Hyperuniformity of generalized random organization models

Zheng Ma
Department of Physics, Princeton University and
Princeton, New Jersey 08544, USA

Salvatore Torquato
Department of Chemistry, Princeton University
Department of Physics, Princeton University
Princeton Institute for the Science and Technology of Materials, Princeton University
Program in Applied and Computational Mathematics, Princeton University, and
Princeton, New Jersey 08544, USA

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Studies of random organization models of monodisperse spherical particles have shown that a hyperuniform state is achievable when the system goes through an absorbing phase transition. Here we investigate to what extent hyperuniformity is preserved when the model is generalized to particles with a size distribution and/or nonspherical shapes. We examine binary disks, disks with a size distribution and hard rectangles of various aspect ratios. We show that the systems are hyperuniform as two-phase media at their respective critical points. This analysis reveals that the redistribution of “mass” of the particles rather than the particle centroids is central to this dynamical process. 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 enables the creation of multifunctional hyperuniform materials with desirable optical, transport and mechanical properties.

Introduction. Periodically driven colloidal suspensions were observed to have a phase transition in terms of the reversibility of the dynamics one decade ago [11, 12]. A simple model named random organization [2], which is an absorbing phase transition model [3, 4], successfully captured the phenomenology. At around the same time, the conception of hyperuniformity, which describes the unusual suppression of number fluctuations of point configurations at large length scales, came to the fore [5]. Hyperuniform point configurations possess a structure factor \( S(k) \) that goes to zero as the wavenumber \( k \) vanishes, i.e.,

\[
\lim_{|k| \to 0} S(k) = 0. \tag{1}
\]

A hyperuniform system in \( d \)-dimensional space \( \mathbb{R}^d \) is poised at a special critical point \([5]\) in which the direct correlation function, defined via the Ornstein-Zernike relation \([6]\), is long-ranged, which is the diametric opposite behavior of traditional critical points in which the total correlation is long-ranged. Since 2003, many different types of disordered hyperuniform systems, including equilibrium and nonequilibrium varieties, have been identified; see the recent review [7].

Surprisingly, it was only recently that Hexner and Levine showed that the critical absorbing states associated with random organization models are actually hyperuniform [8]. They fall in a special class of hyperuniform point configurations called class III [7], where the structure factor scales as a power \( S(k) \sim k^{d-\alpha_N} \) with \( 0 < \alpha_N < 1 \) as \( k \to 0 \), and number variance scales as a power-law \( R^{d-\alpha_N} \). Many variations of such models have been studied [9, 10]. Moreover, experimentally, such protocols suggest that hyperuniform state of matter can be made in a self-organized fashion [11, 12]. However, previous numerical studies have focused on models that are based on monodisperse spherical particles, which is an idealization that may not be achievable nor tunable in practice. Interestingly, it is not known whether multicomponent systems subjected to random-organization dynamics preserves hyperuniformity, and, if so, how it is preserved. If it is preserved, would the multicomponent system also be “multi-hyperuniform” [17, 18] (each species the system consists of is hyperuniform by its own) at the critical point? Should we consider the system as a two-phase medium, and apply the corresponding generalization of hyperuniformity associated with volume-fraction fluctuations [19]? In this case, hyperuniformity is defined for a two-phase medium in terms of the spectral density \( \tilde{\chi}_V(k) \), which approaches zero as the wavenumber \( |k| \) vanishes

\[
\lim_{|k| \to 0} \tilde{\chi}_V(k) = 0. \tag{2}
\]

Another unexplored question is whether the system would maintain hyperuniformity if nonspherical (noncircular) particles are used, in which case the rotational degree of freedom is introduced. This Letter investigates all of these extensions of random organization models and their consequences.

Here we consider the isotropic version of the model and use the volume fraction \( \phi \) as a control parameter [20]. The system starts from a collection of randomly placed particles. At a given instant of time \( t \), any pair of particles that overlap with one another are considered “active” and are given a random kick in the next time step. The system dynamics are followed until a steady-state is established. The eventual number fraction of active par-
particles \( f_a \) is either zero or a positive value, depending on whether \( \phi \) is below or above the critical volume fraction \( \phi_c \). Unless otherwise specified, the amplitude of a kick is always randomly and uniformly chosen between 0 and \( c\sqrt{\phi/N} \), where \( N \) is the total number of particles and the constant \( c \) is \( 1/2\sqrt{\pi} \) for disks and \( 1/4 \) for noncircular particles. The system size used throughout this paper is 100,000 particles and ensemble averages of 10–100 configurations are carried out for all of the reported results.

\[ \gamma = \frac{S(k)}{L(k)} \]

\[ \gamma = 0, 0.33, 0.5, 0.57, 0.67, 0.8, 1 \]

\[ \phi_c \]

\[ \phi \]

\[ \phi \] is always randomly and uniformly chosen between 0 and 1. Unless otherwise specified, the amplitude of a kick is chosen randomly from a uniform distribution centered at 0.

