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Extraordinary Multifunctional Disordered Composites

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14. ABSTRACT Disordered hyperuniform many-body systems are new, exotic states of matter that lie between a crystal and a liquid: they are like perfect crystals in the way they suppress large-scale density fluctuations and yet are statistically isotropic like liquids. Now there is a realization that such states play vital roles in a number of fundamental and applied problems: glass formation, jamming, rigidity, band-gap structures, biology, localization of waves and excitations, selforganization, fluid dynamics, quantum systems, and pure mathematics. This activity strongly indicates that this is an emerging field with potentially explosive growth. The overall purpose of this project is to establish the fundamental understanding and capabilities required to design disordered hyperuniform composites for utilization in aerospace materials, devices and microsystems as multifunctional structures. We derived and applied "cross-property maps that connect combinations of pairs of the aforementioned effective transport and elastic properties to one another for a wide class of dispersions in which the inclusions can have different shapes and sizes and are spatially distributed in a matrix with varying degrees of order/disorder. In another study, we have showed that generalizations of "random organization models" of vibrated granular media are disordered and hyperuniform. Elsewhere, we explored the effective thermal or electrical conductivities and elastic moduli of various 2D ordered and disordered hyperuniform cellular networks. We established the multifunctionality of a class of such low-density networks by demonstrating that they maximize or virtually maximize the effective conductivities and elastic moduli. Finally, we established the first cross-property relations for two-phase composite media 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 that depend on the microstructure via the spectral density and utilizing previous analogous results for effective electromagnetic properties. Many of the disordered hyperuniform composites that we have studied theoretically and computationally can be readily fabricated using 3D printing and lithographic technologies.					
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Final Technical Report

EXTRAORDINARY MULTIFUNCTIONAL DISORDERED COMPOSITES

Grant No. FA9550-18-1-0514

August 1, 2018 - July 31, 2023

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PREPARED FOR THE AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

1 Objectives

Disordered hyperuniform many-body systems are new, exotic states of matter that lie between a crystal and a liquid: they are like perfect crystals in the way they suppress large-scale density fluctuations and yet are statistically isotropic like liquids. Now there is a realization that such states play vital roles in a number of fundamental and applied problems: glass formation, jamming, rigidity, band-gap structures, biology, localization of waves and excitations, self-organization, fluid dynamics, quantum systems, and pure mathematics. This activity strongly indicates that this is an emerging field with potentially explosive growth. The overall purpose of this project is to establish the fundamental understanding and capabilities required to design disordered hyperuniform *composites* for utilization in aerospace materials, devices and microsystems as multifunctional structures.

2 Accomplishments

During the entire grant period, we published a total of 17 peer-reviewed journal articles. The P.I. gave 25 invited lectures on his AFOSR-related research work and was named to an endowed chair.

In a paper published in Physical Review Materials [1], we employed microstructure-dependent analytical expressions for the effective electrical (thermal) conductivity and elastic moduli that accurately model a wide class of three-dimensional isotropic particulate composites (see Fig. 1) for all phase contrasts and volume fractions to derive and apply “cross-property maps”. These maps connect combinations of pairs of the aforementioned effective transport and elastic properties to one another for a wide class of dispersions in which the inclusions can have different shapes and sizes and are spatially distributed in a matrix with varying degrees of order/disorder. We illustrated cross-property maps for a variety of cases (e.g., incompressible or compressible composites with conducting/insulating inclusions that may be rigid or compliant or auxetic) for high inclusion volume fractions. All of the cross-property maps that involve the effective conductivity translate immediately into equivalent results for the effective dielectric constant, magnetic permeability, or diffusion coefficient because they are mathematically analogous. We discussed an example that enables one to design a disordered dispersion with desired values of the effective dielectric constant and effective Young’s modulus. Cross-property maps and their extensions will facilitate the rational design of particulate media with different desirable multifunctional characteristics. Moreover, our investigation has implications for the application of machine-learning and other data-driven approaches for multifunctional materials discovery.

