Extraordinary disordered hyperuniform multifunctional composites

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Abstract
A variety of performance demands are being placed on material systems, including desirable mechanical, thermal, electrical, optical, acoustic and flow properties. The purpose of the present article is to review the emerging field of disordered hyperuniform composites and their novel multifunctional characteristics. Disordered hyperuniform media are exotic amorphous states of matter that are characterized by an anomalous suppression of large-scale volume-fraction fluctuations compared to those in "garden-variety" disordered materials. Such unusual composites can have advantages over their periodic counterparts, such as unique or nearly optimal, direction-independent physical properties and robustness against defects. It will be shown that disordered hyperuniform composites and porous media can be endowed with a broad spectrum of extraordinary physical properties, including photonic, phononic, transport, chemical and mechanical characteristics that are only beginning to be discovered.

Keywords
disordered composites, hyperuniformity, multifunctionality

Introduction
Increasingly, a variety of performance demands are being placed on material systems. In aerospace and space applications these requirements include lightweight component structures that have desirable mechanical, thermal, electrical, optical, acoustic and flow properties. Structural components should be able to carry mechanical loads while having other beneficial performance characteristics. Desirable thermal properties include high thermal conductivity to dissipate heat and thermal expansion characteristics that match the attached components. In the case of porous cellular solids, heat dissipation can be improved by forced convection through the material, but in these instances the fluid permeability of the porous material must be large enough to minimize power requirements for convection. Desirable optical and acoustic properties include materials that can control the propagation of light and sound waves through them. It is difficult to find single homogeneous materials that possess these multifunctional characteristics.

By contrast, composite materials are ideally suited to achieve multifunctionality, since the best features of different materials can be combined to form a new material that has a broad spectrum of desired properties.1–5 These materials may simultaneously perform as ultralight load-bearing structures, enable thermal and/or electrical management, ameliorate crash or blast damage, and have desirable optical and acoustic characteristics. A general goal is the design of composite materials with $N$ different effective properties or responses, which we denote by $K_i^{(1)}, K_i^{(2)}, \ldots, K_i^{(N)}$, given the individual properties of the phases. In principle, one desires to know the region (set) in the multidimensional space of effective properties in which all composites must lie (see Figure 1 for a two-dimensional (2D) illustration). The size and shape of this region depends on the prescribed phase properties as well as how much microstructural information is specified. For example, the set of composites with unspecified volume fractions is clearly larger than the set in which the the volume fractions are specified.

The determination of the allowable region is generally a highly complex problem. Cross-property bounds can aid to identify the boundary of the allowable region and numerical topology optimization methods can then be used to find specific microstructures that lie on the boundary, which are extremal solutions. These methods often bias the solutions to be periodic structures with high

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crystallographic symmetries. As we will see below, it can be very beneficial to constrain the optimal solution set to microstructures possessing “correlated disorder”,22 which can have advantages over periodic media, especially an exotic type of disorder within the so-called hyperuniformity class.23,24 The purpose of the present article is to review the emerging field of disordered hyperuniform composites and their novel multifunctional characteristics.

The hyperuniformity concept was introduced and studied nearly two decades ago in the context of many-particle systems.23 Hyperuniform systems are characterized by an anomalous suppression of large-scale density fluctuations compared to “garden-variety” disordered systems. The hyperuniformity concept generalizes the traditional notion of long-range order in many-particle systems to not only include all perfect crystals and perfect quasicrystals, but also exotic amorphous states of matter. Disordered hyperuniform materials can have advantages over crystalline ones, such as unique or nearly optimal, direction-independent physical properties and robustness against defects.16,22,25–42

The hyperuniformity concept was generalized to two-phase heterogeneous media in \( \mathbb{R}^d \),24,43 which include composites, cellular solids and porous media. A two-phase medium in \( \mathbb{R}^d \) is hyperuniform if its local volume-fraction variance \( \sigma_V^2(R) \) associated with a spherical observation window of radius \( R \) decays in the large-\( R \) limit faster than the inverse of the window volume, i.e., \( 1/R^d \); (see Structural characterization of disordered hyperuniform composites for mathematical details). This behavior is to be contrasted with those of “typical” disordered two-phase media for which the variance decays like the inverse of the window volume (see Figure 2).

