Sensitive Microstructural Descriptors of Disordered Heterogeneous Materials Across Length Scales for Materials Discovery

Salvatore Torquato

Department of Chemistry,

Department of Physics,

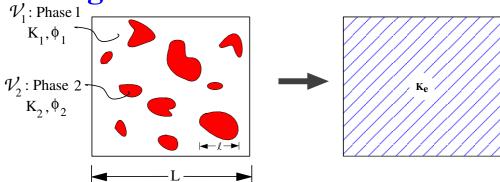
Princeton Institute of Materials,

and Program in Applied & Computational Mathematics

UCLA IPAM Workshop II: Bridging Scales from Atomistic to Continuum in Electrochemical Systems

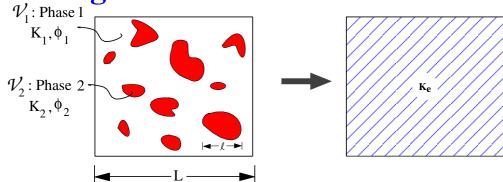
October 7, 2025

Heterogeneous Materials: Preliminaries



Examples: Porous and composite media,, foams, colloids, battery materials, geologic media, biological membranes, animal and plant tissue, etc.

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Effective Properties

- 1. Effective Conductivity Tensor, σ_e
- 2. Effective Diffusion Tensor, D_e
- 3. Effective Stiffness Tensor, C_e
- 4. Effective Viscosity, μ_e
- 5. Mean Survival Time, τ
- 6. Diffusion and Viscous Relaxation Times, T_1, Θ_1
- 7. Fluid Permeability Tensor, k
- 8. Nonlinear Mechanical Properties
- 9. Optical and Other Wave Properties

Seemingly different properties are interrelated: Cross-Property Relations

Effective properties are sensitive to the details of the microstructure, i.e., volume fractions; orientations, sizes and shapes of the phase domains; spatial distribution of the domains; connectedness of the phases, etc.

OUTLINE

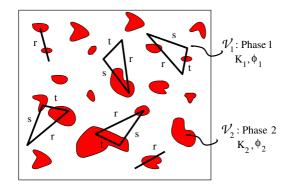
- Statistical Descriptors of Two-Phase Media
- Microstructure-Dependent Estimates on Transport Properties
- Hyperuniform Two-Phase Media
- Quantifying Order/Disorder Across Length Scales

Statistical Descriptors of Two-Phase Media

• A two-phase medium in \mathbb{R}^d is fully statistically characterized by the n-point correlation (probability) functions:

$$S_n^{(i)}(\mathbf{x}_1,...,\mathbf{x}_n) \equiv \left\langle \mathcal{I}^{(i)}(\mathbf{x}_1) ... \mathcal{I}^{(i)}(\mathbf{x}_n) \right\rangle,$$

where $n=1,2,3,\ldots$, angular brackets denote an ensemble average, and $\mathcal{I}^{(i)}(\mathbf{x})$ is the indicator function for phase i=1,2:



The $S_n^{(i)}$'s arise in rigorous expressions (exact expansions and bounds) for the effective conductivity, elastic moduli, diffusion properties, fluid permeability, and optical properties.

Statistical Descriptors of Two-Phase Media

Interfacial n-point Correlation Functions

The interface between the phases of a realization of a two-phase medium is characterized the interface indicator function $\mathcal{M}(\mathbf{x})$ defined as

$$\mathcal{M}(\mathbf{x}) = |\nabla \mathcal{I}^{(1)}(\mathbf{x})| = |\nabla \mathcal{I}^{(2)}(\mathbf{x})| \tag{1}$$

lacksquare One type of interfacial n-point correlation function is defined as

$$F_{sss...}(\mathbf{x}_1,...,\mathbf{x}_n) \equiv \langle \mathcal{M}(\mathbf{x}_1)...\mathcal{M}(\mathbf{x}_n) \rangle$$
.

lacksquare The specific surface s (interface area per unit volume) is the one-point correlation function:

$$s = \langle \mathcal{M}(\mathbf{x}) \rangle.$$

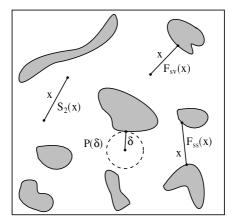
The interfacial two-point or surface-surface correlation function is particularly important:

$$F_{ss}(\mathbf{r}) = \langle \mathcal{M}(\mathbf{x}) \mathcal{M}(\mathbf{x} + \mathbf{r}) \rangle$$

Interfacial n-point correlation functions naturally arise in rigorous bounds on effective properties in which the interface plays a major role, e.g., diffusion-controlled reactions and fluid permeability.

