High-density hyperuniform materials can be transparent: supplementary material

O. LESEUR, R. PIERRAT, AND R. CARMINATI

ESPCI Paris, PSL Research University, CNRS, Institut Langevin, 1 rue Jussieu, F-75005, Paris, France

*Corresponding author: remi.carminati@espci.fr

Published 14 July 2016

This document provides supplementary information to "High-density hyperuniform materials can be transparent," <http://dx.doi.org/10.1364/optica.3.000763>. It contains a brief description of the method used to generate hyperuniform point patterns, and a derivation of the relationship between the averaged structure factor and, respectively, the single-scattering diffuse intensity and the phase function. © 2016 Optical Society of America

<http://dx.doi.org/10.1364/optica.3.000763.s001>

1. METHOD TO GENERATE HYPERUNIFORM POINT PATTERNS

This section is dedicated to the presentation of the method used to generate 2D hyperuniform point patterns. The algorithm is adapted from Refs. [1–3]. Numerically we can only manipulate a finite number of points and to mimic an infinite medium, we divide the space into identical square cells of size L , each containing N points, as shown in Fig. S1. The system is supposed to be L -periodic and we only consider points in one unit cell.

Fig. S1. Periodic system considered to generate hyperuniform point patterns.

The structure factor for the full system is thus given by

$$S_{\text{tot}}(\mathbf{q}) = \lim_{R \rightarrow \infty} \frac{1}{(2R+1)^2} \frac{1}{N} \times \left| \sum_{n_x, n_y=-R}^R \sum_{j=1}^N \exp[i\mathbf{q} \cdot (n_x L \mathbf{u}_x + n_y L \mathbf{u}_y + \mathbf{r}_j)] \right|^2 \quad (\text{S1})$$

where $\mathbf{u}_x$ and $\mathbf{u}_y$ are unit vectors along the x and y axes respectively. This leads to the following expression

$$S_{\text{tot}}(\mathbf{q}) = \lim_{R \rightarrow \infty} \frac{1}{(2R+1)^2} \left| \frac{\sin[(R+1/2)q_x L]}{\sin(q_x L/2)} \right|^2 \times \left| \frac{\sin[(R+1/2)q_y L]}{\sin(q_y L/2)} \right|^2 S(\mathbf{q}) \quad (\text{S2})$$

where $\mathbf{q} = (q_x, q_y)$ and

$$S(\mathbf{q}) = \frac{1}{N} \left| \sum_{j=1}^N \exp(i\mathbf{q} \cdot \mathbf{r}_j) \right|^2 \quad (\text{S3})$$

is the structure factor of the unit cell. This expression implies that $S_{\text{tot}}(\mathbf{q}) = 0$ for all $\mathbf{q}$ such that q_x and q_y are not multiple of $2\pi/L$. Thus to get $S_{\text{tot}}(\mathbf{q}) = 0$ for all $\mathbf{q} \in \Omega$, we only have to require a vanishing of the last term for any $\mathbf{q} = (2\pi n/L, 2\pi m/L) \in \Omega$, that is

$$B(\mathbf{q}) = \left| \sum_{j=1}^N \exp(i\mathbf{q} \cdot \mathbf{r}_j) \right|^2 = 0 \quad \forall \mathbf{q} \in \Omega' \quad (\text{S4})$$

where Ω' is the ensemble corresponding to all $\mathbf{q} \in \Omega$ such that $\mathbf{q} = (2\pi n/L, 2\pi m/L)$, as shown in Fig. S2. $B(\mathbf{q})$ is always positive or zero. As a result, to make the structure factor vanishing in Ω' , $B(\mathbf{q})$ must reach its global minimum (i.e. zero) for all $\mathbf{q} \in \Omega'$. The simplest way to implement this is to define a potential ϕ by

$$\phi(\mathbf{r}_1, \dots, \mathbf{r}_N) = \sum_{\mathbf{q} \in \Omega'} B(\mathbf{q}) \quad (\text{S5})$$

and to minimize this potential. In the case of a square domain Ω with size K , this leads to the following expression for ϕ :

$$\phi(\mathbf{r}_1, \dots, \mathbf{r}_N) = \sum_{j=1}^N \sum_{l=1}^N \frac{\sin \left[(2P+1)\pi(x_j - x_l)/L \right]}{\sin \left[\pi(x_j - x_l)/L \right]} \times \frac{\sin \left[(2P+1)\pi(y_j - y_l)/L \right]}{\sin \left[\pi(y_j - y_l)/L \right]} \quad (\text{S6})$$

where $P = E[KL/(4\pi)]$.