In this article, we review progress that has been made to generate and characterize multifunctional disordered two-phase composites and porous media. In Structural characterization of disordered hyperuniform composites, we collect basic definitions and background on hyperuniform and nonhyperuniform two-phase media. In Forward approaches to generating disordered hyperuniform two-phase media and Inverse approaches to generating disordered hyperuniform two-phase media, we review developments in generating disordered hyperuniform two-phase media using forward and inverse approaches, respectively. In Order metrics for disordered hyperuniform two-phase media, we describe order metrics that enable a rank ordering of disordered hyperuniform two-phase media. In Novel multifunctional disordered hyperuniform composites and porous media, we review the current knowledge about the extraordinary multifunctional characteristics of disordered hyperuniform composites and porous media. Finally, In Conclusions and outlook, we make concluding remarks and

**Figure 1.** Schematic illustrating the allowable region in which all composites with specified phase properties must lie for the case of two different effective properties, \( K^{(1)} \) and \( K^{(2)} \), as adapted from Ref. 19. Importantly, this allowable region depends on the type of microstructural information that is specified.

**Figure 2.** Schematics indicating a circular observation window of radius \( R \) in two dimensions and its centroid \( x_0 \) for a “typical” disordered nonhyperuniform (a), periodic (b), and disordered hyperuniform (c) media. In each of these examples, the phase volume fraction within the window will fluctuate as the window position varies. Whereas the local variance \( \sigma_V^2(R) \) for the nonhyperuniform medium decays like \( 1/R^2 \) for large \( R \), it decays like \( 1/R^3 \) in both the periodic and disordered hyperuniform examples. In space dimension \( d \) and for large \( R \), \( \sigma_V^2(R) \) scales like \( 1/R^d \) and \( 1/R^{d+1} \) for nonhyperuniform and hyperuniform media, respectively.
 Structural characterization of disordered hyperuniform composites

For two-phase heterogeneous media in $d$-dimensional Euclidean space $\mathbb{R}^d$, hyperuniformity can be defined by the following infinite-wavelength condition on the spectral density $\tilde{\chi}_V(k)$, i.e.

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

where $k$ is the wavevector. The spectral density $\tilde{\chi}_V(k)$ is the Fourier transform of the autocovariance function $\chi_V(r) = S^{(2)}_i(r) - \phi_i^2$, where $\phi_i$ is the volume fraction of phase $i$, and $S^{(2)}_i(r)$ gives the probability of finding two points separated by $r$ in phase $i$ at the same time. The two-point descriptor in Fourier space can be easily obtained for general microstructures either theoretically, computationally, or via scattering experiments. The distinctions between the spectral densities for examples of 2D hyperuniform and nonhyperuniform media can be vividly seen in the top panel of Figure 3.

Hyperuniformity of two-phase media can be also defined in terms of the local volume-fraction variance $\sigma^2_V(R)$ associated with a spherical window of radius $R$. Specifically, a hyperuniform two-phase system is one in which $\sigma^2_V(R)$ decays faster than $R^{-d}$ in the large-$R$ regime, i.e.

$$\lim_{R \to \infty} R^d \sigma^2_V(R) = 0$$

In addition to having a direct-space representation, the local variance $\sigma^2_V(R)$ has the following Fourier representation in terms of the spectral density $\tilde{\chi}_V(k)$:

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

where $v_1(R) = \pi^{d/2} R^d / \Gamma(d/2 + 1)$ is the volume of a $d$-dimensional sphere of radius $R$, $\Gamma(x)$ is the gamma function

$$\tilde{\alpha}_2(k;R) = 2^d \pi^{d/2} \Gamma \left( \frac{d}{2} + 1 \right) \frac{J_{d/2}(kR)^2}{k^d}$$

is the Fourier transform of the scaled intersection volume of two spheres of radius $R$ whose centers are separated by a distance $r$, and $k \equiv |k|$ is the wavenumber. The bottom panel of Figure 3 depicts the local variances corresponding to the spectral densities shown in the top panel.