Some 2-Point Correlation Functions

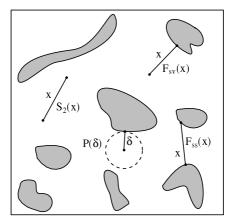
Two-Point Functions for General Media



- ullet $S_2^{(i)}(r)$: two-point correlation function for phase i.
- $F_{sv}(r)$: surface-void correlation function
- ullet $F_{ss}(r)$: surface-surface correlation function
- ullet $P(\delta)$: pore-size probability density function

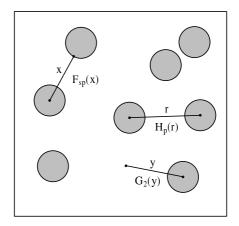
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Two-Point Functions for Particulate Media



- $F_{sp}(r)$: surface-particle correlation function
- ullet $G_2(r)$: void-particle surface-void correlation function
- ullet $H_P(r)$: nearest-neighbor probability density function

Spectral Density

lacksquare The autocovariance function $\chi_V^{}({f r})$ is defined by

$$\chi_V(\mathbf{r}) \equiv S_2^{(1)}(\mathbf{r}) - \phi_1^2 = S_2^{(2)}(\mathbf{r}) - \phi_2^2.$$

- The nonnegative spectral density $\tilde{\chi}_V(\mathbf{k})$ at wavevector \mathbf{k} , which can be obtained from scattering experiments, is the Fourier transform of $\chi_V(\mathbf{r})$.
- m P In the large-k limit, it decays as an inverse power law with coefficient prop. to specific surface s:

$$\tilde{\chi}_V(\mathbf{k}) \sim \frac{\gamma(d) s}{k^{d+1}}, \qquad k \to \infty.$$

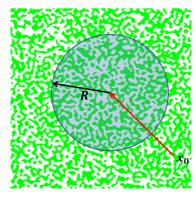
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Local volume-fraction variance $\sigma_V^2(R)$ within a spherical window of radius R is given in terms of $\chi_V({\bf r})$ or $\tilde{\chi}_V({\bf k})$ (Lu and Torquato, 1990; Zachary and Torquato, 2009):

$$\sigma_V^2(R) = \frac{1}{v_1(R)} \int_{\mathbb{R}^d} \chi_V(\mathbf{r}) \,\alpha_2(r; R) d\mathbf{r}$$
$$= \frac{1}{v_1(R)(2\pi)^d} \int_{\mathbb{R}^d} \tilde{\chi}_V(\mathbf{k}) \tilde{\alpha}_2(k; R) d\mathbf{k},$$

where $v_1(R)$ is the volume of a d-dimensional sphere of radius R, $\alpha_2(r;R)$ is the scaled volume common to 2 windows separated by a distance r and $\tilde{\alpha}_2(k;R)$ is its Fourier transform.

Transport Properties of Fluid Saturated Porous Media

NMR Measurements

- Nuclear magnetic resonance (NMR) relaxation times of porous media provide useful probes of the pore-phase microstructure.
 - Mean survival time, au
 - **Diffusion relaxation times,** T_1, T_2, T_3, \ldots , where T_1 is the largest or principal relaxation time.

Conduction Measurements

- Consider a porous medium whose pore space is filled with an electrically conducting fluid of conductivity σ_1 and a solid phase that is perfectly insulating (σ_2 = 0).
 - We consider dimensionless effective conductivity

$$\frac{\sigma_e}{\sigma_1} = \mathcal{F}^{-1}$$

where \mathcal{F} is the formation factor, which a measure of the tortuosity or degree of "windiness for electrical transport pathways in the pore phase.

Stokes Flow and Fluid Permeability

- Consider Stokes flow through a porous medium.
 - ullet Fluid permeability, k
 - ▶ Viscous relaxation times, $\Theta_1, \Theta_2, \Theta_3, \ldots$, where Θ_1 is the largest or principal relaxation time.