Studies 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. We have investigated to what extent hyperuniformity is preserved when the model is generalized to particles with a size distribution and/or

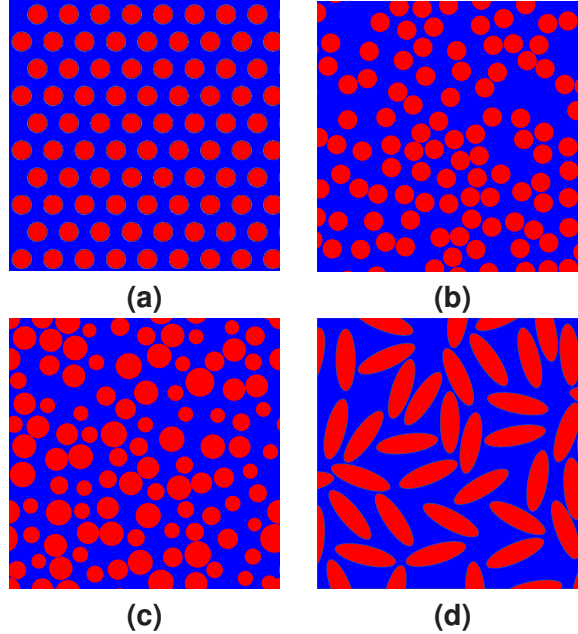


Figure 1: *Select representative examples of the wide class of dispersions of nonoverlapping inclusions in a matrix for which formulas for the effective conductivity, bulk modulus and shear modulus and resulting cross-property relations apply. (a) Ordered dispersion of identical spheres (disks). (b) Disordered dispersion of identical spheres (disks). (c) Disordered dispersion of spheres (disks) of different sizes. (d) Disordered dispersion of identical ellipsoids (ellipses) with random orientations. At a fixed volume fraction and away from close-packed states, ζ_2 is generally smallest for (a), next smallest for (b), larger for (c) and largest for (d). Qualitatively, the same trends apply to the three-point parameter η_2 , which arises in rigorous three-point bounds estimates on the effective shear modulus G_e .*

nonspherical shapes [2]. We began by examining binary disks in two dimensions and demonstrate that their critical states are hyperuniform as two-phase media, but not hyperuniform nor multihyperuniform as point patterns formed by the particle centroids. We further confirmed the generality of our findings by studying particles with a continuous size distribution (see Fig. 2). Finally, to study the effect of rotational degrees of freedom, we extended our model to noncircular particles, namely, hard rectangles with various aspect ratios, including the hard-needle limit (see Fig. 2). Although these systems exhibit only short-range orientational order, hyperuniformity is still preserved. Our analysis reveals that the redistribution of the “mass” of the particles rather than the particle centroids is central to this dynamical process. The consideration of the “active volume fraction” of generalized random organization models may help to resolve which universality class they belong to and hence may lead to a deeper theoretical understanding of absorbing-state models. Our 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 in turn potentially enables the creation of multifunctional hyperuniform materials with desirable optical, transport,

