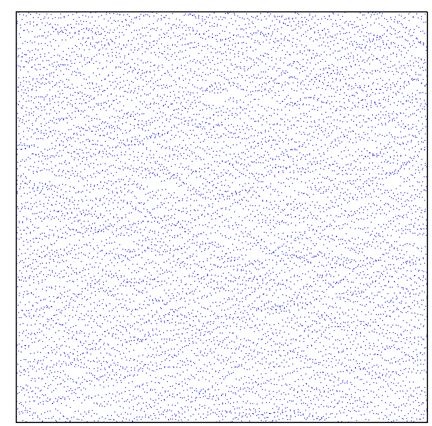

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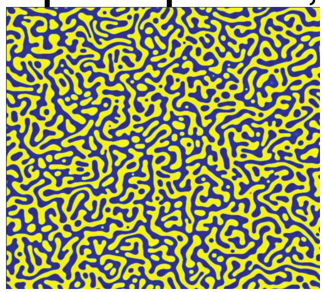

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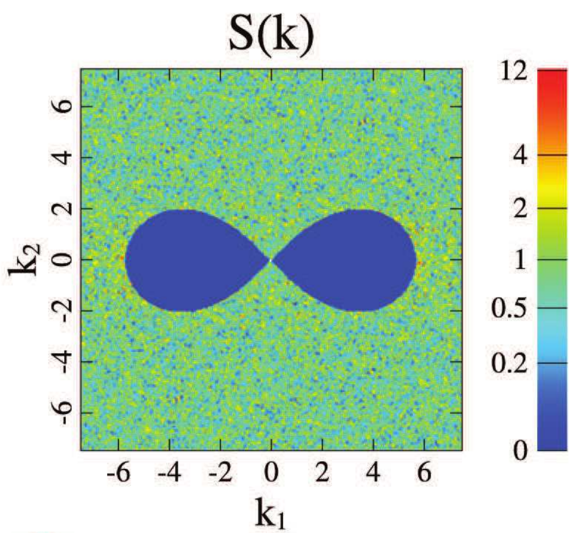

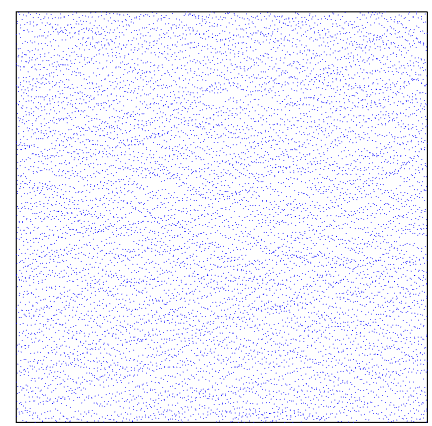

- Treatment of spin systems, both classical [Chertkov et al., PRB (2016)] and quantum-mechanical [Crowley, Laumann & Gopalakrishnan, PRB (2019)]

Examples of Disordered Hyperuniform Systems

Physical Examples

- **Disordered classical ground states**: Uche et al. PRE (2004); Batten et al., PRE (2015)
- **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)
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Nearly Hyperuniform Disordered Systems

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

Randomly Perturbed Crystals and Their Order/Disorder

Klatt, Kim and Torquato, PRE (2018)

- A common way to introduce disorder into an otherwise ordered system such as a perfect crystal or quasicrystal is to randomly perturb the particle positions of that system.
- The structure factor $S(\mathbf{k})$ for a uniformly randomized lattice (URL) is

$$S(\mathbf{k}) = 1 - |\tilde{f}(\mathbf{k})|^2 + |\tilde{f}(\mathbf{k})|^2 S_{\mathcal{L}}(\mathbf{k}),$$

  where $S_{\mathcal{L}}(\mathbf{k})$ - structure factor of unperturbed lattice $\mathcal{L}$ and $\tilde{f}$ is FT of displacement PDF f.
- For most a, Bragg peaks are present, which is far from disordered!
- Certain a make second term vanish, i.e., Bragg peaks are cloaked!, yielding $S(k) \sim k^2$ (class I).

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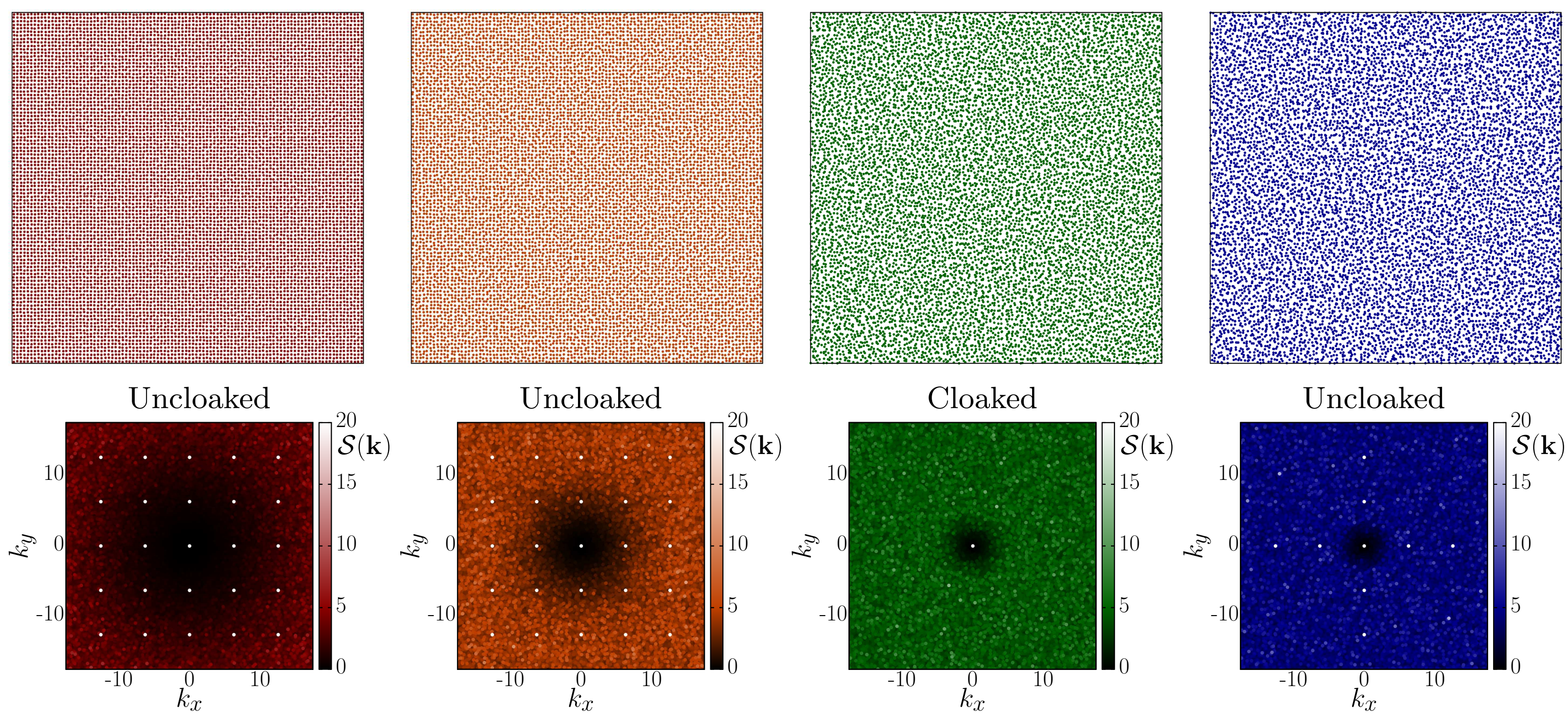