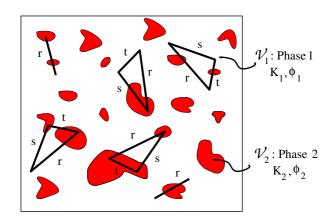
Microstructure-Dependent Estimates on Transport Properties

Exact Representation of Effective Dynamic Dielectric Tensor

Nonlocal strong-contrast expansions for effective dynamic dielectric tensor $\varepsilon_e(\mathbf{k}_q)$ that exactly account for complete microstructural information and hence multiple scattering to all orders for the range of wavenumbers for which our extended homogenization theory applies, i.e., $0 \leq |\mathbf{k}_q| \ell \lesssim 1$ (Torquato and Kim, PRX, 2021):

$$\phi_p^2 \beta_{pq}^2 [\boldsymbol{\varepsilon}_e(\mathbf{k}_q) + (d-1)\boldsymbol{\varepsilon}_q \boldsymbol{I}] \cdot [\boldsymbol{\varepsilon}_e(\mathbf{k}_q) - \boldsymbol{\varepsilon}_q \boldsymbol{I}]^{-1} = \phi_p \beta_{pq} \boldsymbol{I} - \sum_{n=2}^{\infty} \boldsymbol{A}_n^{(p)}(\mathbf{k}_q) \beta_{pq}^n,$$

where $\beta_{pq}=f(\varepsilon_p,\varepsilon_q)$ is a generalized "polarizability", $\boldsymbol{A}_n^{(p)}(\mathbf{k}_q)$ is a wavevector-dependent second-rank tensor that is a functional involving the set of correlation functions $S_1^{(p)},S_2^{(p)},\cdots,S_n^{(p)}$ and products of the dyadic Green's function $\boldsymbol{H}^{(q)}(\mathbf{r})$.



- Due to the rapid convergence, even for large contrasts, their lower-order truncations yield accurate closed-form approximate formulas for $\varepsilon_e(\mathbf{k}_q)$.
- Such nonlocal formulas are resummed representations of the expansions that still accurately capture multiple scattering to all orders.

Microstructure-Dependent Bounds on Transport Properties

NMR Time Scales

The mean survival time τ in the diffusion-controlled limit is bounded from above by an integral over the spectral density (Torquato, 2020):

$$D\tau \leq \frac{1}{\phi_1\,\phi_2^2}\,\ell_P^2,$$

$$\ell_P^2 = \int_0^\infty \chi_V(r)\,rdr = \frac{1}{2\pi^2}\int_0^\infty \tilde\chi_V(k)dk.$$

where

It is bounded from below in terms of the mean pore size (Torquato and Avellaneda, 1991):

$$\tau \ge \frac{\langle \delta \rangle^2}{D}.$$

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Formation Factor

Derived a tight lower bound on ${\mathcal F}$ for any 3D porous medium that accounts for up to four-point correlation function information (Torquato, 2020):

$$\mathcal{F} \geq \frac{1 + \frac{1}{2} \frac{\gamma_2}{\zeta_2} - \frac{1}{2} \zeta_2 + \left(\frac{1}{2} + \frac{1}{2} \zeta_2 + \frac{1}{4} \frac{\gamma_2}{\zeta_2}\right) \phi_2}{1 + \frac{1}{2} \frac{\gamma_2}{\zeta_2} - \frac{1}{2} \zeta_2 + \left(-1 + \frac{1}{2} \zeta_2 - \frac{1}{2} \frac{\gamma_2}{\zeta_2}\right) \phi_2},$$

$$\zeta_2 = \zeta_2 [S_1^{(2)}, S_2^{(2)}, S_3^{(2)}] \text{: three-point microstructural parameter}$$

$$\gamma_2 = \gamma_2 [S_1^{(2)}, S_2^{(2)}, S_3^{(2)}, S_3^{(2)}] \text{: four-point microstructural parameter}$$

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Fluid Permeability

Permeability is bounded from above in terms of the same length scale ℓ_P (Torquato, 2020):

$$k \le \frac{2}{3\phi_2^2} \,\ell_P^2,$$

While this two-point "void" bound is not tight, it correctly rank orders the permeabilities of different models

Link Between Permeability and Diffusion Parameters

Exact relation (Avellaneda and Torquato, 1991):

$$k = \frac{\mathcal{L}^2}{8\mathcal{F}},$$

where is \mathcal{L} is a length parameter that is a sum over the times $\Theta_1.\Theta_2,\ldots$

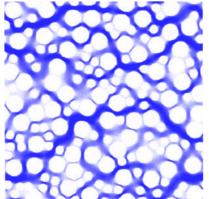


Figure 1: Laminar flow through the void space of a two-dimensional hard-disk packing. The brightness of the color indicates the magnitude of the flow velocity.