**FIG. 1.** (a) A representative image of a configuration of a 2D binary disk system in an absorbing state with the small-to-large particle size ratio \( \gamma = 0.5 \) and the relative number fraction of small disks \( x = 0.5 \). (b) Critical volume fraction \( \phi_c \) as a function of \( \gamma \) with fixed \( x = 0.5 \). The smooth curve is fit well by a polynomial of degree 4 (blue curve). Interestingly, it is seen that there is a local minimum at \( \gamma \approx 0.2 \).

**Mixture of Disks.** We begin by considering binary hard-disk systems in two dimensions. We let \( \gamma = R_S/R_L \) denote the small-to-large particle size ratio and \( x = N_S/(N_S + N_L) \) denote the relative number fraction of small disks. Since the parameter space is infinite, we restrict our study to two regimes: one in which we vary \( \gamma \) at fixed \( x = 0.5 \) and another in which we vary \( x \) at fixed \( \gamma = 0.5 \). Since the conclusions that we reach for the two different regimes are similar, we focus on the one in which the small-disk fraction is fixed and leave the results of the other regime to Supplemental Material [21].

We study a sequence of systems with \( \gamma \) ranging from 0 to 1 and identify their critical absorbing states respectively. An example of a 2D binary disk system in an absorbing state is shown in Fig. 1(a). We find that the critical volume fraction \( \phi_c \) varies nonmonotonically over the entire range of \( \gamma \), as shown in Fig. 1(b). This also holds for systems with fixed \( \gamma \) (see Supplemental Material [21]), suggesting that the function \( \phi_c(x, \gamma) \) is a smooth function bounded in a small interval. Note that as \( \gamma \) decreases away from 1, \( \phi_c \) decreases over a certain wide range of \( \gamma \). However when \( \gamma = 0 \), the small particles become effectively points and the system behaves like a monodisperse system again. As \( \gamma \) increases away from 0, \( \phi_c \) decreases again such that there is a local minimum at around \( \gamma = 0.2 \).

To ascertain the possible hyperuniformity exhibited by these critical absorbing states, we first compute the structure factors associated with the particle centroids for each species as well as for the whole system for these systems in Fig. 2. As the size discrepancy increases (\( \gamma \) decreases), the larger particles become more and more ordered, while the smaller particles goes in the opposite direction. The overall long-range density fluctuations increase as size discrepancy increases, as evidenced by a structure factor at the origin \( S(k = 0) \) that increases. Observe that there is always a small kink at small wavenumbers.

**FIG. 2.** Structure factors for large particles \( S_L(k) \), small particles \( S_S(k) \) and the whole system \( S(k) \) for different size ratios with fixed \( x = 0.5 \). Note that as the size discrepancy increases, large particles become more uniformly distributed while small particles become more disordered, but none of them are hyperuniform.

Clearly, these binary systems are not hyperuniform if they are seen as point patterns determined by the particle centroids, as the case for monodisperse disks. However, by applying the appropriate generalization of hyperuniformity to two-phase systems [19], we find that the binary systems at criticality are indeed hyperuniform. We com-
pute the spectral densities $\tilde{\chi}_v(k)$ of the resulting two-phase systems by taking the space interior to particles as one phase and the space exterior as the other. The results are shown in Fig. 3. Notice that while the spectral densities for different compositions vary greatly (especially for $k$ larger than the first peak, see Supplemental Material [21] for a rescaled plot), they all go to zero as $k \to 0$, meaning that they are all hyperuniform with respect to volume-fraction fluctuations. The scaling behavior of spectral densities, i.e., $\tilde{\chi}_v(k) \sim k^{\alpha_v}$ as $k \to 0$, is shown in the log-log plot of the small-$k$ region in Fig. 3(b). Interestingly, it further reveals that all of the spectral densities go to zero with the same scaling [22], implying that these systems have similar large scale structures despite their different compositions. We find the exponent $\alpha_v = 0.42 \pm 0.04$, which is consistent with previously reported values [8, 15, 21] (see Supplemental Material [21] for the details of the fitting procedure). The hyperuniformity of the resulting two-phase media, as quantified by the spectral density, along with the nonhyperuniformity of particle centroids, implies that the rearrangement of particles to suppress their volume-fraction (“mass”) fluctuations rather than the number fluctuations is at the heart of random organization dynamics. Since the former implies the latter in the monodisperse case, this discovery could not have been made if we had treated number fluctuations associated with the centroids of the particles, as was done in previous studies of monodisperse spheres (circles).