Fig. S2. Ensemble Ω' of points where the structure factor of the elementary cell should vanish to get a hyperuniform point pattern.

2. RELATIONSHIP BETWEEN THE AVERAGE DIFFUSE INTENSITY AND THE STRUCTURE FACTOR IN THE SINGLE-SCATTERING REGIME

In this section, we derive the relationship between the average diffuse intensity and the structure factor in the single scattering regime. We consider a 3D geometry, but the derivation is essentially the same in 2D (and can be easily adapted). The average diffuse intensity is given by the difference between the average intensity and the intensity of the average field (ballistic component) through the relation

$$I_{\text{diff}}(\theta, \omega) = \left\langle |E_{\text{sca}}(\theta, \omega)|^2 \right\rangle - |\langle E_{\text{sca}}(\theta, \omega) \rangle|^2 \propto \left[\left\langle \left| \sum_{j=1}^N \exp[i\mathbf{q} \cdot \mathbf{r}_j] \right|^2 \right\rangle - \left| \left\langle \sum_{j=1}^N \exp[i\mathbf{q} \cdot \mathbf{r}_j] \right\rangle \right|^2 \right] |E_0|^2 \quad (\text{S7})$$

where $\mathbf{q} = \mathbf{k}_s - \mathbf{k}_i$. The first term corresponds to the average structure factor and is given by

$$\left\langle \left| \sum_{j=1}^N \exp[i\mathbf{q} \cdot \mathbf{r}_j] \right|^2 \right\rangle = N \langle S(\mathbf{q}) \rangle. \quad (\text{S8})$$

The second term is more complex. To have a more explicit expression, we have to consider the probability $P(\mathbf{r}) = V^{-1}$ of having a scatterer at position $\mathbf{r}$ which gives

$$\begin{aligned} \left| \left\langle \sum_{j=1}^N \exp[i\mathbf{q} \cdot \mathbf{r}_j] \right\rangle \right|^2 &= \left| \sum_{j=1}^N \langle \exp[i\mathbf{q} \cdot \mathbf{r}_j] \rangle \right|^2 \\ &= \left| \sum_{j=1}^N \int \exp[i\mathbf{q} \cdot \mathbf{r}_j] P(\mathbf{r}_j) d^3 \mathbf{r}_j \right|^2 \\ &= \frac{N^2}{V^2} \left| \int \exp[i\mathbf{q} \cdot \mathbf{r}] d^3 \mathbf{r} \right|^2. \end{aligned} \quad (\text{S9})$$

We now define Θ as

$$\Theta(\mathbf{r}) = \begin{cases} 1 & \text{if } \mathbf{r} \in V \\ 0 & \text{everywhere else.} \end{cases} \quad (\text{S10})$$

Thus Θ is a window function, the Fourier transform of which gives the second term in the average diffuse intensity. Finally, we obtain

$$I_{\text{diff}}(\theta, \omega) \propto N \langle S(\mathbf{q}) \rangle |E_0|^2 - \frac{N^2}{V^2} |\Theta(\mathbf{q})|^2 |E_0|^2 \quad (\text{S11})$$

which corresponds to Eq. (4) of the main text.

3. RELATIONSHIP BETWEEN THE PHASE FUNCTION AND THE STRUCTURE FACTOR

In this section, we derive the relationship between the phase function that enters the radiative transfer equation (RTE) [4] and the structure factor in a 3D geometry (the derivation can be easily adapted to 2D). We recall that the phase function corresponds to the radiation pattern of a single scattering event in a macroscopic sense (after averaging over disorder), and the RTE is a transport equation for the specific intensity (averaged over disorder as well). The RTE can be derived from first principles (Maxwell equations) [5, 6]. More precisely, the starting point of the derivation is the Bethe-Salpeter equation [7–9] which, in statistically homogeneous medium, reads as

