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Disordered multihyperuniformity derived from binary plasmas

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Disordered multihyperuniform many-particle systems are exotic amorphous states that allow exquisite color sensing capabilities due to their anomalous suppression of density fluctuations for distinct subsets of particles, as recently evidenced in photoreceptor mosaics in avian retina. Motivated by this biological finding, we present the first statistical-mechanical model that rigorously achieves disordered multihyperuniform many-body systems by tuning interactions in binary mixtures of non-additive hard-disk plasmas. We demonstrate that multihyperuniformity competes with phase separation and stabilizes a clustered phase. Our work provides a systematic means to generate disordered multihyperuniform solids, and hence the lays the groundwork to explore their potentially novel photonic, phononic, electronic and transport properties.

A hyperuniform state of matter is characterized by an anomalous suppression of density fluctuations at large length scales compared to the fluctuations in typical disordered point configurations, such as atomic positions in ideal gases, liquids, and glasses. A hyperuniform many-particle system in d-dimensional Euclidean space $\mathbb{R}^d$ at number density ρ is one in which the structure factor $S(\mathbf{Q}) \equiv 1 + \rho\tilde{h}(\mathbf{Q})$ tends to zero as the wavenumber $Q \equiv |\mathbf{Q}|$ tends to zero [1], i.e.,

$$\lim_{Q\to 0} S(\mathbf{Q}) = 0, \tag{1}$$

where $\tilde{h}(\mathbf{Q})$ is the Fourier transform of the total correlation function $h(\mathbf{r}) = g_2(\mathbf{r}) - 1$ and $g_2(\mathbf{r})$ is the pair correlation function. Hyperuniform many-body systems include crystals, quasicrystals, and certain exotic disordered systems [1, 2]. Disordered hyperuniform states lie between a crystal and liquid: they behave like perfect crystals in the manner in which they suppress large-scale density fluctuations and yet, like liquids and glasses, are statistically isotropic without Bragg peaks [3]. Due to their novel structural and physical properties, these exotic states of amorphous matter have been the subject of many recent investigations [2–17].

Disordered *multihyperuniform* many-body systems are a remarkable class of disordered hyperuniform states of matter that were first identified in the photoreceptor patterns in avian retina [7]. A disordered multihyperuniform point configuration is not only disordered and hyperuniform but contains multiple distinct subsets of the entire point configuration that are themselves hyperuniform. This twist on standard hyperuniformity is presumably partly responsible for the acute color sensing ability of birds with their five different cone photoreceptor subpopulations, each of which is separately hyperuniform. An open question since that time has been the identification of a theoretical model of interacting particles with disordered multihyperuniform states. In this Communication, we provide the first statistical-mechanical model of a many-body system that rigorously achieves disordered multihyperuniformity by tuning interactions in binary mixtures of hard-disk plasmas.

Hyperuniform systems can be regarded to be at an "inverted" critical point in which the direct correlation function $c(\mathbf{r})$, defined through the Ornstein-Zernike equation, is long-ranged and the total correlation function $h(\mathbf{r})$ is short-ranged, which is the diametric opposite of the behaviors of these functions at thermal critical points [1]. For a large class of disordered hyperuniform systems [10]

$$\lim_{Q\to 0} S(Q) \propto Q^{\alpha} \tag{2}$$

with $\alpha > 0$. This can be thought of as an "inverted" critical point and through the Ornstein-Zernike equation, this can be shown to determine the low-Q behavior of the Fourier transform of the direct correlation function $c(\mathbf{r})$ [1]:

$$\lim_{Q\to 0} \tilde{c}(Q) \propto Q^{-\alpha}. \tag{3}$$

Importantly, at large pair separations, the direct correlation $c(\mathbf{r})$ is equal to the negative of the pair potential $u(\mathbf{r})$ of a many-body system in equilibrium, and hence the small-Q behavior of the Fourier transform of the potential obeys the following power-law form:

$$\lim_{Q\to 0} \tilde{u}(Q) \propto -Q^{-\alpha}. \tag{4}$$

We see that condition (4) dictates what type of interactions can lead to a hyperuniform system in equilibrium. Such a situation is found for two- and three-dimensional Coulomb plasmas[18–22], for which $\alpha = 2$ and 1, respectively. Interestingly, the small-Q behavior of the structure factors of the distribution of avian photoreceptors is similar with the latter following linear/quadratic dependence [7]. If the system was to be modeled using coarse-grained effective interactions, these should then be plasma-like.

