Experimental evidence for transparency, band gaps and Anderson localization in two-dimensional hyperuniform disordered photonic materials

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The recent prediction of full photonic band gaps in amorphous dielectric materials [1–4] is surprising since it defies the picture of bandgap formation by reflections and interferences from Bragg planes in a periodically repeating environment [5]. Numerical work on stealthy hyperuniform solids also suggested that disordered photonic materials can display additional transport regimes: the same material can show transparency, photon diffusion, Anderson localization, or a full bandgap, depending on the frequency ν of the electromagnetic wave [6]. Here we demonstrate experimentally, using microwaves, that the density of states and the different transport properties can be observed and quantified in a two-dimensional hyperuniform disordered array of cylinders with high dielectric permittivity. Interestingly, we find a second weaker bandgap at higher frequencies, which can be linked to higher-order spatial correlations and the second peak in the structure factor. Our results emphasize the importance of spatial correlations for the formation of photonic band gaps [3].

In analogy to electronic semiconductors, dielectric materials in a periodic, quasiperiodic, or amorphous configuration can all display full band gaps [1–5,10]. For the latter materials, due to the absence of long range order, the band gap has been associated with local resonances of the scatterers or correlated scattering clusters, which is reminiscent of the tight-binding model in electronic semiconductors [11]. In contrast to electrons, however, there exist no bound photon states making this analogy questionable. Other proposals have linked the opening of a gap directly to the suppression of density fluctuations on large length scales, known as stealthy hyperuniformity (SHU) [12]. While the precise origin of a bandgap in an amorphous dielectric material is yet unknown, the transport properties inside the gap are well understood [3,5,13]. In both periodic and non-periodic bandgap materials, an incident light wave enters by a finite distance L_B, called the Bragg length, and is then totally reflected. For a slab of thickness L, the wave can tunnel through the material with a probability T ∼ e−L/L_B. However, outside the gap, the transport properties differ strongly. Photonic crystals either reflect, diffract into Bragg peaks, or they are transparent, which is a direct consequence of long-range order and the corresponding sharp Bragg maxima in the structure factor S(k). The situation is entirely different for amorphous materials, which scatter light strongly over a broad range of k. Recent numerical work has revealed that this leads to a rich transport phase diagram for amorphous bandgap materials—with regions of transparency, Anderson localization, and light diffusion—not present in ordered materials [4]. In contrast to disordered photonic crystals, discussed for example in the celebrated article by Sajeev John in 1987 [6], the diffuse scattering and localization observed outside the gap is not a consequence of imperfections, but an inherent feature of the amorphous material [3]. Introduced in 2003 [14], stealthy hyperuniformity provides an elegant way to construct such idealized disordered materials with finely tunable correlations encoded by the degree of stealthiness χ, ranging from 0 → 0.5 before the onset of crystallisation [14].

Thirty years after John’s seminal work on the interplay between photonic band gap formation and strong localization in disordered dielectric lattices [6], a controlled experimental study of the optical transport properties in between ordered and disordered states of matter is still lacking. Here, we present experimental results obtained for a 2D system composed of high index dielectric cylinders in air [15] placed according to SHU point patterns [1]. To probe the different transport regimes experimentally, we conduct measurements in the microwave regime. Our samples consist of about N ≈ 200 cylindrical scatterers (dielectric permittivity ε ≃ 36, radius r = 3 mm, height h = 5 mm; the Mie scattering efficiency of such a cylinder is shown in Supplementary information, Fig. S1) placed in an aluminum 2D cavity (50 × 50 × 0.5 cm²) on a SHU point pattern (on a square of size of approximately 25 × 25 cm²) generated by simulating an annealing relaxation scheme [3] (see Fig. 1(a)). We perform measurements on five different configurations χ = 0.15, 0.25, 0.30, 0.40 and a triangular lattice. For all the samples studied, we kept the number density constant (ρ = 0.3136 cm⁻²). The point patterns and the structure factors of the samples are shown in the supplementary Fig. S2. The cavity can be considered as two-dimensional for the microwaves frequencies ν < 10 GHz studied. Under this condition, only the first transverse magnetic mode, TM₀, exists in air: the electric field is perpendicular to the plane, and the field amplitude is uniform over the cavity height [16]. We mimic an infinite 2D system by placing absorbing carbon loaded
polyurethane foam (LS-14 from Emerson & Cuming) between the sample and the metallic walls of the cavity. We raster the cavity with a mobile antenna that is inserted by a robotic arm through holes drilled into the upper plate with a diameter 2mm, on a $5 \times 5 \text{mm}^2$ grid unit cell. Considering the sample size, and the fact that we are not able to penetrate the cavity at the holes above the scatterers, we end up with about $\sim 2700$ measured positions. The antenna is weakly coupled to the field because it extends into the cavity by 1mm whereas the height of the cavity is 5mm. This allows us to minimize the perturbation of the field induced by the monopole antenna at different positions in the cavity [17].