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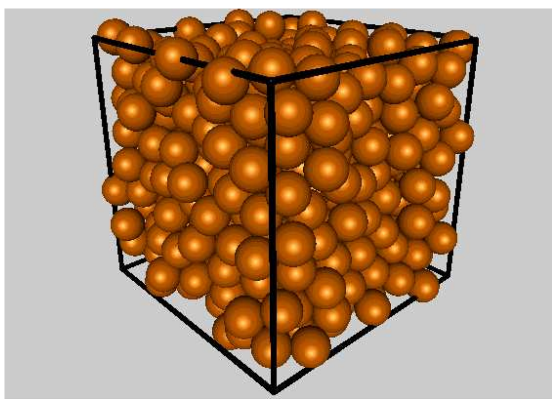
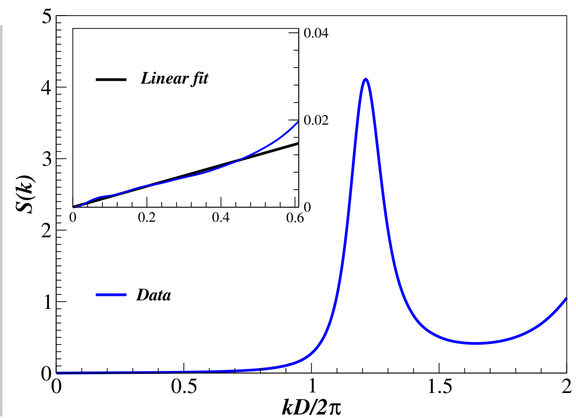
- **Such packings of identical spheres have been shown to be hyperuniform with quasi-long-range (QLR) pair correlations in which $h(r)$ decays as $-1/r^4$ (Donev, Stillinger & Torquato, PRL, 2005).**





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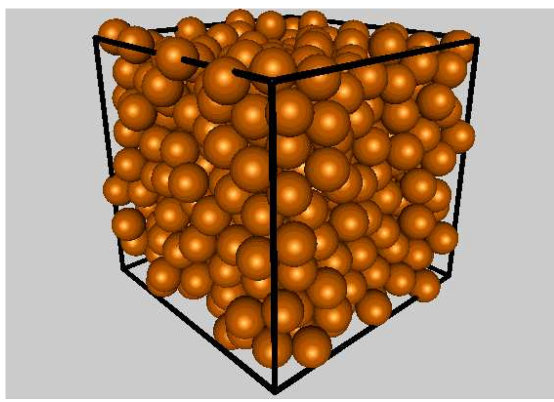
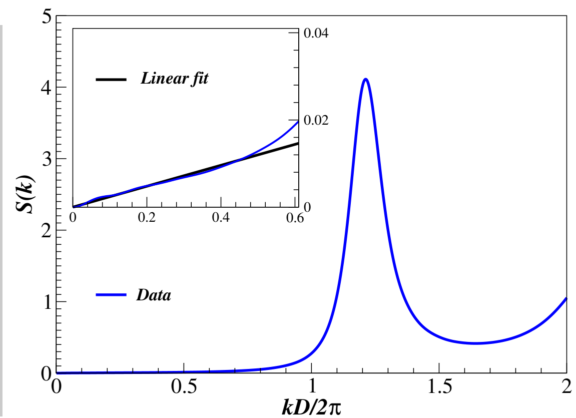




This is to be contrasted with the hard-sphere fluid with correlations that decay exponentially fast. Contradicts frozen-liquid picture of a glass.

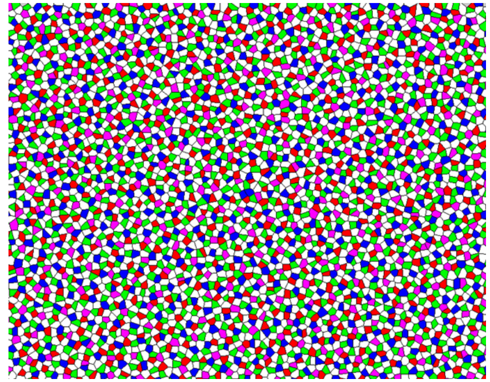
- **Apparently, hyperuniform QLR correlations with decay $-1/r^{d+1}$ are a universal feature of general MRJ packings in $\mathbb{R}^d$.**

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

Multihyperuniformity: In the Eye of a Chicken



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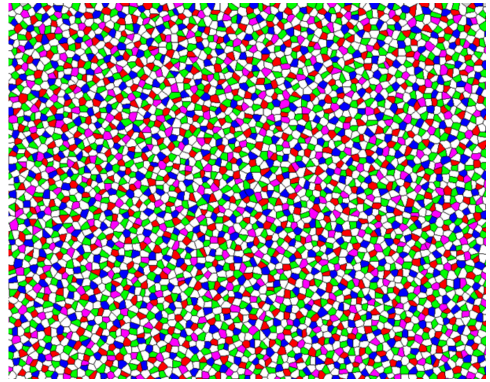

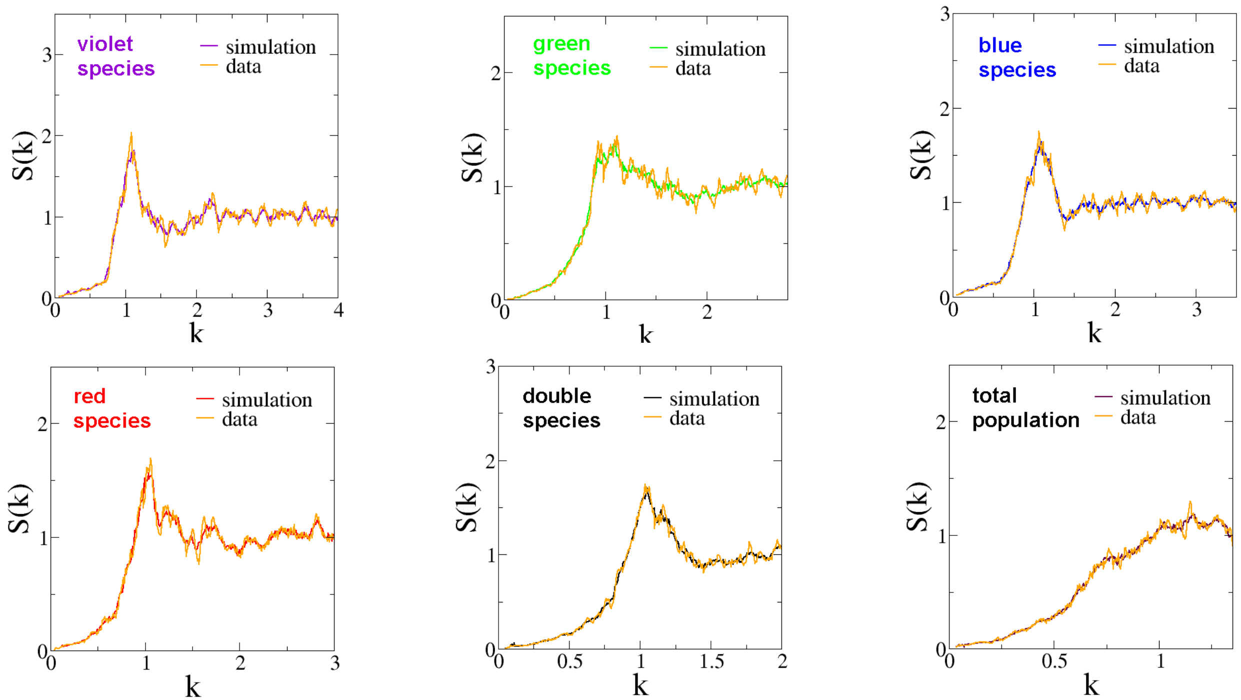

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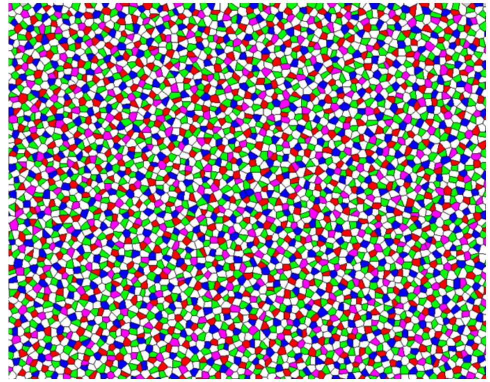