$$\begin{aligned} \langle E(\mathbf{r}, \omega) E^*(\boldsymbol{\rho}, \omega) \rangle &= \langle E(\mathbf{r}, \omega) \rangle \langle E^*(\boldsymbol{\rho}, \omega) \rangle \\ &+ \int \langle G(\mathbf{r} - \mathbf{r}', \omega) \rangle \langle G^*(\boldsymbol{\rho} - \boldsymbol{\rho}', \omega) \rangle \Gamma(\mathbf{r}', \mathbf{r}'', \boldsymbol{\rho}', \boldsymbol{\rho}'', \omega) \\ &\times \langle E(\mathbf{r}'', \omega) E^*(\boldsymbol{\rho}'', \omega) \rangle d^3 \mathbf{r}' d^3 \mathbf{r}'' d^3 \boldsymbol{\rho}' d^3 \boldsymbol{\rho}'' \end{aligned} \quad (\text{S12})$$

where $\langle \dots \rangle$ denotes the ensemble average over the configurations of disorder, $\langle E(\mathbf{r}, \omega) E^*(\boldsymbol{\rho}, \omega) \rangle$ being the spatial correlation of the field and $\langle G(\mathbf{r} - \mathbf{r}', \omega) \rangle$ the average Green function. Γ is called the intensity vertex operator. It is a very complex object that contains all multiple scattering events that cannot be factorized when averaging over disorder. In particular, Γ contains the information on the spatial correlations of the disordered medium. From Eq. (S12), and considering a dilute system with

$k_0 \ell_B \gg 1$, one can derive the RTE, and in particular the expression of the phase function that reads

$$p(\mathbf{u}, \mathbf{u}') = A \tilde{\Gamma}(k_r \mathbf{u}, k_r \mathbf{u}', k_r \mathbf{u}, k_r \mathbf{u}', \omega) \quad (\text{S13})$$

where the constant factor A is such that $\int p(\mathbf{u}, \mathbf{u}') d\mathbf{u} = 4\pi$. $k_r = \sqrt{\Re[k_{\text{eff}}^2]}$ where $k_{\text{eff}} = k_0 + i/(2\ell)$ is the effective wave vector (i.e. the wave vector appearing in the wave equation for the average field). In a dilute system, we have $k_r \sim k_0$. $\tilde{\Gamma}$ is the reduced vertex operator in Fourier space, given by

$$\Gamma(\mathbf{k}, \mathbf{k}', \boldsymbol{\kappa}, \boldsymbol{\kappa}', \omega) = (2\pi)^3 \delta(\mathbf{k} - \mathbf{k}' - \boldsymbol{\kappa} + \boldsymbol{\kappa}') \times \tilde{\Gamma}(\mathbf{k}, \mathbf{k}', \boldsymbol{\kappa}, \boldsymbol{\kappa}', \omega). \quad (\text{S14})$$

In a dilute system, it is convenient to use a Taylor expansion of Γ with respect to the small parameter $[k_0 \ell_B]^{-1}$. In the case of a correlated medium (such as a hyperuniform material), the first two terms are needed, and the expansion is

$$\Gamma = \begin{array}{c} \bigcirc \\ | \\ \bigcirc \end{array} + \begin{array}{c} \bigcirc \\ - \\ \bigcirc \end{array} \quad (\text{S15})$$

where the first term corresponds to a scattering process for the field and its complex conjugate involving the same scatterer (solid line between both circles representing the scatterers, the upper row being related to the field and the lower row to its conjugate). The second term describes a scattering process involving two different but correlated scatterers (dashed line). Analytically, for subwavelength scatterers with polarizability $\alpha(\omega)$, we obtain

$$\Gamma(\mathbf{r}', \mathbf{r}'', \boldsymbol{\rho}', \boldsymbol{\rho}'', \omega) = \frac{N}{V} k_0^4 |\alpha(\omega)|^2 \delta(\mathbf{r}'' - \mathbf{r}') \delta(\boldsymbol{\rho}'' - \boldsymbol{\rho}') \times \left[\delta(\boldsymbol{\rho}' - \mathbf{r}') + \frac{N-1}{V} h(\boldsymbol{\rho}' - \mathbf{r}') \right] \quad (\text{S16})$$

where h is the pair correlation function [10]. This leads to the following expression for the phase function:

$$p(\mathbf{u}, \mathbf{u}') = \frac{AN}{V} k_0^4 |\alpha(\omega)|^2 \left\{ 1 + \frac{N-1}{V} h[k_0(\mathbf{u} - \mathbf{u}')] \right\}. \quad (\text{S17})$$