Rigorous bounds (Torquato, 1990; Avellaneda and Torquato, 1991):

$$k \leq D\phi_1 \tau$$
,

$$k \le \frac{DT_1}{\mathcal{F}}$$

Approximation valid when pore space is well connected (Torquato, 2020)

$$k \approx \phi_1 \frac{\langle \delta^2 \rangle}{\mathcal{F}}$$

Predictions of Permeability Approximation

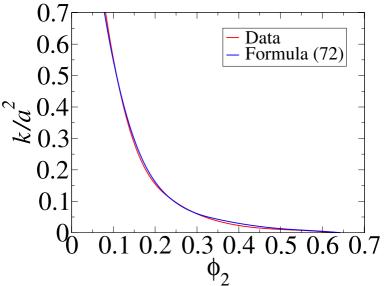


Figure 2: Comparison of computer-simulation data for the fluid permeability of BCC packings as a function of ϕ_2 to the predictions of the approximation formula.

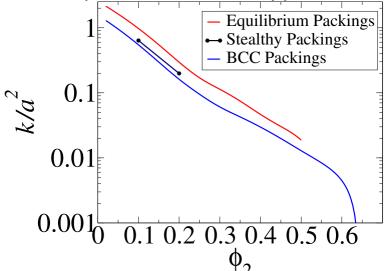
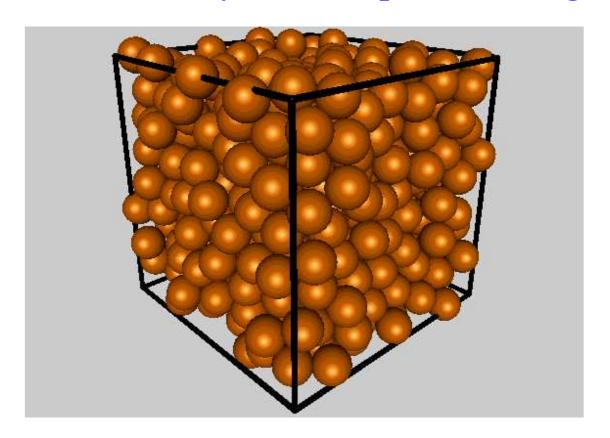


Figure 3: Estimates of the dimensionless fluid permeability k/a^2 as a function of ϕ_2 predicted by the approximation formula for the disordered stealthy, equilibrium, and BCC packings.

Permeability of MRJ Sphere Packings



- In the approximation formula for the permeability of the MRJ packing yields $k/a^2 \approx 0.010$ at porosity $\phi_1=0.374$, where a is the radius of a sphere. This is to be contrasted with the corresponding BCC packing with $k/a^2 \approx 0.0016$, which is much lower.
- This is consistent with the fact that the pore space in the MRJ packing is more localized and less uniformly dynamically connected than that of the BCC packing.

Hyperuniformity of Two-Phase Media

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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.
- This enables a new structural classification scheme that encompasses all of these states according to this generalization of long-range order.

Disordered Hyperuniform States

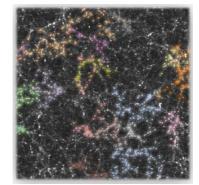
- Disordered hyperuniform many-particle systems can be regarded to be new ideal states of matter in that they
 - 1. behave more like crystals or quasicrystals in the way they suppress large-scale density fluctuations, and yet are also like liquids and glasses, since they are statistically isotropic structures 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.

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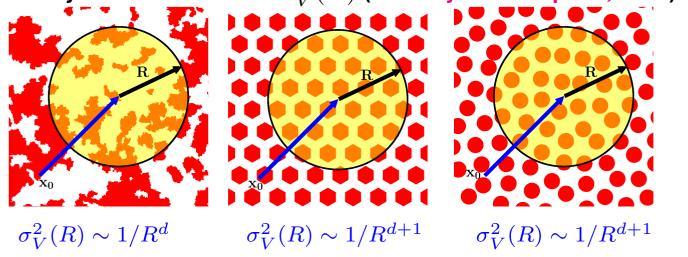
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- Existence of disordered hyperuniform states forces us to re-think what we mean by "disorder" and on what length scales.
- Such exotic correlated disordered states arise in a myriad of contexts:
 - Classical equilibrium liquids and ground states
 - Classical nonequilibrium systems
 - Quantum systems
 - Sphere packings
 - Random matrices
 - Dynamical systems and quantum chaos
 - **Number theory** (e.g., prime numbers; Torquato et al. J. Phys. A, 2019)
 - Biological systems
 - Novel materials
 - Large-scale structure of the Universe (Philcox & Torquato, PRX, 2023)