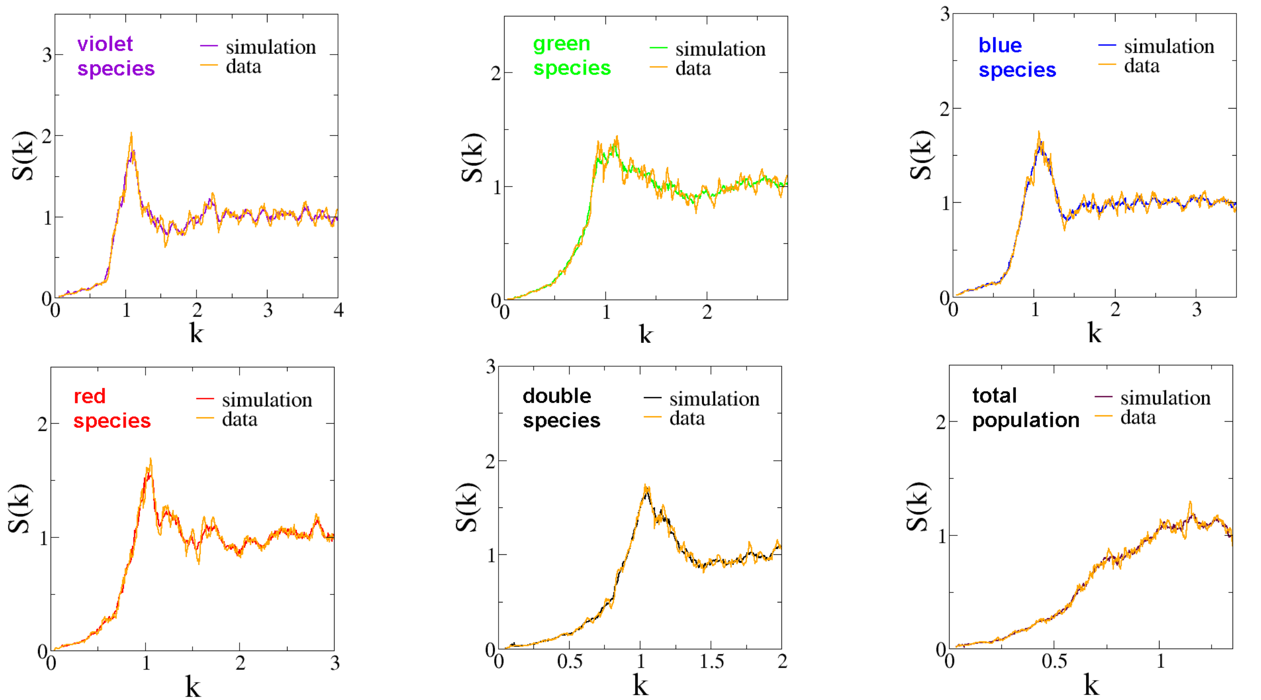

- Disordered mosaics of **both total population and individual cone types** are effectively **hyperuniform**, which had been **never** observed in any system before. We call this **multi-hyperuniformity** (**Jiao, Corbo & Torquato, PRE 2014**).
- Recently showed that **multihyperuniformity** can be **rigorously** achieved via **hard-disk plasmas** (**Lomba, Weis and Torquato, PRE 2018**).

Classical Disordered "Stealthy" Hyperuniform Ground States

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

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

- Consider N particles with configuration $\mathbf{r}^N$ in a flat torus $\mathbf{T}$ with a pair potential $v(\mathbf{r})$ that is **bounded** with Fourier transform $\tilde{v}(\mathbf{k})$. The **total potential energy** is

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

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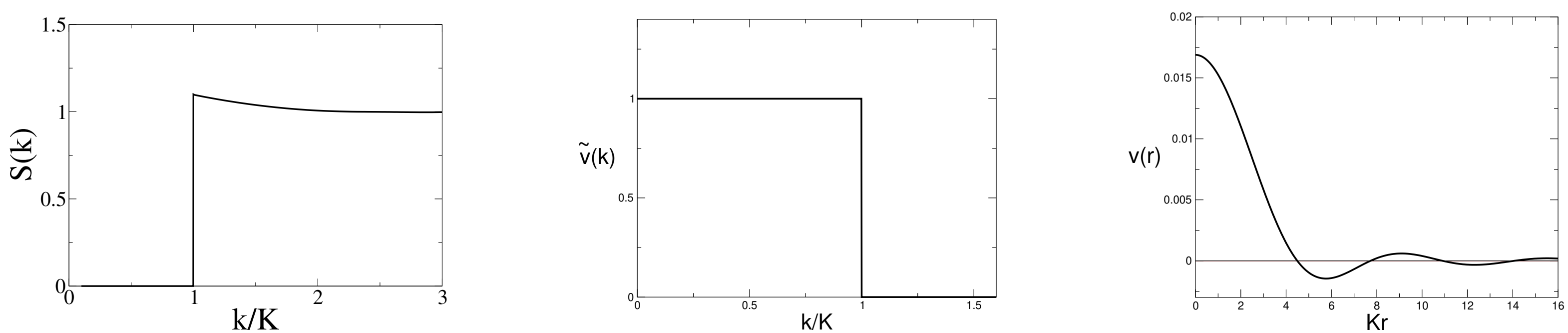

- These **exotic class I hyperuniform** ground states are called **"stealthy"** and when **disordered are highly degenerate** - classical analogs of **quantum spin liquids**.
- **Direct-space** potentials are **long-ranged**, reminiscent of **Friedel oscillations** of the electron density in a variety of systems.

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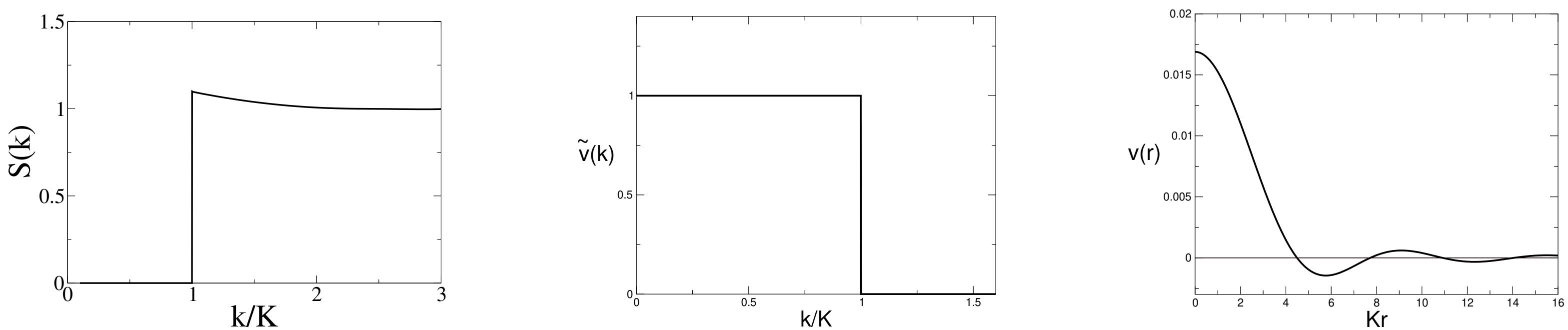

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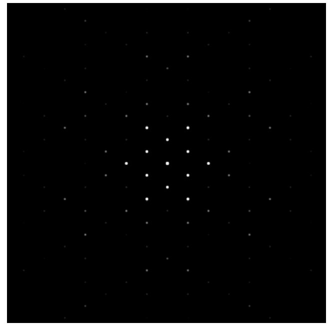
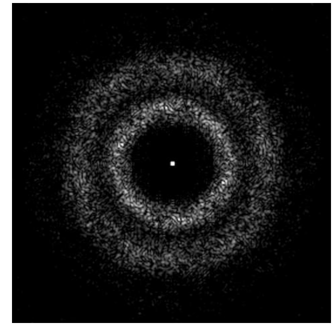
- Stealthy patterns can be **tuned** by varying size of the **"exclusion region"**, measured by parameter χ: ratio of # of **constrained degrees of freedom** to the total # of degrees of freedom, $d(N-1)$.

