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Designing disordered hyperuniform two-phase materials with novel physical properties

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ABSTRACT

Heterogeneous materials consisting of different phases are ideally suited to achieve a broad spectrum of desirable bulk physical properties by combining the best features of the constituents through the strategic spatial arrangement of the different phases. Disordered hyperuniform heterogeneous materials are new, exotic amorphous matter that behave like crystals in the manner in which they suppress volume-fraction fluctuations at large length scales, and yet are isotropic with no Bragg peaks. In this paper, we formulate for the first time a Fourier-space numerical construction procedure to design at will a wide class of disordered hyperuniform two-phase materials with prescribed spectral densities, which enables one to tune the degree and length scales at which this suppression occurs. We demonstrate that the anomalous suppression of volume-fraction fluctuations in such two-phase materials endow them with novel and often optimal transport and electromagnetic properties. Specifically, we construct a family of phase-inversion-symmetric materials with variable topological connectedness properties that remarkably achieves a well-known explicit formula for the effective electrical (thermal) conductivity. Moreover, we design disordered stealthy hyperuniform dispersion that possesses nearly optimal effective conductivity while being statistically isotropic. Interestingly, all of our designed materials are transparent to electromagnetic radiation for certain wavelengths, which is a common attribute of all hyperuniform materials. Our constructed materials can be readily realized by 3D printing and lithographic technologies. We expect that our designs will be potentially useful for energy-saving materials, batteries and aerospace applications.

1. Introduction

Heterogeneous materials that consist of different phases (or constituent materials) abound in nature and synthetic products, such as composites, polymer blends, porous media, and powders [1–4]. In many instances, the length scale of the inhomogeneities is much smaller than the macroscopic length scale of the material, and microscopically the material can be viewed as a homogeneous material with macroscopic or *effective* properties [1,5–8]. It has been shown that given the individual phases, the effective properties of the materials are uniquely determined by microstructure of the phases [1]. Consequently, the discovery of novel guiding principles to arrange the constituents presents a promising path to design and realize materials with a broad spectrum of exotic and desirable properties by combining the best features of the constituents. The concept of disordered hyperuniformity provides guiding design principles for the creation of materials with singular performance characteristics, as we will demonstrate in this work.

The notion of hyperuniformity was first introduced in the context of many-particle systems over a decade ago [9]. Hyperuniform many-body systems are those characterized by an anomalous suppression of density fluctuations at long wavelengths relative to those in typical disordered systems such as ideal gases, liquids and structural glasses. All perfect crystals and perfect quasicrystals, and certain special disordered systems are hyperuniform [9,10]. Disordered hyperuniform many-particle systems are exotic amorphous state of matter that lie between crystal and liquid states: they behave like crystals in the way that they suppress

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density fluctuations at very large length scales, and yet they are statistically isotropic with no Bragg peaks. There is a special type of hyperuniformity called disordered stealthy hyperuniformity, characterized by the absence of scattering within a range of small wavenumbers around the origin in the Fourier space [11,12].

The exotic structural features of disordered hyperuniform systems appear to endow such systems with novel physical properties. For example, disordered hyperuniform dielectric networks were found to possess complete photonic band gaps comparable in size to photonic crystals [13,14]. Interestingly, such networks are isotropic, i.e., electromagnetic radiation propagates through the networks independent of the direction, which is an advantage over photonic crystals, and thus makes them suitable for applications such as lasers, sensors, waveguides, and optical microcircuits [14]. Moreover, disordered hyperuniform patterns can have optimal color-sensing capabilities, as evidenced by avian photoreceptors [15]. Recently it was revealed that the electronic band gap of amorphous silicon widens as it tends toward a hyperuniform state [16]. In the context of superconductors, it was shown that hyperuniform pinning site geometries exhibit enhanced pinning [17], which is robust over a wide range of parameters. In addition, there is evidence suggesting that disordered hyperuniform particulate media possess nearly optimal transport properties while maintaining isotropy [18].

These tantalizing findings have provided an impetus for researchers to discover and/or synthesize new disordered hyperuniform systems. We now know that disordered hyperuniformity arises in both equilibrium and nonequilibrium systems across space dimensions; e.g., maximally random jammed hard-particle packings [19–22], driven nonequilibrium granular and colloidal systems [23,24], dynamical processes in ultracold atoms [25], geometry of neuronal tracts [26], immune system receptors [27] and polymer-grafted nanoparticle systems [28]. The reader is referred to Refs. [29] and [30] for a comprehensive review of disordered hyperuniform systems that have been discovered so far.