However, to achieve multihyperuniformity, one must consider particle mixtures, which is more involved if the overall hyperuniformity condition (1) is to be satisfied by the partial structure factors of each mixture component. In this connection, it was recently shown by the authors that superimposing a long-range Coulomb repulsion on a non-additive hard disk (NAHD) mixture induces hyperuniformity only on a global scale, and not for the structure of the mixture components [23]. In this Letter, we ascertain the conditions under which the interactions can be tuned to induce a multihyperuniform structure.

In general, in a binary mixture, the partial structure factors $S_{ij}(Q)$ can be expressed in terms of the Fourier transform of the corresponding total and direct correlation functions, $h_{ij}(r)$ and $c_{ij}(r)$, using the Ornstein-Zernike relation:

$$S_{ii}(Q) = x_i(1+\rho x_i \tilde{h}_{ii}(Q)) = x_i \frac{1-\rho_j \tilde{c}_{jj}}{|\mathbf{I}-\mathbf{C}|}$$
$$S_{ij}(Q) = \rho x_i x_j \tilde{h}_{ij}(Q) = \frac{\rho x_i x_j \tilde{c}_{ij}}{|\mathbf{I}-\mathbf{C}|} \qquad (5)$$

with $i \neq j$, and

$$|\mathbf{I}-\mathbf{C}| = 1-\rho_1\tilde{c}_{11}-\rho_2\tilde{c}_{22}+\rho_1\rho_2(\tilde{c}_{11}\tilde{c}_{22}-\tilde{c}_{12}^2), \qquad (6)$$

where the ρ_i are the partial number densities of each component, x_i the corresponding mole fractions, and $\rho = \rho_1+\rho_2$. Intuitively, one can require each component interaction to display the same singular behavior at $Q \to 0$ as found in disordered single component hyperuniform systems, namely $\lim_{Q\to 0} \tilde{c}_{ij}(Q) \sim -\beta\tilde{u}_{ij}(Q) \propto -\eta_{ij}Q^{-\alpha}$. Here η_{ij} is a constant dependent on the type of interaction and $\beta = 1/(k_BT)$ as usual. For standard Coulomb interactions, one has $\eta_{ij} \propto z_i z_j$, by which $\eta_{ij} = \sqrt{\eta_{ii}\eta_{jj}}$. In contrast to the single-component case, one observes that the denominator (6) contains a quadratic term that, in principle, can guarantee that $\lim_{Q\to 0} S_{ij} = 0$, $\forall(i,j)$, if

$$\lim_{Q\to 0} \beta^2 \left(\tilde{u}_{11}(Q)\tilde{u}_{22}(Q) - \tilde{u}_{12}(Q)^2\right) \neq 0. \qquad (7)$$

Here we have made use of the large-r asymptotic behavior of the direct correlation function $c_{ij}(r)$. If instead, the equality in expression (7) - basically one of Lorentz-Berthelot's mixing rules - is fulfilled, then Eqs. (5) lead to non-vanishing partial structure factors for $Q \to 0$, even if the total structure factor complies with (1) [23]. In what follows, we will show for a simple mixture model in two dimensions how multihyperuniformity can be achieved by a simple tuning of the cross interaction long-range behavior so that it satisfies the inequality (7).