Disordered Stealthy Hyperuniform Ground States

- Stealthy systems have **hybrid crystal-liquid** nature.



Crystal



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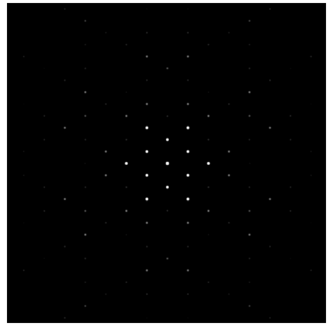

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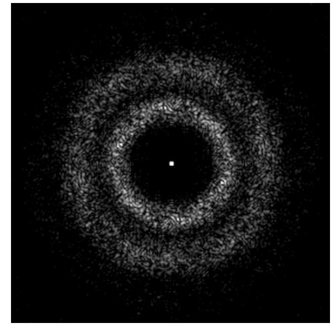
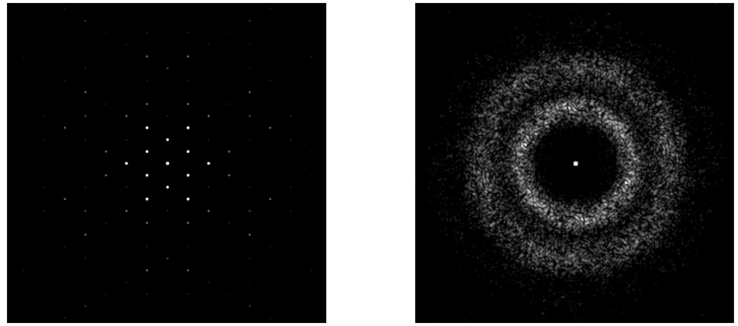


Stealthy

- A **statistical-mechanical theory** for stealthy ground states is nontrivial because **dimensionality of the configuration space decreases with** χ. Such a theory for the thermodynamics and structure has been proposed [Torquato, Zhang and Stillinger, PRX (2015)].

Disordered Stealthy Hyperuniform Ground States

- Stealthy systems have **hybrid crystal-liquid** nature.



Crystal Stealthy

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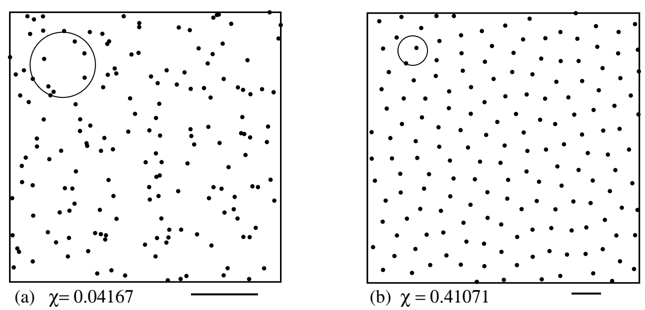
- For $0 \leq \chi < 0.5$, 2D and 3D ground states are **highly degenerate, disordered and isotropic**.

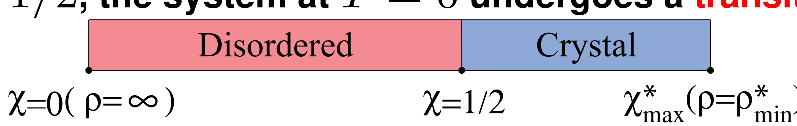


As χ increases, **short-range order increases**; see animations.

- We proposed new **metric** to quantify **order across length scales**:

$$\tau = \frac{1}{(2\pi)^d D^d} \int_{|\mathbf{k}| \leq K} [S(\mathbf{k}) - 1]^2 d\mathbf{k},$$

- As χ increases above $1/2$, the system at $T = 0$ undergoes a **transition to ordered phases**.



Disordered Stealthy Hyperuniform Ground States and Novel Material

Florescu, Torquato and Steinhardt, PNAS (2009)

- About 1.5 decades ago, it was believed that Bragg scattering was required to achieve cellular solids with complete photonic band gaps (PBGs).

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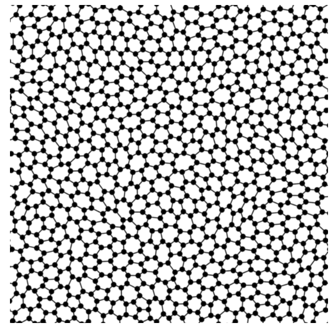
- Mapped disordered, isotropic "stealthy" ground-state configurations into disordered 2D dielectric trivalent networks via a Delaunnay centroidal tessellation.



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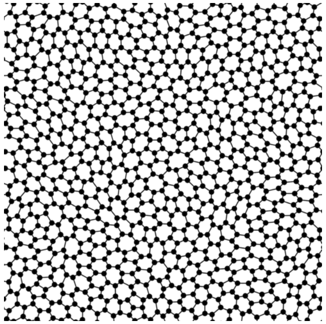
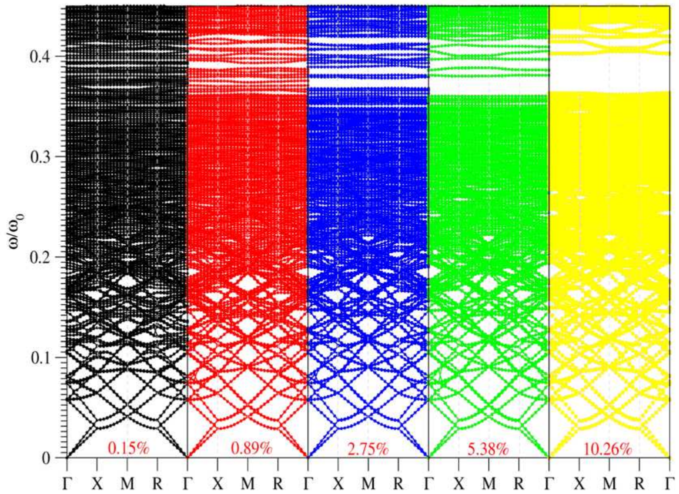


- This enabled us to computationally design photonic materials with **large complete** (both polarizations and all directions) **band gaps**.

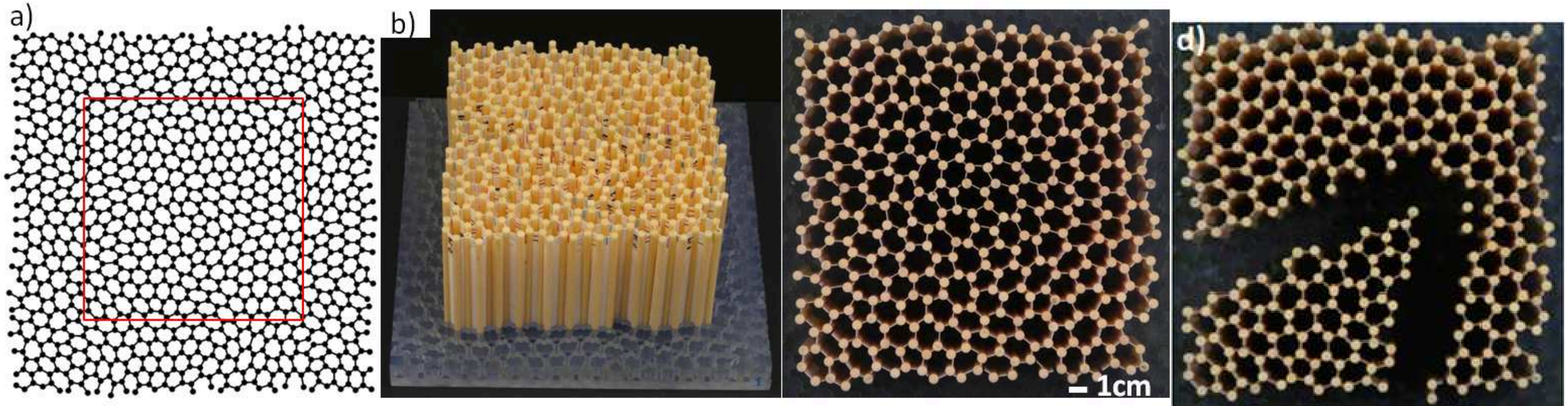


$$\chi = 0.1, \; 0.2, \; 0.3, \; 0.4, \; 0.5$$

Can now compute τ order metric $= 0.02, 0.04, 0.13, \; 0.42, \; 2.72$ (Note: $\tau \longrightarrow \infty$ for crystal)

Disordered Stealthy Hyperuniform Ground States and Novel Materials

- These network material designs have been fabricated and tested for microwave regime: Man, Florescu et. al.. PNAS (2013).