To further generalize our findings, we investigate disks with a continuous size distribution (with radius chosen from a uniform distribution $U(R, 1.5R)$); see Fig. 4(a) for a representative image. We find again that the two-phase medium at the critical point is hyperuniform, as shown in Fig. 4(f).

Non-spherical/noncircular Particle Shapes. Our findings thus far have revealed that the random organization mechanism provides a robust means of generating hyperuniform systems consist of a variety of mixtures of spherical (circular) particles. This naturally leads us to consider a wider class of particle shapes. Specifically, we extend this model to noncircular particles [23], namely hard rectangles of certain aspect ratios $L/W$ (where $L$ is the length and $W$ is the width of the rectangle), including the hard-needle limit. This allows us to study the effect of rotational degree of freedom. For every random kick, we also need to include a random rotation $\delta \theta$ of the active particle. Here we chose $\delta \theta$ uniformly from $[-\theta_0, \theta_0]$. We did not find fundamental differences between different choices of $\theta_0$ except for a shift of the critical point, and thus we use $\theta_0 = \pi/2$ for the ensuing discussion.

Representative images of squares ($L/W = 1$) and needles ($L/W = \infty$) in absorbing states are shown in Fig. 4(b) and (c). For rectangles, we find that the critical packing fraction $\phi_c$ decreases monotonically as the aspect ratio $L/W$ increases. This is not surprising, since in the limit $L/W \to \infty$, the problem is reduced to the random organization of hard needles. Since the packing fraction is diminishing to zero in this limit, a more proper quantity to focus on is $\rho_c L^2$, which we refer to as a critical reduced density. We plot the critical reduced density as a function of $L/W$ in Fig. 4(d), one can see that it converges as $L/W \to \infty$.

Unlike the quasi-long-range correlation found in the XY model [24] and two-dimensional hard rods at equilibrium [25], the orientational correlation function $g_\theta(r) = \langle \cos(2(\theta(0) - \theta(r))) \rangle$ for hard needles decays to zero almost immediately beyond $r = L$ (see Fig. 4(e)). This definitively demonstrates that only short-range orientational order is present in these noncircular particle systems. Note that the critical reduced density is well below the isotropic-nematic transition density [20, 27]. Indeed, we did not observe any nematic order in these systems. We theoretically determined the orientational correlation function based on hard needles in equilibrium in the dilute limit, which agrees qualitatively well with our simulations, as shown in Fig. 4(e); see Supplemental Material [21] for the details of the theoretical derivation.

The spectral densities (see Fig. 4(f)) show that for these noncircular particles, hyperuniformity is still preserved. For hard needles, it is crucial to employ the spectral density for interface to obtain the result [19, 28]. Interestingly, as the aspect ratio $L/W$ increases, one can
clearly see the destruction of short-range order from the increasingly diffusive tails in the spectral densities. This can also be seen from the damped oscillations in the structure factors (see Supplemental Material [21]). The reason is that less symmetric particles would broaden the distribution of short-range particle-particle spacing. However, as one can see from the Fig. 4(f), despite the fact that the spectral density profiles appear to be quite different from one another, they all exhibit the same small-$k$ behavior [29].

**Discussion.** The results of our investigation expand our understanding of random organization as a model for absorbing phase transitions of continuum media by going beyond the previously studied monodisperse systems. The more general treatment of the systems as two-phase media has led us to the proposition that “active volume” is the appropriate general quantity to examine rather than the “active particle numbers” (see Supplemental Material [21] for additional data). Moreover, our work demonstrates that random organization provides a robust and versatile means of generating a wide class of disordered hyperuniform two-phase media. This includes a broad spectrum of mixtures of particles (discrete and continuous) of circular or noncircular shape. Practically, our findings imply that hyperuniform materials can be made in the laboratory without the need to use monodisperse particles, which eases the preparation process. Moreover, different structures can be made in the laboratory by changing the composition of periodically driven colloids via their size and shape distributions. These structures will maintain hyperuniformity in the small-$k$ region, while functional form of the spectral densities away from the origin can vary widely. Note the spectral density (or equivalently its corresponding direct-space two-point correlation function [30]) controls a variety of different effective properties of two-phase media, including the effective dielectric tensor [31], fluid permeability [32], mean survival time [33], structural-color characteristics [34], and mechanical properties [35].
among other quantities. Thus, the tunability of the functional form of the spectral density and associated structures enables the creation of multifunctional hyperuniform materials by self-organization.