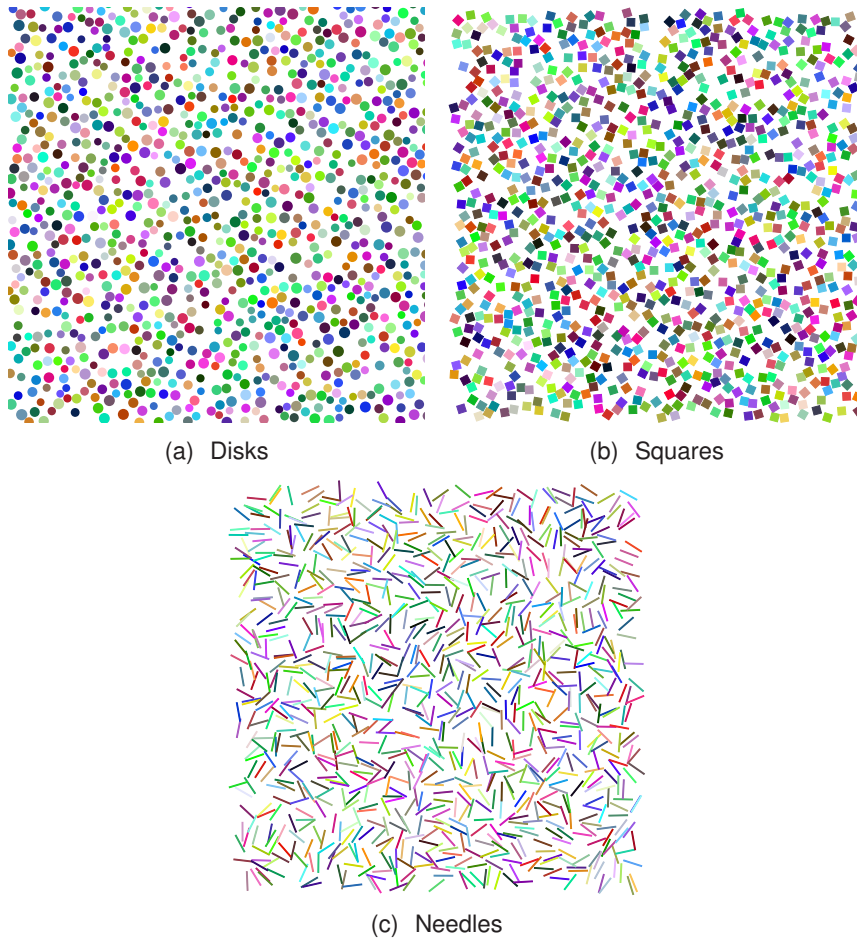


Figure 2: (a) A representative image of a configuration of a 2D disk system with continuous size distribution in an absorbing state. (b) A representative plot of a configuration of a 2D square system in an absorbing state. (c) A representative image of a configuration of a 2D hard-needle system in an absorbing state.

and mechanical properties.

In another investigation [3], we explored the effective thermal or electrical conductivities and elastic moduli of various 2D ordered and disordered hyperuniform cellular networks (see Fig. 3). We established the multifunctionality of a class of such low-density networks by demonstrating that they maximize or virtually maximize the effective conductivities and elastic moduli. This is accomplished using the machinery of homogenization theory, including optimal bounds and cross-property bounds, and statistical mechanics. We 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, we showed 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, we demonstrated

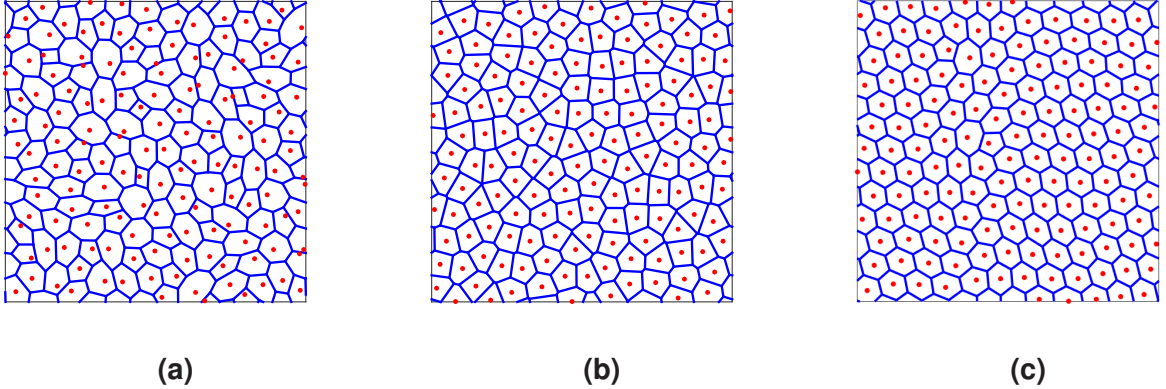


Figure 3: Representative disordered stealthy cellular network structures mapped from various point patterns. (a) Delaunay-centroidal network of stealthy point pattern with $\chi = 0.3$. (b) Voronoi network of stealthy point pattern with $\chi = 0.4$. (c) Delaunay-centroidal network of stealthy point pattern with $\chi = 0.49$.

that the effective conductivity and elastic moduli of various disordered networks derived from disordered “stealthy” hyperuniform point patterns possess virtually optimal values. Interestingly, the optimal networks for conductivity are also optimal for 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. In summary, we have identified ordered and disordered hyperuniform low-weight cellular networks that are multifunctional with respect to transport (e.g., heat dissipation and fluid transport), mechanical and electromagnetic properties, which can be readily fabricated using 3D printing and lithographic technologies.