Because band gaps are isotropic, such photonic materials offer advantages over photonic crystals (e.g., free-form waveguides).

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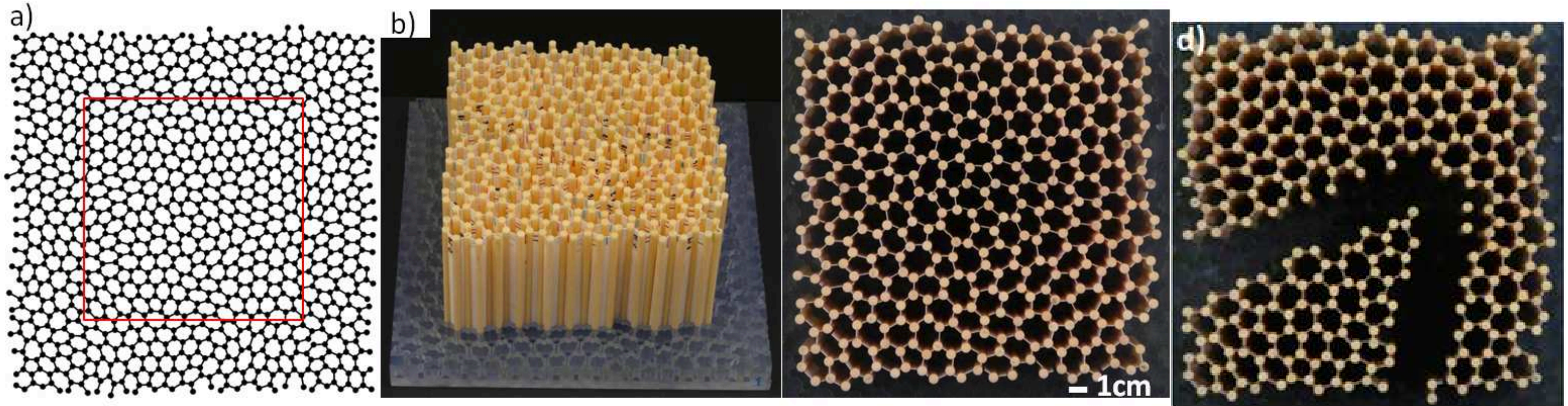

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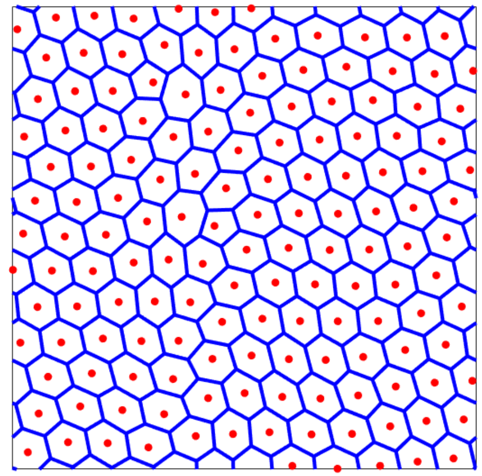
Subsequent Wave Propagation Studies

- High-density transparent 2D stealthy hyperuniform (SH) materials: Leseur et al. Optica (2016).
- For TM polarization, 2D SH materials show a rich "phase diagram" as a function of χ - transparency, diffusive, PBG and localization regimes: Froufe-Pérez et al. PNAS (2017).
- Recent analogous 3D study showing weak-localization and transparency regimes depending on χ: Sgrignuoli, Torquato and Dal Negro, PRB (2021).
- 2D SH materials are nearly optimal wave absorbers: Bigourdan et al. Opt. Exp. (2018).
- SH composite lens can dramatically reduce back scattering relative to its periodic counterparts: Zhang et al. APL (2019).
- Predictive nonlocal theory of effective dynamic dielectric constant across the first 3 dimensions for general composite microstructures, including SH materials: Torquato and Kim, PRX (2020).
- Over 65% sunlight absorption in a 1 μm Si Slab with SH texture: Tavakoli et al. ACS Phot. (2022)

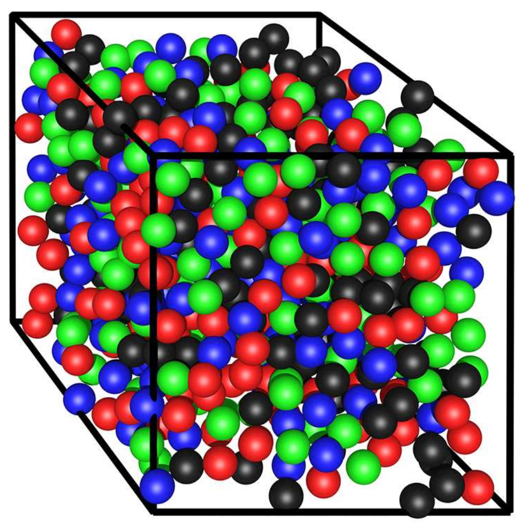
Disordered Stealthy Hyperuniform Materials with Optimal Transport/Elastic Propert

Optimal Effective Conductivity & Elastic Moduli in 2D High-χ Stealthy Networks



Chen & Torquato, Multifunctional Materials (2018)

Optimal Effective Diffusivity in Decorated 3D Stealthy Patterns



Zhang, Stillinger& Torquato, J. Chem. Phys. (2016)

WHY DO DISORDERED STEALTHY HYPERUNIFORM MATERIALS WITH SUFFICIENTLY HIGH χ VALUES YIELD DESIRABLE PHYSICAL PROPERTIES?

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MATERIALS WITH SUFFICIENTLY HIGH χ VALUES

YIELD DESIRABLE PHYSICAL PROPERTIES?

They are rotationally invariant disordered materials that maximally suppress density fluctuations from intermediate to infinite wave-lengths and the "bounded-hole" property, i.e., holes of arbitrarily large size are prohibited in thermodynamic limit (Torquato, Physics Reports 2017).

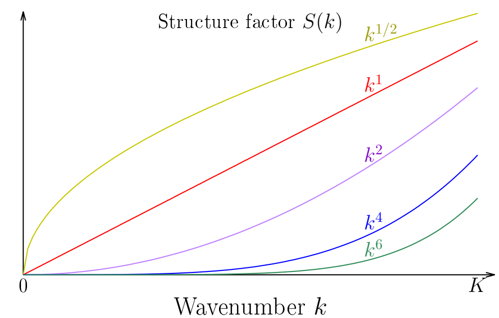
- **We derived (Zhang, Stillinger & Torquato, Soft Matter 2017) that the maximum hole size R_{max} is bounded from above for any d by**

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

- **This bound was proved by Ghosh and Lebowitz, Comm. Math. Phys. (2018)**

Targeted Spectra $S(k) \sim k^\alpha$

- **Can target scaling of structure factor $S(k) \sim k^\alpha$ for $k \to 0$ using collective-coordinate optimization procedure.**