As in the case of hyperuniform point configurations, there are three different scaling regimes (classes) that describe the associated large-$R$ behaviors of the volume-fraction variance when the spectral density goes to zero with the following power-law scaling:

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

namely,

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

where the exponent $\alpha$ is a positive constant. Classes I and III are the strongest and weakest forms of hyperuniformity, respectively. Stealthy hyperuniform media are also of class I.
and are defined to be those that possess zero-scattering intensity for a set of wavevectors around the origin,\textsuperscript{46} i.e.
\[
\tilde{\chi}_V(\mathbf{k}) = 0 \quad \text{for } 0 \leq |\mathbf{k}| \leq K \tag{7}
\]

Examples of such media are periodic packings of spheres as well as unusual disordered sphere packings derived from stealthy point patterns.\textsuperscript{30,46}

By contrast, for any nonhyperuniform two-phase system, it is straightforward to show, using a similar analysis as for point configurations,\textsuperscript{47} that the local variance has the following large-$R$ scaling behaviors
\[
\sigma^2_V(R) \sim \begin{cases} 
R^{-d}, & a = 0 \quad \text{(typical nonhyperuniform)} \\
R^{-(d+a)}, & -d < a < 0 \quad \text{(anti-hyperuniform)} 
\end{cases} \tag{8}
\]

For a “typical” nonhyperuniform system, $\tilde{\chi}_V(0)$ is bounded.\textsuperscript{24} In anti-hyperuniform systems, $\tilde{\chi}_V(0)$ is unbounded, i.e.
\[
\lim_{|\mathbf{k}| \to 0} \tilde{\chi}_V(\mathbf{k}) = +\infty \tag{9}
\]

and hence are diametrically opposite to hyperuniform systems. Anti-hyperuniform systems include systems at thermal critical points (e.g. liquid-vapor and magnetic critical points),\textsuperscript{48,49} fractals,\textsuperscript{50} disordered non-fractals,\textsuperscript{51} and certain substitution tilings.\textsuperscript{52}

### Forward approaches to generating disordered hyperuniform two-phase media

Here we describe “forward” (direct) approaches that have yielded disordered hyperuniform particulate media. Jammed as well as unjammed states are briefly reviewed.

Torquato and Stillinger\textsuperscript{23} suggested that certain defect-free strictly jammed (i.e. mechanically stable) packings of identical spheres are hyperuniform. Specifically, they conjectured that any strictly jammed saturated infinite packing of identical spheres is hyperuniform. A saturated packing of hard spheres is one in which there is no space available to add another sphere. This conjecture was confirmed by Donev et al.\textsuperscript{54} via a numerically generated maximally random jammed (MRJ) packing\textsuperscript{55,56} of $10^6$ hard spheres in three dimensions. Subsequently, the hyperuniformity of other MRJ hard-particle packings, including nonspherical particle shapes, was established across dimensions.\textsuperscript{57–68} Jammed athermal soft-sphere models of granular media,\textsuperscript{69,70} jammed thermal colloidal packings,\textsuperscript{71,72} and jammed bidisperse emulsions\textsuperscript{73} were also shown to be effectively hyperuniform. The singular transport and electromagnetic properties of MRJ packings of spheres\textsuperscript{74} and superballs\textsuperscript{68} have also been investigated.

Periodically driven colloidal suspensions were observed to have a phase transition in terms of the reversibility of the dynamics one decade ago.\textsuperscript{75} Random organization models capture the salient physics of how driven systems can self-organize.\textsuperscript{76} A subsequent study of random organization models of monodisperse (i.e. identical) spherical particles have shown that a hyperuniform state is achievable when a granular system goes through an absorbing phase transition to a critical state.\textsuperscript{77} Many variants of such models and systems have been studied numerically.\textsuperscript{78–82} To what extent is hyperuniformity is preserved when the model is generalized to particles with a size distribution and/or nonspherical shapes? This question was probed in a recent study\textsuperscript{53} by examining disks with a size distribution, needle-like shapes and squares in two dimensions and it was demonstrated that their critical states are hyperuniform as two-phase media (see Figure 4). These results suggest that general particle systems subject to random organization can be a robust way to fabricate a wide class of hyperuniform states of matter by tuning the structures via different particle-size and -shape distributions. This tunability
capacity in turn potentially enables the creation of multifunctional hyperuniform materials with desirable optical, transport, and mechanical properties.