In a paper published in *Advances in Water Resources* [4], we were concerned with the estimation of the effective transport characteristics of fluid-saturated porous media via rigorous microstructure-property relations. We were particularly interested in predicting the formation factor \mathcal{F} , mean survival time τ , principal NMR (diffusion) relaxation time T_1 , principal viscous relaxation time Θ_1 , and fluid permeability k . To do so, we employed rigorous methods to estimate the fluid permeability and these other transport properties of “hyperuniform” and nonhyperuniform models of porous media from microstructural information. Disordered hyperuniform materials are exotic amorphous states of matter that have attracted great attention in the physical, mathematical and biological science but little is known about their fluid transport characteristics. In carrying out this investigation, we not only drew from ideas and results of the emerging field of hyperuniformity, but from homogenization theory, statistical geometry, differential equations (spectrum of Laplace and Stokes operators), and the covering and quantizer problems of discrete geometry. Among other results, we derive a Fourier representation of a classic rigorous upper bound on the fluid permeability that depends on the spectral density to infer how the permeabilities of hyperuniform porous media perform relative to those of nonhyperuniform ones; see Fig. 4. We 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 implications for their permeabilities. Rigorous bounds on transport properties 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 that determine the transport properties of general porous media. Our findings also have implications for the design of porous materials with desirable transport properties.

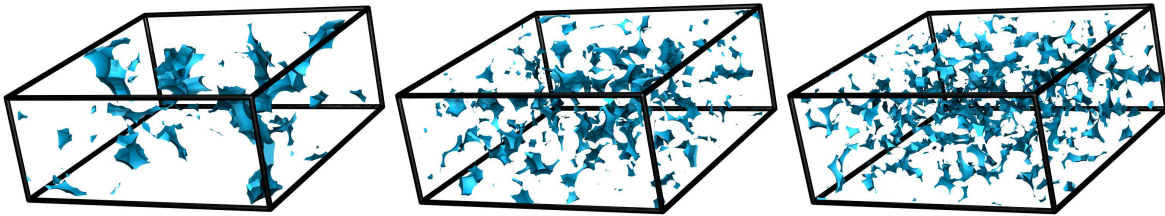


Figure 4: Images of the void space after applying a dilation operation to three different sphere packings: A nonhyperuniform equilibrium packing(left panel), a hyperuniform MRJ packing (middle panel) and a disordered stealthy packing (right panel).

The quantitative structure–property relationships are crucial for the understanding and prediction of the physical properties of complex materials. For fluid flow in porous materials, characterizing the geometry of the pore microstructure facilitates prediction of permeability, a key property that has been extensively studied in material science, geophysics and chemical engineering. In another study [5], we considered the predictability of different structural descriptors via both linear regressions and neural networks. A large data set of 30,000 virtual, porous microstructures of different types, including both granular and continuous solid phases, is created for this end. We computed permeabilities of these structures using the lattice Boltzmann method, and characterized the pore space geometry using one-point correlation functions (porosity, specific surface), two-point surface-surface, surface-void, and void-void correlation functions, as well as the geodesic tortuosity as an implicit descriptor. Then, we study the prediction of the permeability using different combinations of these descriptors. We obtained significant improvements of performance when compared to a Kozeny-Carman regression with only lowest-order descriptors (porosity and specific surface). We found that combining all three two-point correlation functions and tortuosity provides the best prediction of permeability, with the void-void correlation function being the most informative individual descriptor. Moreover, the combination of porosity, specific surface, and geodesic tortuosity was shown to provide very good predictive performance. This shows that higher-order correlation functions are extremely useful for forming a general model for predicting physical properties of complex materials. Additionally, our results suggest that artificial neural networks are superior to the more conventional regression methods for establishing quantitative structure–property relationships. We made the data and code used publicly available to facilitate further development of permeability prediction methods.