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

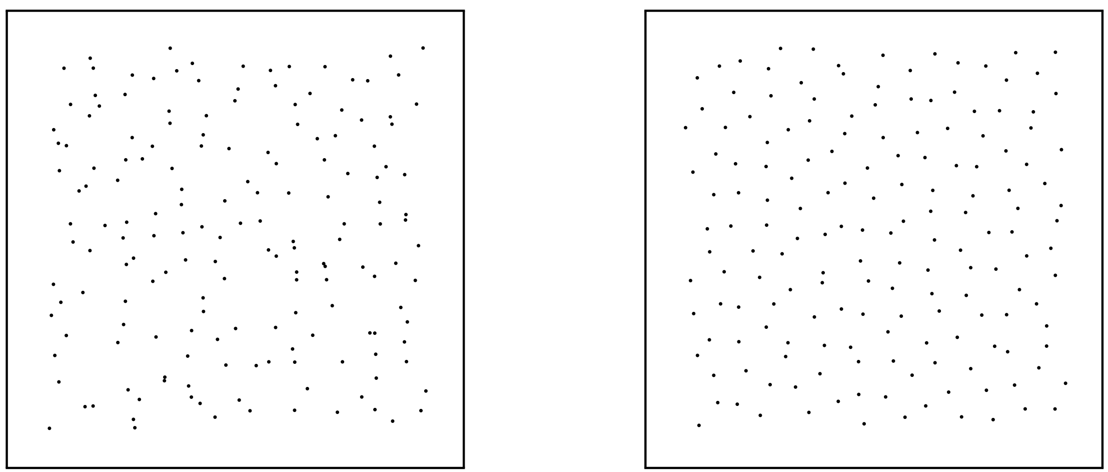


Figure 1: **One of them is for $S(k) \sim k^6$ and other for $S(k) \sim k$.**

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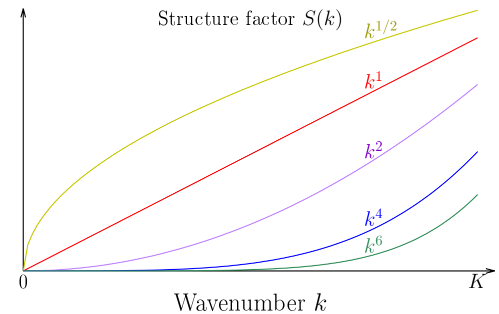

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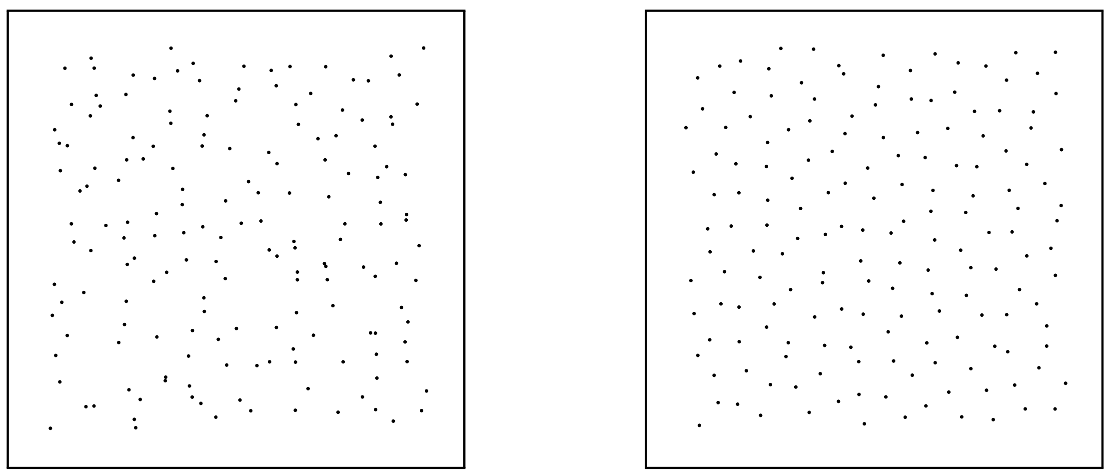

Figure 1: One of them is for $S(k) \sim k^6$ and other for $S(k) \sim k$.

- **This procedure leads to the perfect glass paradigm, corresponding to disordered hyperuniform ground states mechanically stable and eliminate the possibilities of crystalline and quasicrystalline phases from the ground-state manifold.**

Multifunctional Composites for Elastic and Electromagnetic Wave Propagation

Kim and Torquato, Proc. Nat. Acad. Sci. (2020)
Kim and Torquato, New Journal of Physics (2020)
Torquato and Kim, Physical Review X (2021)

- Derived exact expansions for effective dynamic elastic and electromagnetic wave characteristics of composites of arbitrary microstructures that apply well beyond the quasistatic (long-wavelength) regime.
- Extracted accurate formulas for effective dynamic dielectric constant & elastic moduli, each which depends on microstructure via spectral density $\tilde{\chi}_V(\mathbf{k})$.

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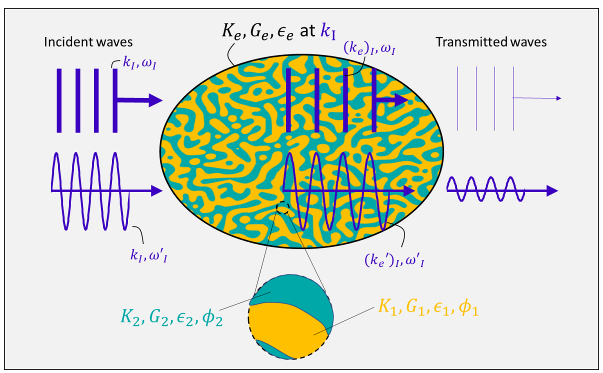
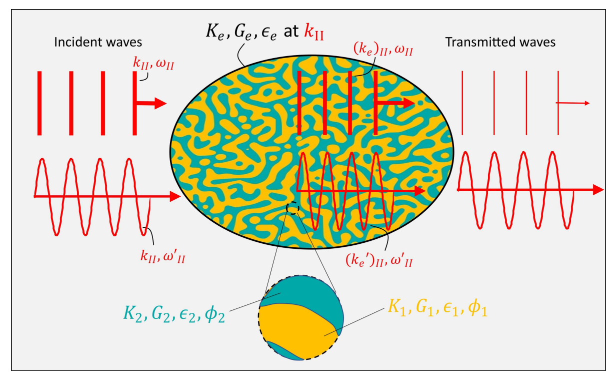
- Extracted accurate formulas for effective dynamic dielectric constant & elastic moduli, each which depends on microstructure via spectral density $\tilde{\chi}_V(\mathbf{k})$.
- By eliminating this common microstructural quantity, found "cross-property relations" that link effective elastic and electromagnetic wave characteristics to one another, facilitating multifunctional design.





Left panel: Both elastic and electromagnetic waves can be attenuated due to scattering.
Right panel: Composite attenuates elastic waves but is transparent to electromagnetic waves.

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

Do PBGs of Disordered Dielectric Networks Persist in Thermodynamic Limit?

CONJECTURE: High-χ stealthy hyperuniformity is a necessary condition for an isotropic disordered network to have a complete photonic band gap in the thermodynamic limit. (Torquato, Physics Reports 2017)

This conjecture is based on

- **Structural "uniformity" from very large to intermediate length scales: Batten, Stillinger& Torquato, J. Appl. Phys. (2008)**
- **High-χ opens up complete PBGs for finite systems: Florescu, Torquato & Steinhardt, PNAS (2009)**
- **"Bounded-hole" property in thermodynamic limit: Zhang, Stillinger& Torquato, Soft Matter (2017)**