While there has been growing interest in disordered hyperuniform materials states, an obstacle has been an inability to produce large samples that are perfectly hyperuniform due to practical limitations of conventional numerical and experimental methods. To overcome these limitations, a general theoretical methodology has been developed to construct perfectly hyperuniform packings in \( d \)-dimensional Euclidean space \( \mathbb{R}^d \). Specifically, beginning with an initial general tessellation of space by disjoint cells that meets a “bounded-cell” condition, hard particles are placed inside each cell such that the local-cell particle packing fractions are identical to the global packing fraction; see Figure 5. It was proved that the constructed packings with a polydispersity in size in \( \mathbb{R}^d \) are perfectly hyperuniform of class I in the infinite-sample-size limit, even though the initial point configuration that underlies the Voronoi tessellation is nonhyperuniform. Implementing this methodology in sphere tessellations of space (requiring spheres down to the infinitesimally small), establishes the hyperuniformity of the classical Hashin-Shtrikman multiscale coated-spheres structures, which are known to be two-phase microstructures that possess optimal effective transport and elastic properties. Figure 6 shows portions of 2D and 3D hyperuniform polydisperse packings that were converted from the corresponding Voronoi tessellations of nonhyperuniform random sequential addition (RSA) packings. These computationally-designed microstructures can be fabricated via either photolithographic and 3D-printing techniques.

Inverse approaches to generating disordered hyperuniform two-phase media

Here, we describe inverse optimization techniques that enable one to design microstructures with targeted spectral densities. These procedures include the capacity to tune the value of the power-law exponent \( \alpha > 0 \), defined by relation (5), for nonstealthy hyperuniform media as well as to design stealthy hyperuniform media, defined by relation (7).

The Yeong-Torquato stochastic optimization procedure\(^{92,93}\) is a popular algorithm that has been employed to construct or reconstruct digitized multi-phase media from a prescribed set of different correlation functions in physical (direct) space. A fictitious “energy” is defined to be a sum of squared differences between the target and simulated correlation function. The Yeong-Torquato procedure treats the construction or reconstruction task as an energy-minimization problem that it solves via simulated annealing. The Yeong-Torquato procedure was generalized to construct disordered hyperuniform materials with desirable effective macroscopic properties but from targeted structural information in Fourier (reciprocal) space, namely, the spectral density \( \tilde{\chi}_V(k) \). Specifically, the fictitious “energy” \( E \) of the system in \( d \)-dimensional Euclidean space \( \mathbb{R}^d \) is defined as the following sum over wavevectors

\[
E = \sum_k \left[ \frac{\tilde{\chi}_V(k)}{I^d} - \frac{\tilde{\chi}_{V,0}(k)}{I^d} \right]^2
\]

where the sum is over discrete wave vectors \( k \), \( \tilde{\chi}_V(k) \) and \( \tilde{\chi}_{V,0}(k) \) are the spectral densities of the target and (re) constructed microstructures, respectively, \( d \) is the space dimension, and \( I \) is the relevant characteristic length of the system used to scale the spectral densities such that they are dimensionless. As in the standard Yeong-Torquato procedure,\(^{92,93}\) the simulated-annealing method is used to minimize the energy (10). It was demonstrated that one can design nonstealthy hyperuniform media and stealthy hyperuniform media with this Fourier-based inverse technique.\(^{35}\) Such in-silico designed microstructures can be readily realized by 3D printing and lithographic technologies.\(^{89}\)

Figure 7 shows designed realizations of digitized nonstealthy hyperuniform media with prescribed values of the power-law exponent \( \alpha > 0 \), defined by relation (5), at different values of the phase volume fraction \( \phi \). It is seen that these designed materials possess a variety of morphologies: as \( \phi \) increases for fixed \( \alpha \), the microstructures transition from particulate media consisting of isolated “particles” to labyrinth-like microstructures. Moreover, as \( \alpha \) increases for fixed \( \phi \), short-range order increases in these hyperuniform materials.