To get the relationship with the structure factor, we now explicitly write $\langle S \rangle$ in terms of the pair correlation function. We have

$$\begin{aligned} \langle S(\mathbf{q}) \rangle &= \frac{1}{N} \left\langle \left| \sum_{j=1}^N \exp[i\mathbf{q} \cdot \mathbf{r}_j] \right|^2 \right\rangle \\ &= 1 + \frac{1}{N} \left\langle \sum_{j=1}^N \sum_{\substack{k=1 \\ k \neq j}}^N \exp[i\mathbf{q} \cdot (\mathbf{r}_j - \mathbf{r}_k)] \right\rangle \\ &= 1 + \frac{1}{N} \int_{V^2} \sum_{j=1}^N \sum_{\substack{k=1 \\ k \neq j}}^N \exp[i\mathbf{q} \cdot (\mathbf{r}_j - \mathbf{r}_k)] \\ &\quad \times P(\mathbf{r}_j) P(\mathbf{r}_k) [1 + h(\mathbf{r}_j, \mathbf{r}_k)] d^3 \mathbf{r}_j d^3 \mathbf{r}_k \\ &= 1 + \frac{N-1}{V^2} \int_{V^2} \exp[i\mathbf{q} \cdot (\mathbf{r}_1 - \mathbf{r}_2)] \\ &\quad \times [1 + h(\mathbf{r}_1 - \mathbf{r}_2)] d^3 \mathbf{r}_1 d^3 \mathbf{r}_2 \\ &= 1 + \frac{N-1}{V} h(\mathbf{q}) + \frac{N-1}{V^2} |\Theta(\mathbf{q})|^2. \end{aligned} \quad (\text{S18})$$

From Eqs. (17) and (18), we finally obtain Eq. (5) of the main text:

$$p(\mathbf{u}, \mathbf{u}') \propto \langle S[k_0(\mathbf{u} - \mathbf{u}')] \rangle - \frac{N-1}{V^2} |\Theta[k_0(\mathbf{u} - \mathbf{u}')]|^2. \quad (\text{S19})$$

The appearance of the correction term in the right-hand side results from the definition of the phase function, that does not involve the same diagrams as the structure factor. In the phase function, or equivalently in the intensity vertex, we only take into account irreducible diagrams (i.e. diagrams that can not be factorized after the average process). In the structure factor, we have an additional diagram corresponding to two different scatterers that are not correlated, so that the expression of the averaged structure factor reads

$$\langle S \rangle = \begin{array}{c} \bigcirc \\ | \\ \bigcirc \end{array} + \begin{array}{c} \bigcirc \\ - \\ \bigcirc \end{array} + \begin{array}{c} \bigcirc \\ \cdot \\ \bigcirc \end{array}. \quad (\text{S20})$$

REFERENCES

1. O. U. Uche, F. H. Stillinger, and S. Torquato, "Constraints on collective density variables: Two dimensions," *Phys. Rev. E* **70**, 046122 (2004).
2. O. U. Uche, S. Torquato, and F. H. Stillinger, "Collective coordinate control of density distributions," *Phys. Rev. E* **74**, 031104 (2006).
3. R. D. Batten, F. H. Stillinger, and S. Torquato, "Classical disordered ground states: Super-ideal gases and stealth and equi-luminous materials," *J. Appl. Phys.* **104**, 033504 (2008).
4. S. Chandrasekhar, *Radiative Transfer* (Dover, New-York, 1950).
5. Y. N. Barabanenkov and V. M. Finkel'berg, "Radiation transport equation for correlated scatterers," *Sov. Phys. JETP* **26**, 587 (1968).
6. S. M. Rytov, Y. A. Kravtsov, and V. I. Tatarskii, *Principles of Statistical Radiophysics*, vol. 4 (Springer-Verlag, Berlin, 1989).
7. P. Sheng, *Introduction to Wave Scattering, Localization and Mesoscopic Phenomena* (Academic Press, New York, 1995).
8. L. A. Apresyan and Y. A. Kravtsov, *Radiation Transfer: Statistical and Wave Aspects* (Gordon and Breach Publishers, Amsterdam, 1996).
9. E. Akkermans and G. Montambaux, *Mesoscopic Physics of Electrons and Photons* (Cambridge University Press, Cambridge, 2007).
10. S. Torquato and F. H. Stillinger, "Local density fluctuations, hyperuniformity, and order metrics," *Phys. Rev. E* **68**, 041113 (2003).