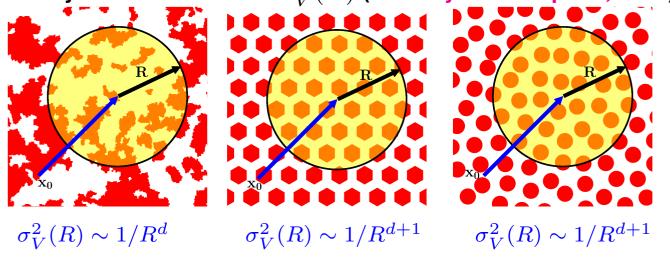
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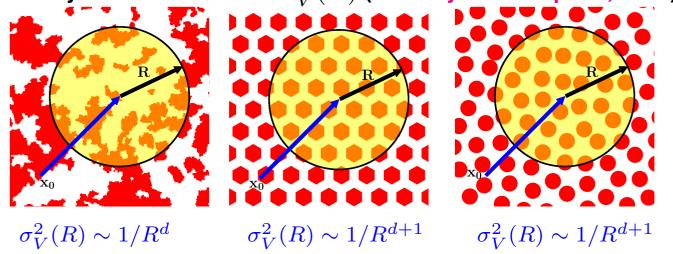


- For typical disordered media, volume-fraction variance $\sigma_V^2(R)$ for large R goes to zero like R^{-d} .
- For hyperuniform two-phase media, $\sigma_V^2(R)$ goes to zero faster than R^{-d} , equivalent to following condition on spectral density $\tilde{\chi}_V(\mathbf{k})$:

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Similarly, variance in surface-area fluctuations $\sigma_S^2(R)$ also goes to zero faster than R^{-d} for hyperuniform media or, equivalently (Torquato, PRE, 2016)

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

General Hyperuniform Scaling Behaviors

Consider power-law spectral density

$$\tilde{\chi}_V(\mathbf{k}) \sim |\mathbf{k}|^{\alpha}, \quad (|\mathbf{k}| \to \mathbf{0})$$

lacksquare Can prove following large-R scalings (Torquato, Phys. Rep. 2018):

$$\sigma_V^2(R) \sim \begin{cases} R^{-(d+1)}, & \alpha > 1 & \text{(Class I)} \\ R^{-(d+1)} \ln R, & \alpha = 1 & \text{(Class II)} \\ R^{-(d+\alpha)}, & 0 < \alpha < 1 & \text{(Class III)}. \end{cases}$$

- Classes I and III are the strongest and weakest forms of hyperuniformity, respectively. Class I media include all crystals, many quasicrystals and exotic disordered media.
- Stealthy hyperuniform media are also of class I:

$$\tilde{\chi}_V(\mathbf{k}) = 0 \qquad \text{for } 0 \le |\mathbf{k}| \le K.$$

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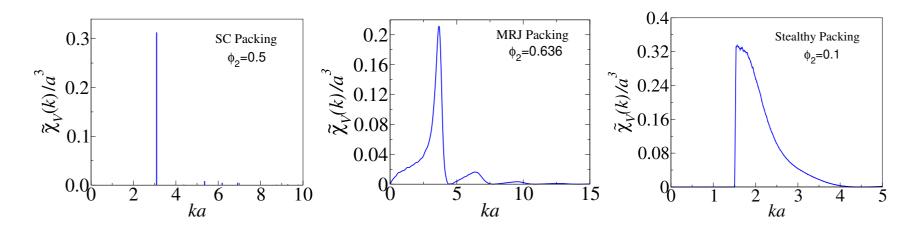
General Nonhyperuniform Scaling Behaviors

$$\sigma_V^2(R) \sim \begin{cases} R^{-d}, & \alpha = 0 \text{ (typical nonhyperuniform)} \\ R^{-(d+\alpha)}, & -d < \alpha < 0 \text{ (antihyperuniform)}. \end{cases}$$

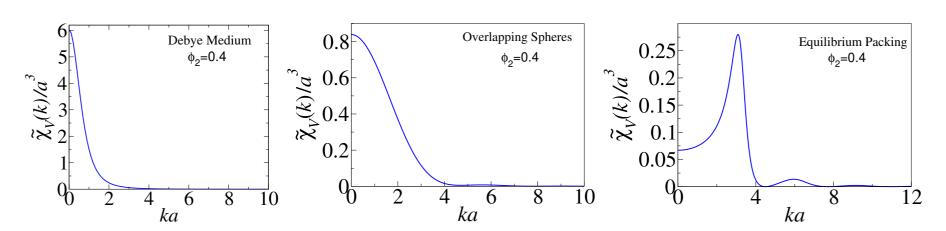
- For a "typical" nonhyperuniform media, $\tilde{\chi}_V(0)$ is bounded, but is unbounded for antihyperuniform media (e.g., fractal structures).
- Thus, can classify all translationally invariant states of matter according to their large-scale fluctuations (Torquato, PRE, 2021).