The “collective-coordinate” optimization procedure represents a powerful reciprocal-space-based approach to generate disordered stealthy hyperuniform point configurations as well as nonstealthy hyperuniform point configurations in \( d \)-dimensional Euclidean space \( \mathbb{R}^d \). In the case of the former, their degree of the stealthiness can be tuned by varying a dimensionless parameter \( \chi \), which measures the relative number of independently constrained degrees of freedom for wavenumbers up to the cut-off value \( K \). For the range \( 0 < \chi < 1/2 \), the stealthy states are disordered, and degree of short-range order increases as \( \chi \) approaches 1/2.\(^{106}\) Such stealthy point patterns can be decorated by nonoverlapping spheres, enabling the generation of non-digitized stealthy sphere packings, which have been used to model disordered two-phase composites that are both stealthy and hyperuniform, as defined by relation (7). As we will see in Novel multifunctional disordered hyperuniform composites and porous media, stealthy hyperuniform composites are endowed with novel multifunctional characteristics.
Figure 5. (a) A Voronoi tesselation of a nonhyperuniform disordered point configuration. (b) The disordered hyperuniform packing of spheres with a size distribution that results by adding particles in the Voronoi tessellation while ensuring that the local cell packing fraction is equal to global packing fraction. (c) A tessellation of space by spheres. (d) The Hashin-Shtrikman composite sphere assemblage that results by adding particles in the sphere tessellation while ensuring that the local cell packing fraction is equal to global packing fraction. These images are adapted from those in Ref. 83.

Figure 6. (a) A portion of a hyperuniform disk packing that was converted from a 2D RSA packing with the packing fraction $\phi_{\text{init}} = 0.41025$. (b) A portion of a hyperuniform sphere packing that was converted from a 3D saturated RSA packing with the packing fraction $\phi_{\text{init}} = 0.288$. This figure is adapted from Ref. 83.
Order metrics for disordered hyperuniform two-phase media

An outstanding open problem is the determination of appropriate “order metrics” to characterize the degree of large-scale order of both hyperuniform and nonhyperuniform media. This task is a highly challenging due to the infinite variety of possible two-phase microstructures (geometries and topologies). To begin such a program, the local variance $\sigma^2_V(R)$ was recently studied for a certain subset of class I hyperuniform media, including 2D periodic cellular networks as well as 2D periodic and disordered/irregular packings, some of which maximize their effective transport and elastic properties. In particular, Kim and Torquato evaluated the local variance $\sigma^2_V(R)$ as a function of the window radius $R$. They also computed the hyperuniformity order metric $B_Y$, i.e., the implied coefficient multiplying $R^{-(d+1)}$ in (6), for all of these class I 2D models to rank them according to their degree of order at a fixed volume fraction. The smaller is the value of $B_Y$, the more ordered is the microstructure with respect to large-scale volume-fraction fluctuations. Among the cellular networks considered, the honeycomb networks have the minimal values of the hyperuniformity order metrics $B_Y$ across all volume fractions. Among all structures studied there, triangular-lattice packings of circular disks have the minimal values of the order metric for almost all volume fractions.

It is desired to formulate order metrics to characterize the degree of order/disorder of the microstructures of two-phase media in $d$-dimensional Euclidean space $\mathbb{R}^d$ across length scales. It has recently been proposed that the local volume-fraction variance $\sigma^2_V(R)$ be used as an order metric for disordered and ordered two-phase media across all length scales by tracking it as a function of $R$. The local volume-fraction variance $\sigma^2_V(R)$ as a function of $R$ was determined for 22 different models across the first three space dimensions, including both hyperuniform and nonhyperuniform systems with varying degrees of short- and long-range order. It was found that the local volume-fraction variance as well as the asymptotic coefficients and integral measures derived from it provide reasonably robust and sensitive order metrics to categorize disordered and ordered two-phase media across all length scales. Such order metrics could be employed to accelerate the discovery of novel heterogeneous materials by tailoring their degree of order/disorder.