Fluid flow through porous media plays a crucial role in many applications and technologies. Our aim in another article published in Physical Review E [6] was to find convenient yet reliable estimates of the fluid permeability based on the structural and topological characteristics of the complex tortuous pore space. Such predictions can facilitate the design of porous media with desirable transport properties. For porous media with a well-connected pore space, a recent study by the P.I. suggested the second moment of the pore-size distribution as a convenient alternative to the often-used critical pore radius, which requires a sophisticated percolation analysis. We determined both descriptors for disordered (hyperuniform and nonhyperuniform) and ordered model microstructures, including maximally random jammed (MRJ) spheres, overlapping spheres, equilibrium hard spheres, quantizer configurations, and lattice packings. Interestingly, we found that the second moment of the pore-size distribution is — to a good approximation — proportional to the critical pore radius. In fact, in contrast to the latter, the former predicts the correct ranking of the permeability for our models. Moreover, we found that the hyperuniform structures, which are characterized by an anomalous suppression of volume-fraction fluctuations, tend to have lower values of the permeability, including MRJ sphere packings, quantizer configurations, and BCC sphere packings.

While the classification of hyperuniform point configurations has received considerable attention, much less is known about the classification of hyperuniform two-phase heterogeneous media, which include composites, porous media, foams, cellular solids, colloidal suspensions, and polymer blends. In a paper published in Physical Review E [7], we began such a program for certain two-dimensional models of hyperuniform two-phase media by ascertaining their local volume-fraction variances and the associated hyperuniformity order metrics B_V . This is a highly challenging task because the geometries and topologies of the phases are generally much richer and more complex than point-configuration arrangements, and one must ascertain a broadly applicable length scale to make key quantities dimensionless. Therefore, we purposely restricted ourselves to a certain class of two-dimensional periodic cellular networks as well as periodic and disordered or irregular packings of circular disks, some of which maximize their effective transport and elastic properties. Among the cellular networks considered, the honeycomb networks have minimal values of the hyperuniformity order metrics B_V across all volume fractions. On the other hand, for all packings of circular disks examined, the triangular-lattice packings have the smallest values of B_V for the possible range of volume fractions. Among all structures studied here, the triangular-lattice packing of circular disks have the minimal values of the order metric for almost all volume fractions. Our study provides a theoretical foundation for the establishment of hyperuniformity order metrics for general two-phase media and a basis to discover new hyperuniform two-phase systems with desirable bulk physical properties by inverse design procedures

Composites are ideally suited to achieve desirable multifunctional effective properties, since the best properties of different materials can be judiciously combined with designed microstructures. *Cross-property relations*, which rigorously link one effective property of a composite to a different effective property of the same composite, can aid in the optimal design of multifunctional composites. We have established the first cross-property relations for two-phase composite media that link effective elastodynamic and electromagnetic wave characteristics to one another,

including effective wave speeds and attenuation coefficients [8]. This was achieved by using the strong-contrast formalism to first derive exact expressions for the effective dynamic dielectric constant tensor [9] as well as the effective dynamic elastic moduli [10] of two-phase composites that apply beyond the quasistatic regime. From these exact relations, we then extracted accurate approximations formulas for the effective properties that depend on the microstructure via the spectral density, which yielded the cross-property relations. Our cross-property formulas enable us to explore the wave characteristics of a broad class of disordered microstructures, including exotic disordered “hyperuniform” varieties, that can have advantages over crystalline ones, such as nearly optimal, direction-independent properties and robustness against defects. We specifically show 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. Our cross-property relations for effective wave characteristics can be applied to design multifunctional composites, 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. Finally, we discussed how to utilize our approximations for engineering composites with prescribed elastic wave characteristics by appropriate tailoring of the spectral densities; see schematic examples in Fig. 5