Spectral Densities of Hyperuniform and Nonhyperuniform Media

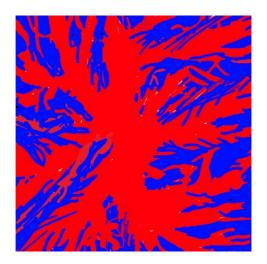
3 Different Hyperuniform Models

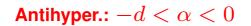


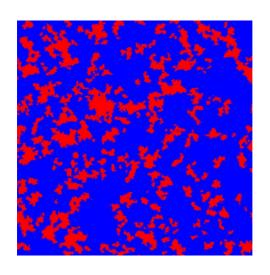
3 Different Nonhyperuniform Models



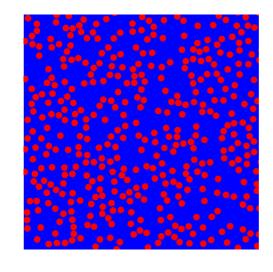
Examples of Microstructures Spanning Across the Possible Spectrum



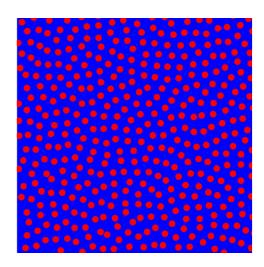




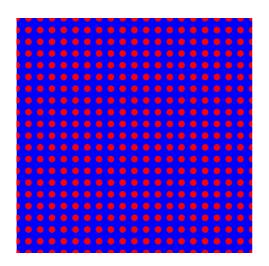
Typical nonhyper.: $\alpha = 0$



Nonstealthy hyper.: $0 < \alpha < \infty$



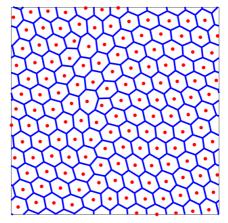
Disordered stealthy hyperuniform: $\alpha = \infty$



Ordered stealthy hyperuniform: $\alpha = \infty$

Optimality of Disordered Hyperuniform Two-Phase Media

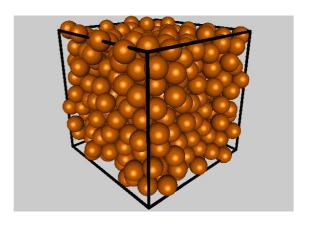
- Disordered hyperuniform dielectric networks have the largest isotropic photonic band gaps: Florescu, Steinhardt and Torquato, PNAS (2009).
- Disordered hyperuniform media have a rich optical "phase diagram" transparency, diffusive, PBG and localization regimes: Froufe-Pérez et al. PNAS (2017).
- Disordered hyperuniform networks have maximal effective thermal (electrical) conductivities as well as maximal effective bulk and shear moduli: Chen & Torquato, Multifunctional Materials (2018)



- Disordered hyperuniform materials are nearly optimal wave absorbers: Bigourdan et al. Opt. Exp. (2018).
- Disordered hyperuniform composite lenses can dramatically reduce back scattering relative to its periodic counterparts: Zhang et al. APL (2019).

Quantifying Order/Disorder Across Length Scales

Quantifying Disorder/Order of Microstructures

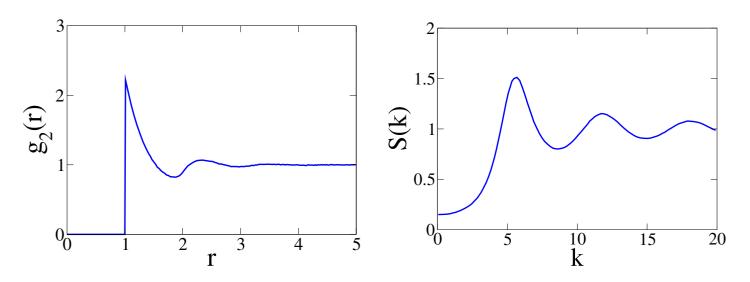




- Must settle for reduced information. For example, scalar order metrics $\psi_1, \psi_2, \psi_3, \ldots$ such that $0 \le \psi_i \le 1$.
- Can order metrics be devised consistent with our intuition:
 - Perfect crystals
 - Perturbed crystals
 - Quasicrystals
 - Highly defective crystals
 - Correlated random systems (e.g., "maximally random jammed" state)
 - Uncorrelated random systems (ideal gases)
- Crucially, should be able detect degree of order across length and time scales.
- Variety of useful translational and orientational order metrics have been introduced (Torquato & Stillinger, Rev. Mod. Phys. 2010).
- Order metrics generally may be tensor quantities.