**Figure 7.** Realizations of disordered hyperuniform two-phase materials for different values of the volume fraction $\phi$ and the positive exponent $\alpha$, defined by the spectral-density scaling law (5). This figure is adapted from Ref. 35.
Novel multifunctional disordered hyperuniform composites and porous media

By mapping relatively large 2D disordered stealthy hyperuniform point configurations, obtained via the collective-coordinate optimization procedure, to certain 2D trivalent dielectric networks via a Delaunay centroidal tessellation, what was thought to be impossible at the time became possible. Specifically, the first disordered network solids to have large complete (both polarizations and blocking all directions) photonic band gaps comparable in size to those in photonic crystals were identified, but with the additional advantage of perfect isotropy. The computational designs consist of trivalent networks of cell walls with circular cylinders at the nodes. The band structure was computed as a function of the degree of stealthiness \( \chi \) (left panel of Figure 8) and the case of \( \chi \) nearly equal to 0.5 (with an accompanying substantial degree of short-range order) leads to the maximal complete band-gap size in disordered hyperuniform dielectric networks. This numerical investigation enabled the design and fabrication of disordered cellular solids with the predicted photonic band-gap characteristics for the microwave regime (right panel of Figure 8), enabling unprecedented free-form waveguide geometries unhindered by crystallinity and anisotropy, and robust to defects. Subsequently, stealthy hyperuniform materials were shown to have novel electromagnetic and elastic wave propagation characteristics, including transparency to long-wavelength radiation, tunable diffusive and localization regimes, enhanced absorption of waves, and singular phononic band gaps.

The effective thermal or electrical conductivities and elastic moduli of various 2D ordered and disordered hyperuniform cellular networks were studied. The multifunctionality of a class of such low-density networks was established by demonstrating that they maximize or virtually maximize the effective conductivities and elastic moduli of certain 2D trivalent dielectric networks via a Delaunay centroidal tessellation.

Figure 8. (a) Band structure for stealthy hyperuniform networks as a function of \( \chi \), as predicted from the computational study in Ref. 25. From left to right, \( \chi = 0.1, 0.2, 0.3, 0.4 \) and 0.5. The relative band-gap size, measured by \( \Delta \omega / \omega \), takes on the largest value of 10.26\% for the rightmost case of \( \chi = 0.5 \). (b) 3D fabrication of the computationally-designed maximal band-gap structure looking down from the top, as adapted from Ref. 110. The solid phase is aluminum oxide.

Figure 9. (a) 2D disordered hyperuniform trivalent network, as adapted from Ref. 115. (b) 3D disordered hyperuniform tetrahedrally-coordinated network, as adapted from Ref. 116.
moduli. This was accomplished by using the machinery of homogenization theory, including optimal bounds and cross-property bounds, and statistical mechanics. It was rigorously proved that anisotropic networks consisting of sets of intersecting parallel channels in the low-density limit, ordered or disordered, possess optimal effective conductivity tensors. For a variety of different disordered networks, it was shown that when short-range and long-range order increases, there is an increase in both the effective conductivity and elastic moduli of the network. Moreover, it was demonstrated that the effective conductivity and elastic moduli of various disordered networks (derived from disordered “stealthy” hyperuniform point patterns), such as the one shown in the left panel of Figure 9, possess virtually optimal values. Interestingly, point patterns, such as the one shown in the left panel of the fluid permeability associated with slow viscous flow through the channels as well as the mean survival time associated with diffusion-controlled reactions in the channels. 3D disordered hyperuniform networks, such as the one shown in right panel of Figure 9, have been shown to have sizable photonic band gaps. In summary, 2D and 3D disordered hyperuniform low-weight cellular networks are multifunctional with respect to transport (e.g. heat dissipation and fluid transport), mechanical and electromagnetic properties, which can be readily fabricated using 2D lithographic and 3D printing technologies.

The theoretical problem of estimating the effective properties of multiphase composite media is an outstanding one and dates back to work by some of the luminaries of science, including Maxwell, Lord Rayleigh, and Einstein. The preponderance of previous theoretical studies have focused on the determination of static effective properties (e.g. dielectric constant, elastic moduli and fluid permeability) using a variety of methods, including approximation schemes, bounding techniques, and exact series-expansion procedures. Much less is known about the theoretical prediction of the effective dynamic dielectric constant tensor $\varepsilon_d(k)$, where $k$ is wavevector of the incident radiation. The strong-contrast formalism has recently been used to derive exact nonlocal expansions for $\varepsilon_d(k)$ 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., $0 \leq |k|/\ell \leq 1$ (where $\ell$ is a characteristic heterogeneity length scale). Due to the fast-convergence properties of such expansions, their lower-order truncations yield accurate closed-form approximate formulas for $\varepsilon_d(k)$ that depend on the spectral density $\tilde{\chi}_r(k)$. It was shown that disordered stealthy hyperuniform particulate composites exhibit novel wave characteristics, including the capacity to act as low-pass filters that transmit waves “isotropically” up to a selected wavenumber or refractive indices that abruptly change over a narrow range of wavenumbers. The aforementioned nonlocal formulas can now be used to accelerate the discovery of novel electromagnetic composites by appropriate tailoring of the spectral densities.