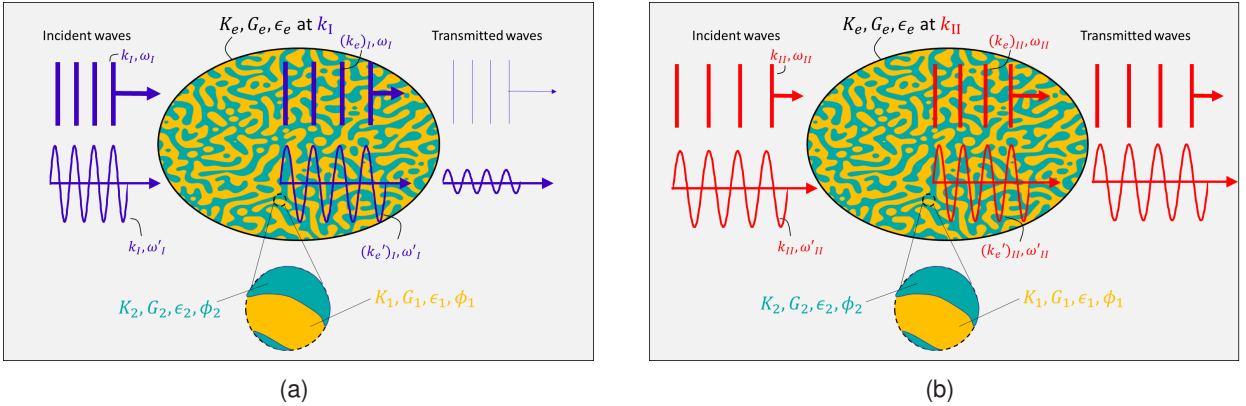


Figure 5: Schematics illustrating elastic and electromagnetic waves at two different wavenumbers (a) k_I and (b) k_{II} incident to, inside of and transmitted from a two-phase heterogeneous material (a large ellipse) consisting of a matrix phase (shown in yellow) and a dispersed phase (shown in cyan). Parallel lines and sinusoidal curves represent elastic and electromagnetic waves, respectively. (a) For an elastic wave with a wavenumber k_I , while the wavefronts inside this material experience microscopic disturbances, they effectively behave like a plane wave inside a homogeneous material with an effective wavenumber $(k_e)_I = \omega_I[(c_e)_L + i(\gamma_e)_L]^{-1}$ 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 ϵ_e . For instance, both elastic and electromagnetic waves are attenuated due to scattering if this composite has a non-zero scattering intensity at k_I . (b) For waves (red) of a wavenumber k_{II} , this composite can be effectively transparent, if it has a zero-scattering intensity at k_{II} .

The hyperuniformity concept provides a unified means to classify all perfect crystals, perfect quasicrystals, and exotic amorphous states of matter according to their capacity to suppress large-scale density fluctuations. While the classification of hyperuniform point configurations has received considerable attention, much less is known about the classification of hyperuniform heterogeneous two-phase media, which include composites, porous media, foams, cellular solids, colloidal suspensions and polymer blends. In a paper published in Physical Review E [11], we began such a program for certain two-dimensional models of hyperuniform two-phase media by ascertaining their local volume-fraction variances $\sigma_V^2(R)$ and the associated hyperuniformity order metrics \overline{B}_V . This is a highly challenging task because the geometries and topologies of the phases are generally much richer and more complex than point-configuration arrangements, and one must ascertain a broadly applicable length scale to make key quantities dimensionless. Therefore, we restricted ourselves to a certain class of 2D periodic cellular networks as well as periodic and disordered/irregular packings of circular disks, some of which maximize their effective transport and elastic properties. Among the cellular networks considered, honeycomb networks were found to have the minimal value of the hyperuniformity order metrics \overline{B}_V across all volume fractions. On the other hand, among all packings of circular disks examined, triangular-lattice packings have the smallest values of \overline{B}_V for the possible range of volume fractions. Among all structures studied here, the triangular-lattice packing of circular disks has the minimal order metric for almost all volume fractions. Our study provides a theoretical foundation for the establishment of hyperuniformity order metrics for general two-phase media and a basis to discover new hyperuniform two-phase systems with desirable bulk physical properties by inverse design procedures.