Our model consists of a symmetric mixture of non-additive hard disks with a two-dimensional Coulomb repulsion added, such that the pair potential is given by

$$\beta u_{ij}(r) = \begin{cases} \infty & r < (1+\Delta(1-\delta_{ij}))\sigma \\ -\gamma_{ij} z_i z_j \Gamma \log r/\sigma & r \geq (1+\Delta(1-\delta_{ij}))\sigma \end{cases} \qquad (8)$$

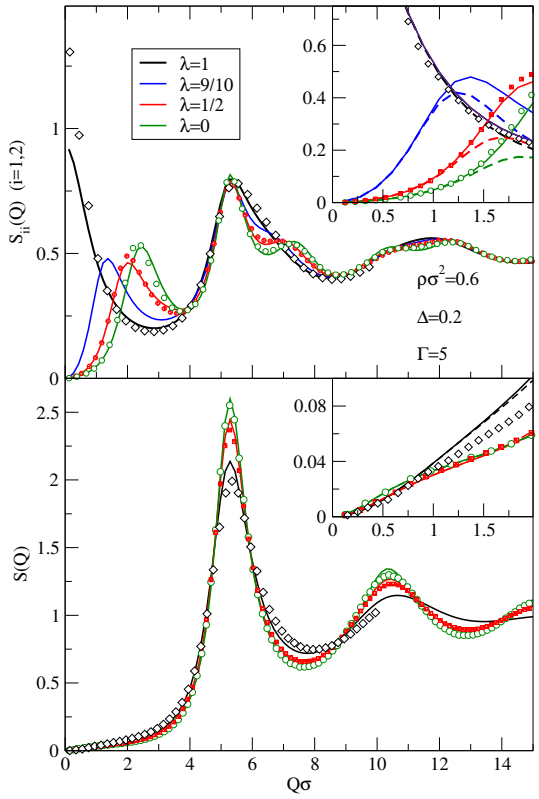

FIG. 1. Total and partial structure factor of the NAHD plasma for various values of the λ parameter tuning the Coulomb interaction between unlike particles. Solid curves correspond to RHNC-PY calculations, symbols denote Monte Carlo data. The multihyperuniformity induced by Coulomb interactions when $\lambda \neq 1$ counteracts phase separation by stabilization of a cluster-like phase with intermediate range order. This is reflected in the prepeak occurring at $Q\sigma \lesssim 2.3$. The insets illustrate the low-Q behavior of $S_{ii}(Q)$ as determined from Eq.(S3) in the SI, and of $S(Q)$ from Eq.(11) –dashed curves–. Note that given the symmetry in the interactions and composition, $S_{11} = S_{22}$.

where $\gamma_{ij} = (\lambda+(1-\lambda)\delta_{ij})$, $\Gamma = \beta e^2$, being e the electron charge (esu units), z_i, the particle charge in e units, δ_{ij} is a Kronecker δ, Δ is the non-additivity parameter, and σ the hard disk diameter between like species. For a plasma system we consider $z_i > 0$ for all species and a corresponding rigid neutralizing background. The key element in Eq. (8) is the λ parameter, which can be tuned in the range $0 \leq \lambda \leq 1$. When $\lambda = 1$ we have the regular situation in which $u_{12} \propto z_1 z_2$. As $\lambda < 1$, the unlike interaction become less repulsive, to go back to the plain hard-core repulsion when $\lambda = 0$. In all cases, when $\lambda \neq 1$, inequality (7) is fulfilled. When $\lambda > 1$ the system becomes unstable, as imbalanced long range interactions between unlike particles drive the mixture towards phase separation. Finally, our system is taken to be a symmetric mixture of total number density, ρ, with $\rho_1 = \rho_2 = \rho/2$ and $z_1 = z_2$.