Cross-property relations for two-phase composite media were recently obtained that link effective elastic and electromagnetic wave characteristics to one another, including effective wave speeds and attenuation coefficients. This was achieved by deriving accurate formulas for the effective elastodynamic properties as well as effective electromagnetic properties, each of which depend on the microstructure via the spectral density. Such formulas enable one to explore the wave characteristics of a broad class of disordered microstructures, including exotic disordered hyperuniform varieties. It was specifically demonstrated that disordered stealthy hyperuniform/nonhyperuniform microstructures exhibit novel elastic wave characteristics that have the potential for future applications, e.g. narrow-band or narrow-band-pass filters that absorb or transmit elastic waves isotropically for a narrow spectrum of frequencies, respectively. These cross-property relations for effective electromagnetic and elastic wave characteristics can be applied to design multifunctional composites (Figure 10), such as exterior components of spacecrafts or building materials that require both excellent stiffness and electromagnetic absorption, and heat-sinks for CPUs that have to efficiently emit thermal radiation and suppress mechanical vibrations, and nondestructive evaluation of the mechanical strength of materials from the effective dielectric response.

The effective transport characteristics of fluid-saturated porous media have been studied using certain rigorous microstructure-property relations. Of particular interest were the predictions of the formation factor $F$, mean survival time $\tau$, principal viscous relaxation time $T_1$, principal viscous relaxation time $\Theta_1$, and fluid permeability $k$ for hyperuniform and nonhyperuniform models of porous media. Among other results, a Fourier representation of a classic rigorous upper bound on the fluid permeability was derived that depends on the spectral density $\tilde{\chi}_r(k)$ to infer how the permeabilities of hyperuniform porous media perform relative to those of nonhyperuniform ones; see Figure 11. It was found that the velocity fields in nonhyperuniform porous media are generally much more localized over the pore space compared to those in their hyperuniform counterparts, which has certain implications for their permeabilities. Rigorous bounds on the transport properties $F$, $\tau$, $T_1$, and $\Theta_1$ suggest a new approximate formula for the fluid permeability that provides reasonably accurate permeability predictions of a certain class of hyperuniform and nonhyperuniform porous media. These comparative studies shed new light on the microstructural characteristics, such as pore-size statistics, in determining the
transport properties of general porous media. In a more recent study, the second moment of the pore-size probability density function was shown to be correlated with the critical pore radius, which contains crucial connectivity information about the pore space. All of these findings have important implications for the design of porous materials with desirable transport properties.

A new dynamic probe of the microstructure of two-phase media has been introduced called the spreadability $S(t)$, which is a measure of the spreadability of diffusion information as a function of time $t$ in any Euclidean space dimension $d$. It is assumed that a solute at $t=0$ is uniformly distributed throughout phase 2 with volume fraction $f_2$, and completely absent from phase 1 with volume fraction $f_1$, and each phase has same diffusion coefficient $D$. The spreadability is the fraction of the total amount of solute present that has diffused into phase 1 at time $t$. In particular, a three-dimensional formula due to Prager was generalized to any dimension in direct space and its Fourier representation was derived. The latter is an

Figure 10. Schematics illustrating elastic and electromagnetic waves at two different wavenumbers (a) $k_1$ and (b) $k_2$, incident to, inside of and transmitted from a two-phase heterogeneous material (a large ellipse) consisting of a matrix phase (shown in yellow) and a dispersed phase (shown in cyan). Parallel lines and sinusoidal curves represent elastic and electromagnetic waves, respectively. (a) For an elastic wave with a wavenumber $k_1$ while the wavefronts inside this material experience microscopic disturbances, they effectively behave like a plane wave inside a homogeneous material with an effective wavenumber $(k_e)$ and effective elastic moduli $K_e$ and $G_e$. Analogously, for an electromagnetic wave, this material behaves like a homogeneous material with an effective dielectric constant $\varepsilon_e$. For instance, both elastic and electromagnetic waves are attenuated due to scattering if this composite has a non-zero scattering intensity at $k_1$. (b) For waves (red) of a wavenumber $k_2$, this composite can be effectively transparent, if it has a zero-scattering intensity at $k_2$. This figure is adapted from Ref. 16.