Detecting Order/Disorder Across Length Scales

• Consider a many-particle system with positive and negative correlations as measured by the pair correlation function $g_2(\mathbf{r})$ and structure factor S(k):



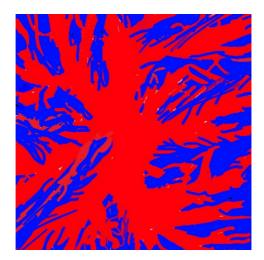
■ The τ order metric characterizes translational order (Torquato, Zhang& Stillinger, PRX, 2015):

$$\tau = \frac{1}{D^d} \int_{\mathbb{R}^d} [g_2(\mathbf{r}) - 1]^2 d\mathbf{r} = \frac{1}{(2\pi)^d D^d} \int_{\mathbb{R}^d} [S(\mathbf{k}) - 1]^2 d\mathbf{k}$$

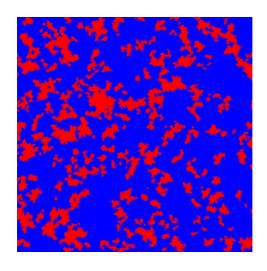
ullet Deviations of the pair functions from unity, whether positive or negative, are picked by au, and has been fruitfully applied to describe a broad spectrum of many-particle systems.

Local Order Metrics Across the Spectrum of Two-Phase Media

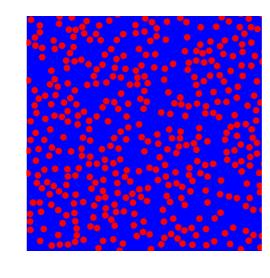
Devising order metrics to characterize and classify microstructures across length scales is a highly challenging task, given the richness of the possible phase geometries and topologies.



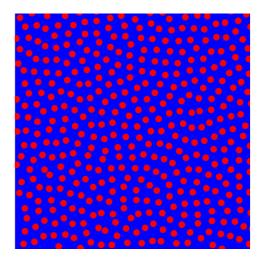
Antihyper.: $-d < \alpha < 0$



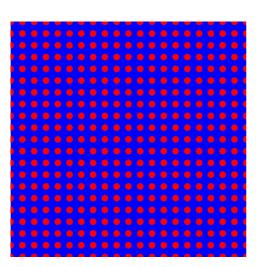
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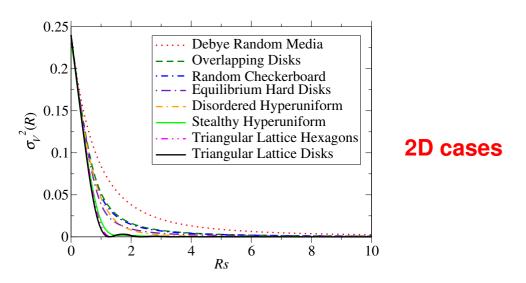


Ordered stealthy hyperuniform: $\alpha = \infty$

Local Order Metrics Across the Spectrum of Two-Phase Media

Torquato, Skolnick and Kim, J. Phys. A: Math. Theory (2022)

- ${\color{red} \blacktriangleright}$ For this purpose, we propose the use of the local volume- fraction variance $\sigma^2_{_V}(R)$ associated with a spherical window of radius R as an local order metric.
- lacksquare We determined $\sigma_V^2(R)$ as a function of R for 22 different models across the first three space dimensions, including both hyperuniform and non-hyperuniform systems with varying degrees of short- and long-range order.

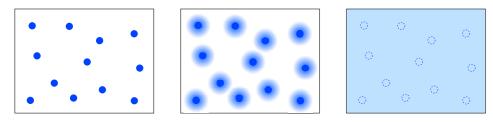


For any particular value of R, the lower (higher) the value of $\sigma_V^2(Rs)$, the greater the degree of order (disorder).