In a paper published in Physical Review Letters [12], we reported on a new discovery concerning the photonic properties of dielectric networks. Through an extensive series of high-precision numerical computations of the optimal complete photonic band gap (PBG) as a function of dielectric contrast α for a variety of crystal and disordered heterostructures (see Fig. 6), we revealed striking universal behaviors of the *gap sensitivity* $S(\alpha) \equiv d\Delta(\alpha)/d\alpha$, the first derivative of the optimal gap-to-midgap ratio $\Delta(\alpha)$. In particular, for all our crystal networks, $S(\alpha)$ takes a universal form that is well approximated by the analytic formula for a one-dimensional quarter-wave stack. Even more surprisingly, the values of $S(\alpha)$ for our disordered hyperuniform networks converge to 1D quarter-wave stack for sufficiently large α . A deeper understanding of the simplicity of this universal behavior may provide fundamental insights about PBG formation and guidance in the design of novel photonic heterostructures.

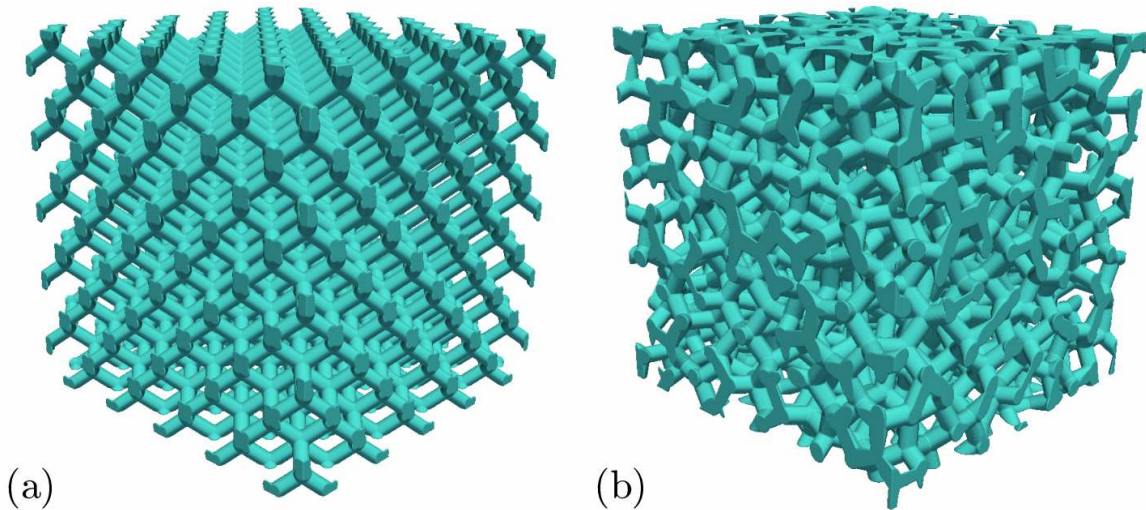


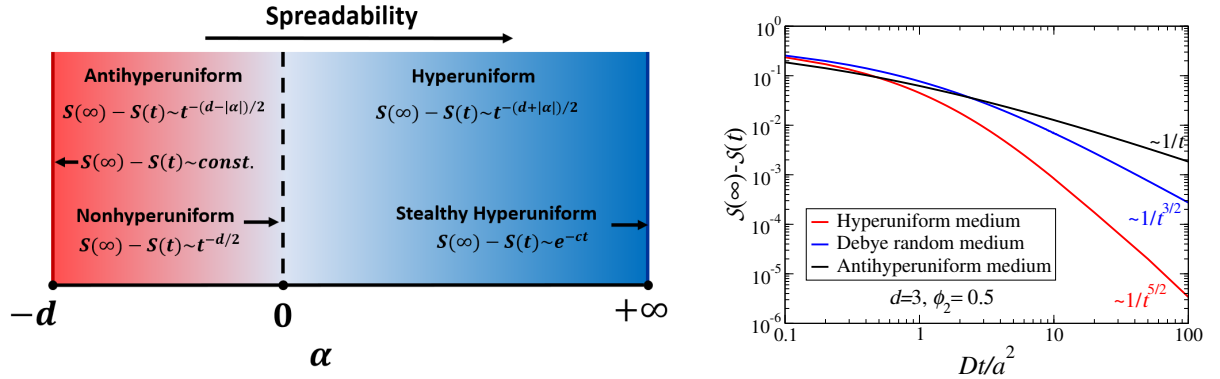
Figure 6: Photonic networks: (a) the crystal diamond network, where cylindrical rods connect nearest neighbors of a diamond lattice, and (b) the disordered nearly hyperuniform network (NHN) model.