Figure 11. Images of the void space after applying a “dilation” operation to three different sphere packings, as adapted from Ref. 130. (a) A nonhyperuniform equilibrium packing. (b) A hyperuniform maximally random jammed packing. (c) A disordered stealthy packing.

Figure 12. Excess spreadabilities versus dimensionless time $Dt/a^2$ for antihyperuniform media (top curve), Debye random medium (middle curve), and disordered hyperuniform media (bottom curve) for $d=3$ and $\phi_2 = 0.5$, as adapted from Ref. 132. The long-time inverse power-law scalings of $S(\infty) - S(t)$ for each of these models is indicated. Here $a$ is a characteristic length scale for each model, as defined in Ref. 132.
exact integral relation for the spreadability $S(t)$ that depends only on the spectral density $\tilde{\chi}_V(k)$:

$$S(\infty) - S(t) = \frac{1}{(2\pi)^d} \int_{\mathbb{R}^d} \tilde{\chi}_V(k) \exp[-k^2 D t] dk \geq 0 \quad (11)$$

where $S(\infty) = \phi_1$. Importantly, the short-, intermediate- and long-time behaviors of $S(t)$ contain crucial small-, intermediate- and large-scale structural characteristics. For hyperuniform media, it was shown that the “excess” spreadability, $S(\infty) - S(t)$, decays to its long-time behavior exponentially faster than that of any nonhyperuniform medium, the “slowest” being antihyperuniform media, as illustrated in Figure 12. It was also shown that there is a remarkable link between the spreadability and NMR pulsed field gradient spin-echo amplitude and diffusion MRI.\(^{132}\) Elsewhere, this new theoretical/experimental tool was applied to characterize many different models and a porous-medium sample.\(^{68,134}\)

**Conclusions and outlook**

We have seen that the exotic hybrid crystal-liquid structural attributes of disordered hyperuniform composites can be endowed with an array of extraordinary physical properties, including photonic, phononic, transport and mechanical characteristics that are only beginning to be discovered. Disordered hyperuniform media can have advantages over their periodic counterparts, such as unique or nearly optimal, direction-independent physical properties and robustness against defects. The field of hyperuniformity is still in its infancy, though, and a deeper fundamental understanding of these unusual states of matter is required in order to realize their full potential for next-generation materials. Future challenges include the further development of forward and inverse computational approaches to generate disordered hyperuniform structures, formulation of improved order metrics to rank order them, and identifying their desirable multifunctional characteristics. These computational designs can subsequently be combined with the 2D lithographic fabrication techniques\(^{91}\) and 3D additive manufacturing techniques\(^{88–90}\) to accelerate the discovery of novel multifunctional hyperuniform two-phase materials.

Cross-property “maps” have recently been introduced to connect combinations of pairs of effective static transport and elastic properties of general particulate media via analytical structure-property formulas.\(^{135}\) Cross-property maps and their extensions will facilitate the rational design of composites with different desirable multifunctional characteristics. In future work, it would be valuable to formulate cross-property maps for the various physical properties described in Novel multifunctional disordered hyperuniform composites and porous media using the corresponding analytical estimates of these properties in order to aid in the multifunctional design of disordered hyperuniform composites.

To complement rigorous approaches to estimate the macroscopic properties of heterogeneous media from the microstructure, data-driven methodologies to establish structure-property relationships are increasingly being employed.\(^{89,136–138}\) The rapid increase in computational resources facilitates the calculation of effective properties for very large data sets (thousands or more) of different microstructures, including those obtained experimentally via 2D and 3D high-resolution imaging techniques.\(^{3,139–142}\) As a result, it has become manageable to generate large numbers of realistic virtual microstructures, and using those to perform exploratory computational screening of structure–property relationships. The application of machine-learning and other data-driven approaches for the discovery of multifunctional disordered hyperuniform composites has yet to be undertaken and hence is a promising avenue for future research.

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