Diffusion Spreadability as a Dynamic Probe of Microstructure

Torquato, Phys. Rev. E (2021)

- At t=0, assumed a solute is uniformly distributed throughout phase 2 with volume fraction ϕ_2 , and completely absent from phase 1 with volume fraction ϕ_1 , and each phase has same diffusion coefficient D.
- Problem: Calculate "spreadability" S(t), fraction of the total amount of solute present that has diffused into phase 1 at time t.



Excess spreadability has following Fourier representation:

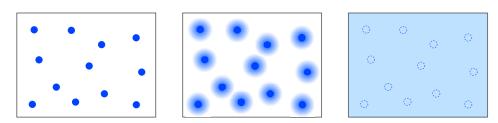
$$\mathcal{S}(\infty) - \mathcal{S}(t) = \frac{1}{(2\pi)^d \phi_2} \int_{\mathbb{R}^d} \tilde{\chi}_V(\mathbf{k}) \exp[-k^2 Dt] d\mathbf{k} \ge 0,$$

where $\tilde{\chi}_{_{V}}(\mathbf{k})$ is the spectral density.

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 $\begin{tabular}{ll} \blacksquare & \textbf{The time-dependent diffusion spreadability } \mathcal{S}(t) \ \text{relates its short-,} \\ & \textbf{intermediate- and long-time behaviors link to the small-, intermediate- and} \\ & \textbf{large-scale structure of heterogeneous materials.} \\ \end{tabular}$

Spreadability Phase Diagram

Consider power-law scaling form for spectral density:

$$\lim_{|\mathbf{k}| \to \mathbf{0}} \tilde{\chi}_V(\mathbf{k}) = B|\mathbf{k}a|^{\alpha} \qquad (|\mathbf{k}| \to \mathbf{0}),$$

where B is a positive constant and $\alpha \in (-d, \infty)$.

m extstyle extstyle

$$S(\infty) - S(t) = \frac{B \Gamma((d+\alpha)/2) \phi_2}{\pi^{d/2} \Gamma(d/2) (Dt/a^2)^{(d+\alpha)/2}} + o\left((Dt/a^2)^{-(d+\alpha)/2}\right) (Dt/a^2 \gg 1).$$

- Large-t behavior is determined by exponent α and dimension d, i.e., excess spreadability decays to zero as a power-law $\frac{1}{t^{(d+\alpha)/2}}$, implying a faster decay as α increases for fixed d and finite α .
- In stealthy limit ($lpha o \infty$), predicted infinitely-fast decay rate implies exponentially fast decay.

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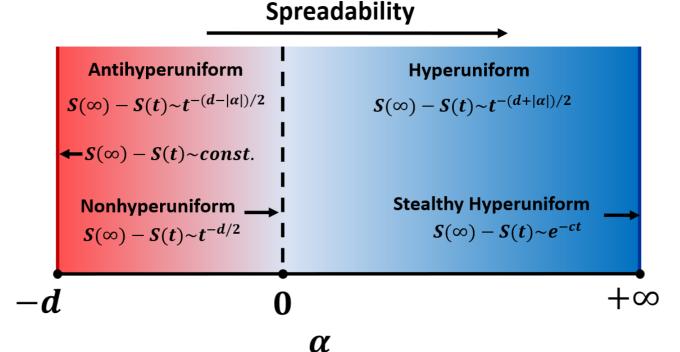
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What stealthy structure maximizes spreadability?

CONCLUSIONS

- There are a plethora of statistical descriptors of microstructures that arise in rigorous relations for various effective properties, leading to accurate estimates.
- Cross-property relations link seemingly different effective properties.
- Hyperuniformity concept provides a unified means of categorizing and characterizing crystals, quasicrystals and special correlated disordered systems according to their large-scale behaviors.
- Disordered hyperuniform materials are ideal states of amorphous matter that often are endowed with novel bulk properties that we are only beginning to discover.
- Spreadability of diffusion information $\mathcal{S}(t)$ across timescales is a powerful tool to dynamically probe and classify all translationally invariant two-phase microstructures across length scales.
- Order metrics have been devised to quantify the degree of order across length scales.
- These general findings have implications for the design of two-phase media with desirable transport, optical and mechanical properties.

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Collaborators

Paul Chaikin (NYU) Weining Man (San Fran. State)

Marian Florescu (Surrey) Murray Skolnick (Princeton)

Yang Jiao (Arizona State) Paul Steinhardt (Princeton)

Jaeuk Kim (Princeton) Frank Stillinger (Princeton)

Michael Klatt (German Aerospace Centre) Ge Zhang (Princeton/Hong Kong)