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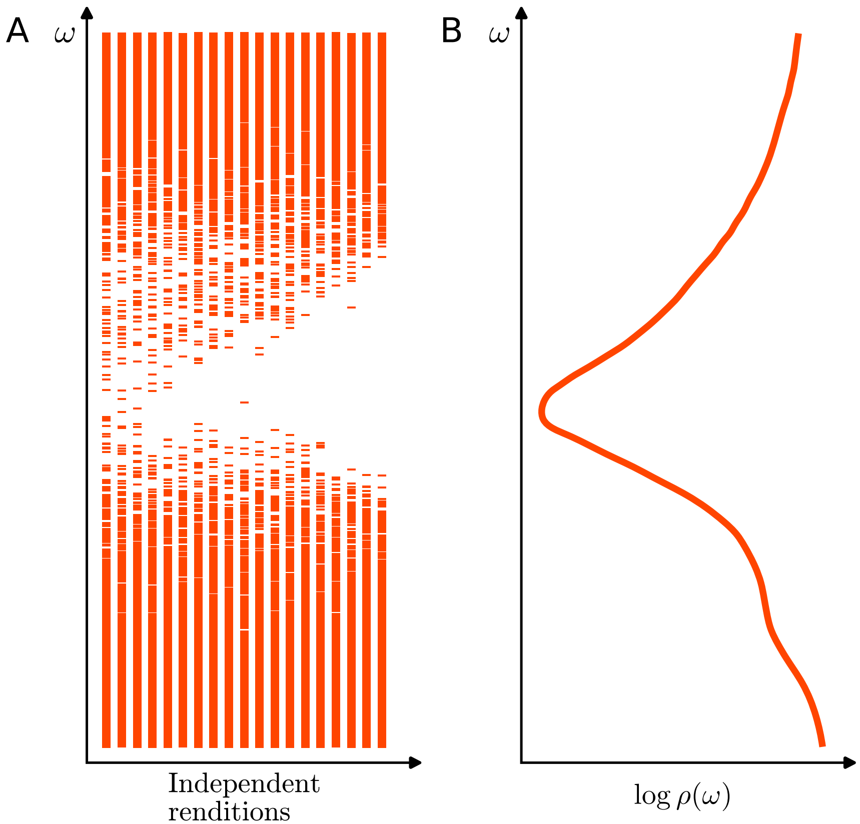
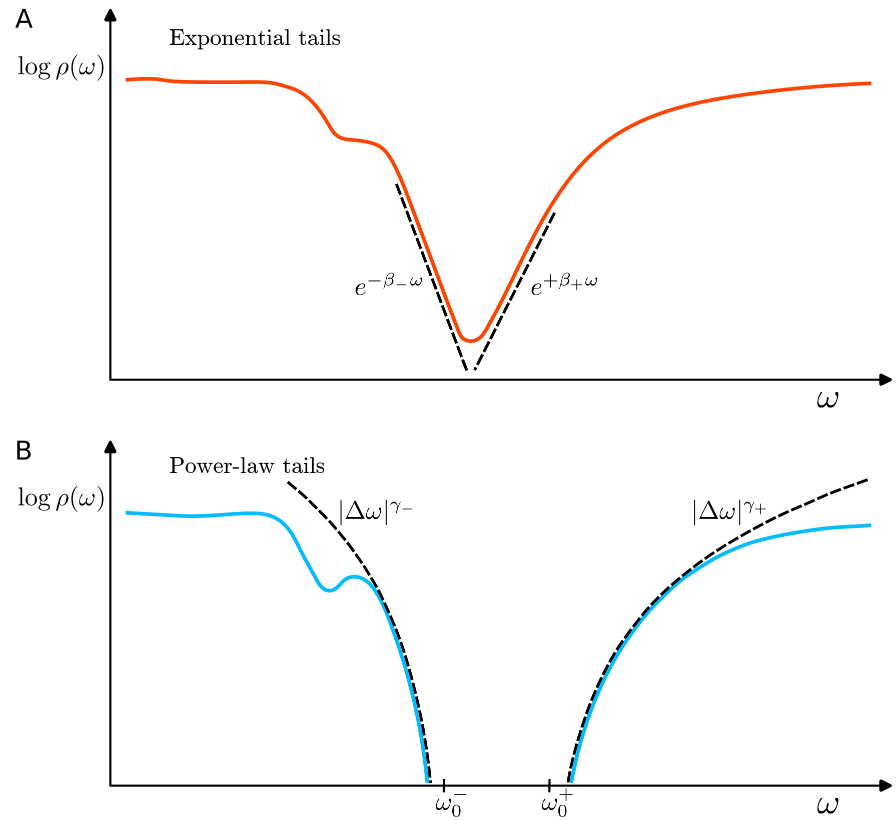
Testing the Conjecture

Klatt, Steinhardt & Torquato, submitted to the PNAS

- We use a two-stage ensemble approach to study the formation of complete PBGs for a sequence of increasingly large systems spanning a broad range of 2D photonic network solids with varying degrees of local and global order, including hyperuniform and nonhyperuniform types.
- Except for high-χ stealthy hyperuniform cases, we discover that the gap in the density of states exhibits exponential tails and the apparent PBGs rapidly close as the system size increases for nearly all disordered networks considered.
- PBGs for high-χ stealthy hyperuniform cases remain open and the band tails exhibit a desirable power-law scaling reminiscent of the PBG behavior of photonic crystals in thermodynamic limit.

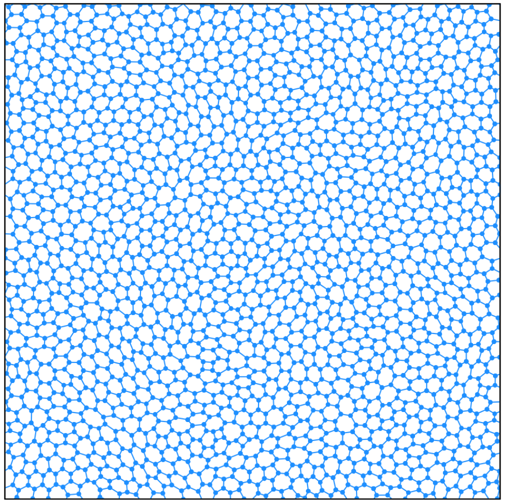
Two-Stage Density of States Ensemble Approach





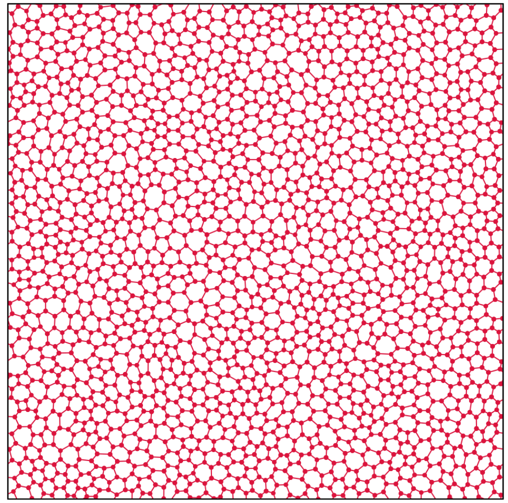
Network Models

Equiluminous



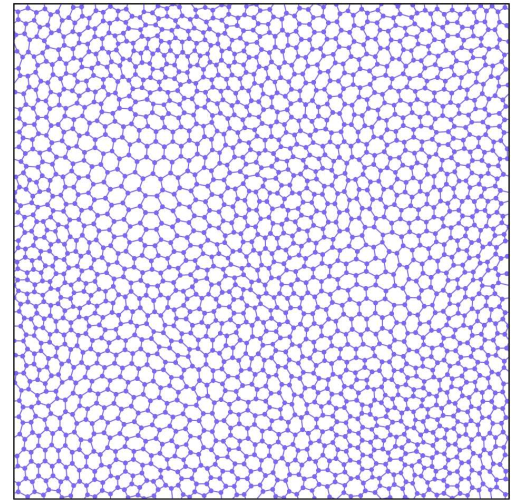
Nonhyperuniform
Nonstealthy
Unbounded holes

Random Sequential Addition



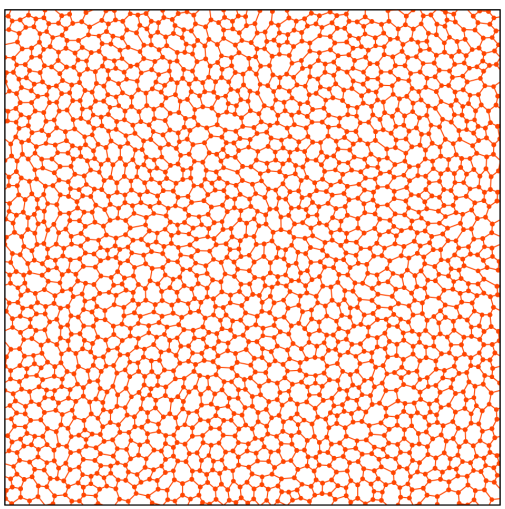
Nonhyperuniform
Nonstealthy
Bounded holes