It is well known that the degeneracy of two-phase microstructures with the same volume fraction and two-point correlation function is generally infinite. To elucidate the degeneracy problem explicitly, we examined Debye random media, which are entirely defined by a purely exponentially decaying two-point correlation function [13]. We structurally discriminated three classes of Debye random media from one another by ascertaining their other statistical descriptors, including the pore-size, surface correlation, chord-length probability density, and lineal-path functions. We found that these three classes of Debye random media are generally distinguished by the aforementioned descriptors, and their microstructures are also visually distinct from one another. Our work further confirms the well-known fact that scattering information is insufficient to determine the effective physical properties of two-phase media. Additionally, our findings demonstrate the importance of the other two-point descriptors considered here in the design of materials with a spectrum of physical properties.

We used an event-driven molecular dynamics algorithm to produce maximally random jammed superball packings for a wide range of deformation parameters [14]. We characterized their large-scale structure by examining the behaviors of their structure factors and spectral densities, as the wave number tends to zero, and find that these packings are effectively hyperuniform for all values of p examined. We showed that the mean width is a useful length scale to make distances dimensionless in order to compare systematically superballs of different shape. Moreover, we computed the complementary cumulative pore-size distribution $F(\delta)$. From $F(\delta)$, we estimated how the fluid permeability, mean survival time, and principal diffusion relaxation time vary as a function of the deformation parameter. Additionally, we computed the diffusion spreadability of

these packings and find that the long-time power-law scaling indicates these packings are hyperuniform. Our results can be used to help inform the design of colloidal or granular materials with targeted densities and transport properties.

A new dynamic probe of the microstructure of two-phase media, called the “spreadability” $S(t)$, was previously introduced by the principal investigator. This quantity provides a measure of the spreadability of diffusion information as a function of time t in any space dimension d . We derived exact integral relations for $S(t)$ that depends only on the spectral density $\tilde{\chi}_V(\mathbf{k})$. The short-, intermediate- and long-time behaviors of $S(t)$ are determined by the small-, intermediate- and large-scale structural characteristics. For hyperuniform media, we show 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. A remarkable link of spreadability to NMR pulsed field gradient spin-echo amplitude and diffusion MRI was shown. We applied this new theoretical/experimental tool to characterize many different models and a porous-medium sample [15]. We also devised an efficient algorithm to compute the spreadability for digitized two-phase media [16].



(a) “Phase diagram” that schematically shows the spectrum of spreadability regimes in terms of the exponent α . (b) Excess spreadability versus dimensionless time for antihyperuniform, typical nonhyperuniform and hyperuniform 3D models.

The principal investigator wrote a journal article to review the emerging field of disordered hyperuniform composites and their novel multifunctional characteristics [17]. 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 are shown to have advantages over their periodic counterparts, such as unique or nearly optimal, direction-independent physical properties and robustness against defects. It was 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.

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3 Impact

Our work has broadened our fundamental understanding of the structure and multifunctionality of disordered hyperuniform composites. The results contribute to this emerging field that is undergoing explosive growth, impacting physics, materials science, mathematics and biology. We expect that the anomalous reduction of volume-fraction fluctuations that disordered hyperuniform composites possess will lead to a new class of materials, devices and microsystems with desirable transport, thermal, mechanical, phononic and chemical properties, including thermal and electrical conductivity, diffusion coefficient, fluid permeability, elastic and viscoelastic moduli, photonic and acoustic band gaps, damage and failure characteristics, and chemical reaction rates. Current 3D additive manufacturing methods will enable one to fabricate our computational designs and hence test them experimentally, which should lead to the discovery of novel multifunctional materials.

4 Personnel Supported and Unsupported

In addition to the principal investigator, Professor Salvatore Torquato, the salaried research group at Princeton University included 7 graduate students: Duyu Chen, Jaek Kim, Zheng Ma, Charles Maher, Jacob Metcalfe, Timothy Middlemas and Murray Skolnick..

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