It is noteworthy that although theoretical understandings of the model are limited [29], it has been shown that the phase transition in certain random organization models belongs to the universality class of directed percolation [20]. Although this is not the focus of this Letter, we computed a critical exponent $\beta \simeq 0.56 \pm 0.02$ (L/$a \propto (\phi - \phi_c)^\beta$), which is reminiscent of directed percolation in 2+1 dimensions [67] (see Supplemental Material [21]).

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[21] See Supplemental Material at [URL] for additional figures and formulas.
[22] For monodisperse spherical particles, we have $\tilde{\chi}_v(k) = \rho(2\pi a/k)^d F_d(k a) S(k)$ [19], where $a$ is the radius and $F_d(k a)$ is the Bessel function. This guarantees that the spectral density scales the same as the structure factor as $k \to 0$, i.e., $\alpha_v = \alpha_s$. However, the relation between $\tilde{\chi}_v(k)$ and partial structure factors become much more complex for polydisperse particles [19], and one might not expect to see the same equality. Thus, it is a significantly more nontrivial result that polydisperse particles somehow arrange themselves in such a way that the spectral densities share the same scaling as the structure factor for monodisperse particles. Interestingly, this is what we see in Fig. 3(b) for different mixtures, despite the fact that their structure factors look quite different in Fig. 2.
Supplemental Material: Hyperuniformity of Generalized Random Organization Models

**BINARY DISKS**

Here we report results for fixed small-to-large particle size ratio \( \gamma = R_S/R_L = 0.5 \). We plot the critical volume fraction \( \phi_c \) as a function of the relative number fraction of small disks \( x = N_S/(N_S + N_L) \) in Fig. S1. Again, we find \( \phi_c \) lies in a small interval \([0.385, 0.410]\). Note there is also a dip at around \( x = 0.8 \).

![Graph of \( \phi_c \) vs. \( x \)](null)

**FIG. S1.** Critical volume fraction \( \phi_c \) as a function of the relative number fraction of small disks \( x = N_S/(N_S + N_L) \) with fixed small to large particle size ratio \( \gamma = 0.5 \).

We show that the critical volume fraction \( \phi_c \) shifts as the composition is varied, and so does the spectral density profile at the critical point. However, the spectral density profiles at the critical points have the same small-\( k \) scaling, implying similar large-scale structures of these configurations. We find that the exponent \( \alpha_V \) given by the least square fit is sensitive to the choice of fitting intervals due to the fluctuations in the small-\( k \) region. In order to obtain a reliable value, we ran over 100 samples of the monodisperse system consists of 100,000 particles, and averaged over the results of 87 samples that finally reached absorbing states. We find that as the fitting interval shrinks from the first 50 \( k \)-points to the first 5 \( k \)-points, the best fitted exponent gradually increases from 0.38 to 0.45, however, the corresponding uncertainty increases from \( \pm 0.005 \) to \( \pm 0.1 \). To balance the bias induced by the large intervals and the fluctuations in the small intervals, we report the result \( 0.42 \pm 0.04 \), which lies in the middle of the two extremes. Here we show the spectral densities rescaled by their first peaks for both fixed \( x = 0.5 \) and \( \gamma = 0.5 \) in Fig. S2. Note that these curves approximately collapse onto a single curve in the small-\( k \) region, while for the region that \( k > k_{peak} \) they vary greatly.

![Graph of spectral densities rescaled by their first peaks](null)

**FIG. S2.** Spectral densities rescaled by their first peaks \( (k_{peak}, \tilde{\chi}_V(k_{peak})) \) for (a) fixed relative number fraction of small disks \( x = 0.5 \) and (b) fixed small-to-large particle size ratio \( \gamma = 0.5 \), as well as a mixture of disks with a continuous size distribution. Note that these curves approximately collapse onto a single curve in the small-\( k \) region.