In order to analyze the low-Q behavior of the partial

structure factors, we perform a small-Q expansion of $\tilde{c}_{ij}$ and separate out the Coulomb term:

$$\tilde{c}_{ij}(Q) \approx \tilde{c}_{ij}^{R}(0) + c_{ij}^{(2)}Q^2 - \gamma_{ij}2\pi\Gamma z^2/Q^2$$
$$c_{ij}^{(2)} = \frac{1}{2}\left.\frac{\partial \tilde{c}_{ij}^{R}(Q)}{\partial Q}\right|_{Q=0}, \quad (9)$$

where, $c_{ij}^{R}(0)$ is the $Q \to 0$ limit of the regular part of the direct correlation function [19]. Inserting (9) into Eq. (5), the low-Q limits of the partial structure factors are obtained. With these, the total structure factor, which in our symmetric case reduces to

$$S(Q) = 2S_{11}(Q) + 2S_{12}(Q), \quad (10)$$

can be expressed as

$$S(Q) \approx \frac{Q^2}{aQ^2 + \pi\rho\Gamma z^2(1+\lambda)} \quad (11)$$

when $Q \to 0$. The concentration-concentration structure factor[24] is defined in terms of the Fourier transform of the local concentration deviation from its average value, $\tilde{C}(Q)$, as

$$S_{cc}(Q) = N\langle\tilde{C}(\mathbf{Q})\tilde{C}(-\mathbf{Q})\rangle$$
$$= x_2^2 S_{11}(Q) + x_1^2 S_{22}(Q) - 2x_1x_2S_{12}(Q)$$

where, N in the number of particles, and $\langle\ldots\rangle$ denotes an ensemble average. In our fully symmetric equimolar case we have

$$S_{cc}(Q) = \frac{1}{2}(S_{11}(Q) - S_{12}(Q)), \quad (12)$$

which leads to ($Q \to 0$)

$$S_{cc}(Q) \approx \frac{Q^2}{bQ^2 + \pi\rho\Gamma z^2(1-\lambda)}. \quad (13)$$

In (11) and (13)

$$a = 1 - \frac{\rho}{2}(\tilde{c}_{11}^{R}(0) + \tilde{c}_{12}^{R}(0) + (c_{11}^{(2)} + c_{12}^{(2)})Q^2)$$
$$b = 1 - \frac{\rho}{2}(\tilde{c}_{11}^{R}(0) - \tilde{c}_{12}^{R}(0) + (c_{11}^{(2)} - c_{12}^{(2)})Q^2).$$

The simple expressions (11) and (13) are valid for all values of λ, and can be seen to accurately reproduce the low-Q behavior of the structure factors in the insets of Figure 1. When $\lambda = 1$ (usual Coulomb charge-charge interaction) one sees that $S(Q) \propto Q^2$ and $S_{cc}(Q) \propto 1/b$ as $Q \to 0$, i.e., the system will only be globally hyperuniform. For all other cases, the total and concentration-concentration structure factors vanish simultaneously as $Q \to 0$, meaning that the system is necessarily multihyperuniform. This is a general result, provided that the long-range part of the interaction satisfies

$$\lim_{Q\to 0} \tilde{u}_{ii}(Q) \propto -Q^{-\alpha}, \quad (14)$$

and inequality (7) holds, the system will display hyperuniformity. Notice that the unlike interactions need not be long-ranged. Neither the interaction symmetry implicit in Eq.(8) nor the equimolar composition are essential to our results. On the other hand, if the interactions do not comply with (7), global hyperuniformity will only occur if the symmetry $\lim_{Q\to 0}(\tilde{u}_{11}(Q) - \tilde{u}_{22}(Q)) = 0$ is preserved[23]. Note that charge asymmetry in Coulomb systems will destroy global hyperuniformity whenever the inequality (7) is not fulfilled.

We have studied our system resorting to the solution of the Reference Hypernetted Chain Equation (RHNC), with a bridge function of a plain NAHD reference fluid obtained in the Percus-Yevick (PY) approximation. Specific numerical procedures are detailed in Ref. [25]. Additionally, we have made extensive use of Monte Carlo simulations, in which long-range interactions have been treated using Ewald summations with conducting boundary conditions [25]; see Ref. [23] for additional details.