Stealthy Nonhyperuniform



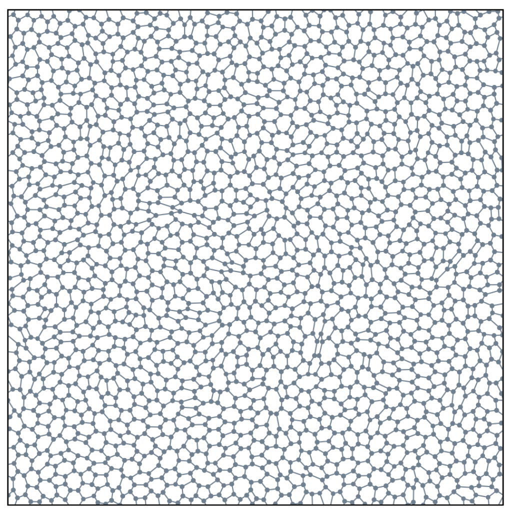
Nonhyperuniform
Stealthy
Unbounded holes

Perfect Glass



Hyperuniform
Nonstealthy
Bounded holes

Low-χ Stealthy Hyperuniform



Hyperuniform
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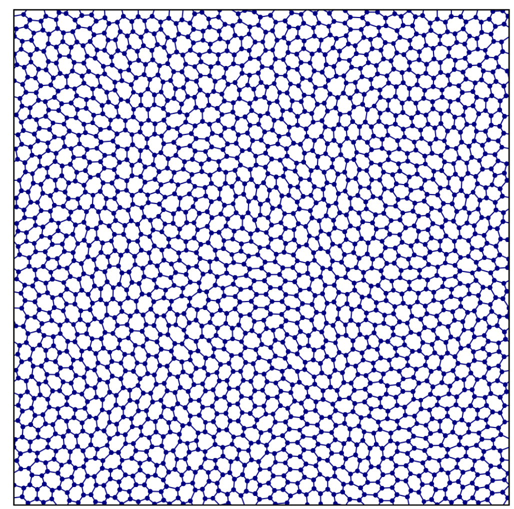

Hyperuniform
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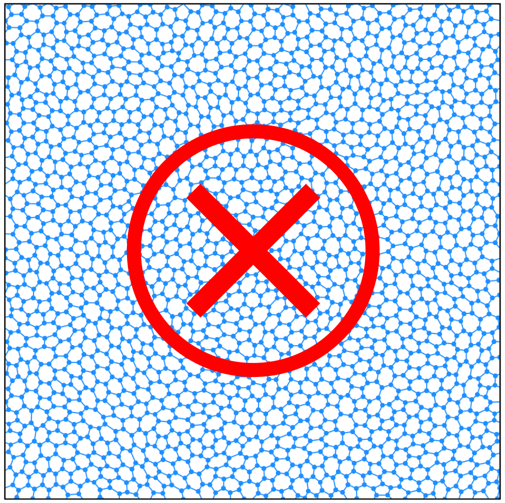
Which Networks Survive? Confirmation of Conjecture

Equiluminous



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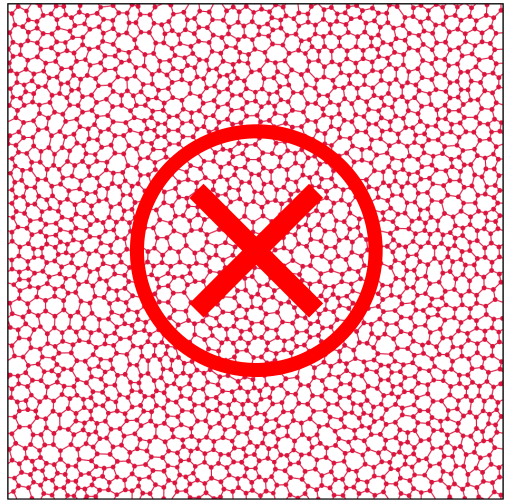

Nonhyperuniform
Nonstealthy
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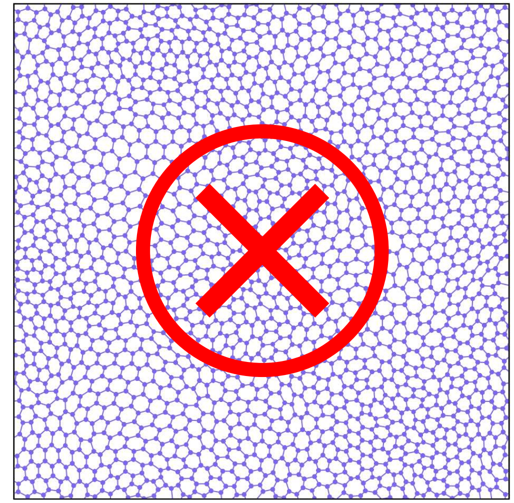
Stealthy Nonhyperuniform



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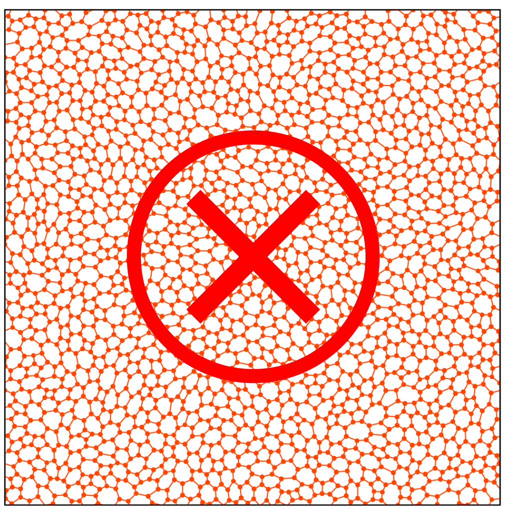

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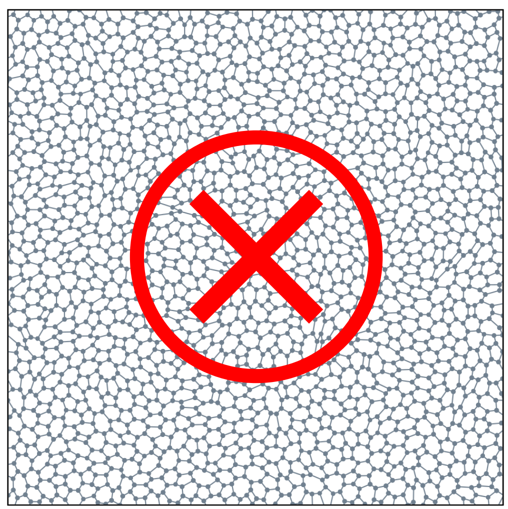

Hyperuniform
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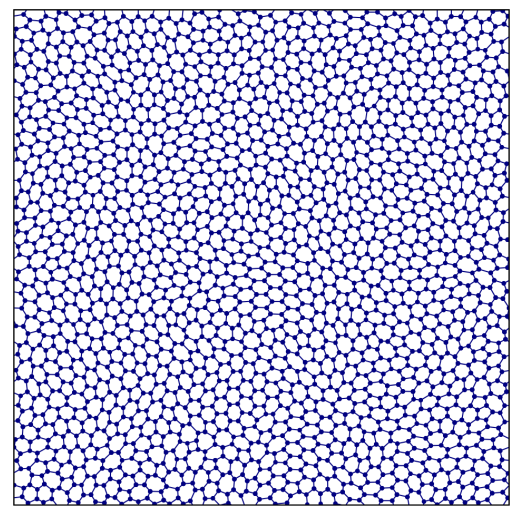

Hyperuniform
Stealthy
Bounded holes

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) and forces us to re-think the meaning of randomness across length scales.
- 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.
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<u>Collaborators</u>

Roberto Car (Princeton)
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Weining Man (San Francisco State)
Erdal Oğŭz (Beijing)
Antonello Scardicchio (ICTP)
Josh Socolar (Duke Univ.)
Fabrizio Sgrignuoli (Boston Univ.)
Paul Steinhardt (Princeton)
Frank Stillinger (Princeton)
Chase Zachary (Princeton)
Ge Zhang (Princeton/Hong Kong)