**STRUCTURE FACTORS FOR PARTICLES OF DIFFERENT SHAPES**

We compare the structure factors of the centroids of rectangles with different values of the aspect ratio \( L/W \), as well as the one for monodisperse disks near their critical points in Fig. S3. The structure factors show that hyperuniformity is still preserved for these non-spherical particles. Notice that the structure factor for squares is very similar to that of disks, the positions of peaks are unchanged while the oscillations are damped. We believe this is a common feature for random organizations of all equilateral polygons. However, as the aspect ratio \( L/W \) increases, one can clearly see the destruction of short-ranged order as evidenced by the damping of oscillations in structure factors. As one can see from the plots, despite the fact that the profiles of structure factors look
quite different, they all exhibit the same small-$k$ behavior, except the one for $L/W = 10$, where $S(k)$ is convex near the origin.

![Graph showing comparison of structure factors for configurations of monodisperse particles with different shapes near critical points under the random organization dynamics.](image)

**FIG. S3.** Comparison of structure factors for configurations of monodisperse particles with different shapes near critical points under the random organization dynamics.

**CRITICAL EXponent $\beta$**

We show the final number fraction of active particles $f_a$ as a function of $(\phi - \phi_c)/\phi_c$ for a few systems in Fig. S4(a). We find the critical exponent $\beta \simeq 0.56 \pm 0.02$, which is reminiscent of directed percolation in 2+1 dimensions. Although for different mixtures $f_a$ approximately have the same scaling, those curves never collapse onto a single one. Interestingly, if “active particles” is replaced by “active volume” $V_a$ (which simply means the volume of the space that overlaps with other particles), then the curves collapse onto a single one, as one can see in Fig. S4(b). This may imply that “active volume” is a more appropriate quantity to investigate. Moreover, by using “active volume”, we find a different exponent $\beta' \simeq 0.68 \pm 0.08$, which is closer to the value of the universality class of conserved directed percolation (a more intuitive candidate for random organization).

![Graph showing number fraction of active particles $f_a$ and rescaled “active volume” as functions of the scaled volume fractions for different small to large particle size ratios $\gamma$ with $x = 0.5$.](image)

**FIG. S4.** (a) The number fraction of active particles $f_a$ and (b) rescaled “active volume” as functions of the scaled volume fractions for different small to large particle size ratios $\gamma$ with $x = 0.5$.

**ORIENTATIONAL CORRELATION FUNCTION**

Suppose the needle has unit length and the distance between the centroids of two needles is $r$. Let

$$\theta_1(x; r) = \sin^{-1} \left( \frac{r \sin(x)}{\sqrt{r^2 - r \cos(x) + 0.25}} \right),$$

$$\theta_2(x; r) = \sin^{-1}(2r \sin(x)).$$

By averaging over all of the feasible configurations of two hard needles, the orientational correlation function $g_\theta(r) = \langle \cos(2(\theta(0) - \theta(r))) \rangle$ can be written as

$$g_\theta(r) = \begin{cases} 
\int_0^{\cos^{-1}(r)} \frac{\sin(2\theta_1(x; r)) + \sin(2\theta_2(x; r))}{\theta_1(x; r) + \theta_2(x; r)} \, dx + \int_{\cos^{-1}(r)}^{\frac{\pi}{2}} \frac{\sin(2\theta_2(x; r))}{\theta_2(x; r)} \, dx, & 0 < r \leq \frac{1}{2}, \\
\int_0^{\cos^{-1}(r)} \frac{\sin(2\theta_1(x; r)) - \sin(2\theta_2(x; r))}{\theta_1(x; r) + \theta_2(x; r)} \, dx + \int_{\cos^{-1}(r)}^{\frac{\pi}{2}} \frac{\sin(2\theta_2(x; r))}{\theta_2(x; r)} \, dx + \int_{\cos^{-1}(r)}^{\frac{\pi}{2} - \cos^{-1}(r)} \frac{\sin(2\theta_2(x; r))}{\theta_2(x; r)} \, dx, & \frac{1}{2} < r \leq \frac{\sqrt{2}}{2}, \\
\int_0^{\cos^{-1}(r)} \frac{\sin(2\theta_1(x; r)) - \sin(2\theta_2(x; r))}{\theta_1(x; r) + \theta_2(x; r)} \, dx, & \frac{\sqrt{2}}{2} < r \leq 1, \\
0, & r > 1.
\end{cases}$$

(5)