A particularly interesting situation occurs when one deals with positive non-additivity. This is a well-known situation in which volume effects in the NAHD system will lead to demixing [26, 27]. Here we will focus on a system with $\rho\sigma^2 = 0.6$, $\Delta = 0.2$ and $\Gamma = 5$ (relatively close to the demixing critical point, located at $\rho_c\sigma^2 = 0.69$ [23]). Interestingly, according to Eq. (13), once $\lambda \neq 1$, long-range concentration-concentration fluctuations will be suppressed (the system becomes multihyperuniform) and hence phase separation is no longer possible. This effect is illustrated in Fig. 1, where the evolution of the partial, $S_{ii}(Q)$, $(i = 1, 2)$ and the total structure factor, $S(Q)$, is displayed for various λ values. For $\lambda = 1$, we see that S_{11} grows rapidly as $Q \to 0$ (in parallel with $S_{cc}(Q)$; see Supplementary Material [28]), a clear indication of the vicinity of the demixing transition. Then, for $\lambda \neq 1$, one observes the presence of a prepeak whose position in Q-space increases as λ decreases. This prepeak is the characteristic signature of intermediate range order [29], and it usually reflects spatial correlations between stable clusters[30]. Indeed, the effect of lowering λ can be readily interpreted in terms of competition between effective interactions. The volume effects induced by the positive non-additivity are equivalent to effective short-range attractions between like particles in the mixture. These effective short-range attractions are responsible for the demixing that takes place when density is increased [27]. Now, lowering λ leaves a long-range repulsion between like particles that is no longer compensated by a similar repulsion between unlike particles. As a result, we have now a system that exhibits a short-range attraction and long-range repulsion (SALR) between like particles, a well-known class of potentials characterized by inducing the presence of stable clusters (as well lamellar and bicontinuous) phases [31–33].

These effects of competing interactions on clustering and phase behavior have been know for a long time, e.g., in the study of protein and colloidal solutions [34]. For simple fluids, the presence of long-range repulsions

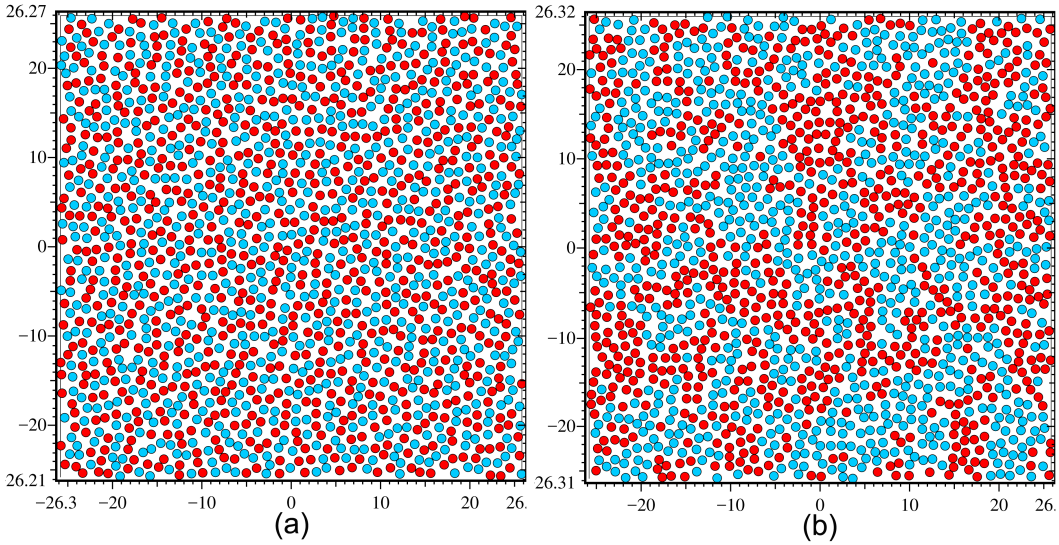

FIG. 2. Snapshots of Monte Carlo configurations of the NAHD two- component plasma with (a) the unlike Coulomb interaction turned off ($\lambda = 0$, left panel), and (b) identical Coulomb interaction between like and unlike particles ($\lambda = 1$, right panel). One sees the build up of mostly linear finite clusters of particles in the multihyperuniform case, i.e. when the unlike Coulomb interaction is turned off ($\lambda = 0$).