Recently the concept of disordered hyperuniformity has been generalized to two-phase heterogeneous materials [10,29,30]. These materials possess suppressed volume-fraction fluctuations at large length scales, and yet are isotropic with no Bragg peaks. This can sometimes offer advantages over periodic structures with high crystallographic symmetries in which the physical properties can have undesirable directional dependence [13,14]. Specifically, the spectral density $\tilde{\chi}_V(\mathbf{k})$ of such system goes to zero as the wavenumber k goes to zero with a power-law scaling [10,19–21,31], i.e.,

$$\tilde{\chi}_V(\mathbf{k}) \sim |\mathbf{k}|^{\alpha}, \tag{1}$$

where α is the scaling exponent. Equivalently, the local volume-fraction variance $\sigma_V^2(R)$ associated with a d-dimensional spherical observation window of radius R possesses the following scaling at large R [10,19–21,31]:

$$\sigma_V^2(R) \sim \begin{cases} R^{-(d+1)}, & \alpha > 1, \\ R^{-(d+1)} \ln R, & \alpha = 1, \\ R^{-(d+\alpha)}, & 0 < \alpha < 1. \end{cases} \quad (R \to \infty) \tag{2}$$

where d is the dimension. Note that in all three cases $\sigma_V^2(R)$ decays more rapidly than the inverse of the window volume, i.e., faster than R^{-d}, which is different from typical disordered two-phase materials.

Our ability to design disordered hyperuniform two-phase materials in a systematic fashion is currently lacking and hence their potential for applications has yet to be explored. In this work, we develop for the first time a Fourier-space based numerical construction procedure to design at will a wide range of disordered hyperuniform two-phase materials by tuning the shape of the spectral density function across phase volume fractions. This is equivalent to tuning the degree and length scales at which there is anomalous suppression of volume-fraction fluctuations in these materials. We note that the Fourier-space setting is the most natural one, since hyperuniformity is defined in Fourier space. This setting is crucial for capturing accurately the long-wavelength, or equivalently, small-wavenumber k behavior. Our designed disordered hyperuniform microstructures include ones with phase-inversion symmetry as well as a stealthy dispersion. We compute the two-point cluster function, which incorporates nontrivial topological connectedness information and is known to provide a discriminating signature of different microstructures [32].

Subsequently, we investigate the effective transport properties and wave-propagation characteristics of these materials. We demonstrate that the anomalous suppression of volume-fraction fluctuations in hyperuniform two-phase materials endow them with novel and often optimal transport and electromagnetic properties. In the case of phase-inversion-symmetric materials, we show that they indeed achieve a well-known explicit formula for the effective electrical (thermal) conductivity. On the other hand, the stealthy dispersion possesses nearly optimal effective conductivity while being statistically isotropic. It is noteworthy that the frequency-dependent effective dielectric constant of any two-phase hyperuniform material cannot have imaginary part, implying that any such material is dissipationless (i.e., transparent) to electromagnetic radiation in the long-wavelength limit. Hence, all of our designed hyperuniform materials possess such characteristics. Moreover, our constructed dispersion is transparent for a range of wavelengths as well.

It is noteworthy that our tailored composite microstructures can be readily realized by 3D printing and lithographic technologies. We expect that our designs will be potentially useful for energy-saving materials [33], batteries [34] and aerospace applications [35].

In Sec. 2, we describe the Fourier-space based construction technique to design disordered hyperuniform two-phase materials. In Sec. 3, we employ our construction technique to generate disordered hyperunform two-phase microstructures with prescribed spectral densities. In Sec. 4 we compute the corresponding effective transport properties and wave-propagation characteristics of the designed two-phase materials. In Sec. 5, we offer concluding marks, and discuss potential application and extension of our results.

2. Fourier-space construction procedure

2.1. Algorithm description

The microstructure of a random multi-phase material is uniquely determined by the indicator functions $\mathscr{I}^{(i)}(\mathbf{x})$ associated with all of the individual phases defined as

$$\mathscr{I}^{(i)}(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \text{ in phase } i \\ 0, & \text{otherwise} \end{cases} \tag{3}$$

where $i = 1, \ldots, q$ and q is the total number of phases [1]. For statistically homogeneous two-phase materials where there are no preferred centers, the two-point probability function $S_2^{(i)}(\mathbf{r})$ measures the probability of finding two points separated by vector displacement $\mathbf{r}$ in phase i [1]. The autocovariance function $\chi_V(\mathbf{r})$ is trivially related to $S_2^{(i)}(\mathbf{r})$ via