is known to inhibit the liquid-vapor transition in favor of the formation of a cluster phase [29, 35]. Our situation is completely analogous, with the uncompensated long-range Coulombic repulsion between like particles inhibiting the demixing transition, stabilizing the transient particle clusters that occur for $\lambda = 1$ close to the consolute point, and thus leading to the presence of a prepeak in $S_{ii}(Q)$. Moreover, these uncompensated Coulomb repulsions turn the system multihyperuniform, as seen in the low-Q behavior of $S_{ii}(Q)$ and $S(Q)$ depicted in Fig. 1. The clustering effect is readily seen in the snapshots shown in Fig. 2 for $\lambda = 1$ and $\lambda = 0$. Observe that the multihyperuniform configuration ($\lambda = 0$) is mostly dominated by the presence of linear particle clusters (chains) of finite size, in contrast with the more extended clusters found for $\lambda = 1$, which is only globally hyperuniform.

The multihyperuniform systems exhibit microsegregation with stable clusters, whereas for $\lambda = 1$ clusters correspond to transient states on the way to the complete phase separation that occurs once the total density is increased by a small amount. This microsegregation (or microheterogeneity) is characteristic of the presence of SALR-type interactions. The average chain-chain separation for $\lambda = 0$ can be estimated from the position of the prepeak in $S_{11}(Q)$, $Q_m \approx 2.3\sigma^{-1}$, as $2\pi/Q_m \approx 2.73\sigma$. Visual inspection of the configuration in Fig. is in accordance with this estimate. Basic energetic considerations can explain the compositional order of Fig. . When $u_{AB}^{LR} < u_{AA}^{LR} = u_{BB}^{LR}$, the simplest way to minimize repulsion between like particles is a chain-like configuration. At the same time, chains of A(B) particles would tend to be surrounded basically by chains of unlike, B(A) particles, with a smaller repulsion (just the hard-core when $\lambda = 0$).

These results are consistent with the cluster size distribution analyis presented in the Supplementary Material [28]. There one can appreciate the presence of a small cluster size maximum only in the multi-hyperuniform case. An additional maximum for percolating clusters in visible in both the $\lambda = 1$ and $\lambda = 0$ cases. Also, it is clear that the expected linear R-dependence of the local partial number fluctuation of the multihyperuniform system, in contrast with the non-monotonic behavior of its counterpart for $\lambda = 1$.

In summary, we have presented the first statistical-mechanical model that rigorously achieves disordered multihyperuniform many-body systems by tuning interactions in binary mixtures of non-additive hard-disk plasmas. Interestingly, multihyperuniformity competes with phase separation and stabilizes a clustered phase. We note here that despite the rather abstract nature of our interaction model, the results herein presented bear a considerable relevance in the general behavior to be expected if realistic effective interactions were to be designed so as to lead to disordered multi-hyperuniform equilibrium states. The extention of this work to multicomponent systems is straightforward and it will be the subject of a forthcoming publication.

The exploration of the potentially novel properties of multihyperuniform materials is a wide open research area. Standard disordered hyperuniform solids have already been shown to be of importance because they have novel photonic, phononic, electronic and transport properties [5, 11, 12, 15–17, 36], despite the lack of translational order. Using designer material techniques, examples have been fabricated in the laboratory on the microwave scale and their photonic properties have been studied [37]. Our work enables one to generate disordered

multihyperuniform dispersions, and then computationally determine their physical properties. Such promising designer materials could then be fabricated using 3D printing technologies.

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