At each grid point $(x, y)$, we measure the complex transmission spectrum $S_{12}(\nu)$ between a fixed antenna (1) placed at the center of the cavity and the mobile antenna (2) using a vector network analyzer or VNA (Agilent E5071C). For the central antenna, we use a higher penetration depth of about 4mm to improve the signal to noise ratio. When placing the ensemble of scatterers, we make sure to maximize the nearest distance between the central antenna and the neighbouring scatterers. A scan of the entire grid takes around 75 hours. Figure 1(b) shows examples of measured spectra $|S_{12}(\nu)|^2$ between the central position 1 and probe position 2 for different $\chi$-values and for different distances $d$ between the antennas. The small transmission values of order $10^{-5}$ or less are because the receiving antenna is weakly coupled to the cavity. The measured spectra consist of a superposition of peaks which are associated to the resonances of the system. We extract their frequency, complex amplitude and width using harmonic inversion as described in ref. [18, 19]. The amplitude of each peak differs from one position to the other and from this we obtain an electric field amplitude map $E_{\nu}(x, y)$ of an eigenmode (see Methods) [20, 21].

In Fig. 2, we plot a histogram of the frequencies of the modes, which is directly proportional to the density of states (DOS). We compare the results for SHU point patterns with different values of $\chi$, to the results obtained for a triangular lattice. As shown in earlier work the triangular lattice is the champion photonic crystal structure.
in 2D, with a gap slightly larger than that of an ordered hyperuniform structure [3, 12]. Our experimental data confirms the two first TM photonic crystal bandgaps predicted by theory [5]. We also find frequency windows without states for the SHU disordered systems. Surprisingly, the second bandgap is also present in the $\chi = 0.4$ disordered system. This finding is in contradiction to previous claims about the origin of bandgaps in disordered photonic materials [14, 22, 23].

To corroborate additional evidence for this interesting observation, we performed band structure calculations, using the same parameters as in the experiment. These numerical data confirm the existence of a second-order bandgap for $\chi > 0.4$, see supplementary Fig. S4. Both the first and the second gap approximately match the Bragg scattering maximum of $S(k)$ of the triangular lattice as shown in supplementary Fig. S2 (supporting earlier proposals, that short-range spatial correlations play a key role for the opening of bandgaps in amorphous photonic materials [3]. Experimentally, we observe a narrow photonic bandgap even for our most disordered sample ($\chi = 0.15$). Our numerical data for a large ensemble of system realizations, however, suggest that the bandgap closes for $\chi \lesssim 0.3$ and reduces to a pseudo-gap with a small but finite density of states. Naturally, variations between different realizations of hyperuniform materials become more pronounced for smaller values of $\chi$ (see supplementary Fig. S5) and moreover the number of states per frequency bin is small for a finite sized system. This can lead to the situation that the central frequency and width of the bandgaps depend on the precise realization of the point pattern, which is a distinct feature of disordered materials not found in crystals. For larger values of $\chi$ these variations are suppressed, and the gap becomes more robust against statistical fluctuations.

We now consider the optical properties of our material outside the gap [4]. The electric field amplitude maps $E_\nu(x, y)$, shown in the first line of Fig. 3 reveal the striking variations in optical transport properties across the spectral range covered by our experiment. Note that we plot the signed amplitude $E_\nu^\pm(x, y) = \text{sgn} \left( \text{Re}(S_{12}) \right) |S_{12}|$, where $S_{12}$ is the transmission deduced from $S_{12}$ after the ad hoc rotation of the global phase making the real and imaginary parts statistically independent [24]. This allows to represent both the real and imaginary parts of the eigenmodes on the same map. At low frequencies, we observe simple square cavity modes as if the medium was homogeneous, which is a remarkable result given the fact that at $\nu \sim 2$ GHz the Boltzmann mean free path $\ell_s(\nu)$ of the cylinder ensemble is almost two orders of magnitude smaller than the system size $L = 25$ cm. In the second line of Fig. S1 with $\ell_s(\nu) = [\sigma_s(\nu) \rho]^{-1}$ given by the total scattering cross section $\sigma_s(\nu)$ and the number density $\rho$. An alternative way to study wave propagation in the SHU material is to monitor the wave emitted by the central antenna as it propagates through the medium in the time domain. By calculating the real part of the Fourier-transform of $S_{12}(\nu) \cdot F_{f_0, \Delta \nu}(\nu)$ (with $F_{f_0, \Delta \nu}$ a bandpass filter of bandwidth $\Delta \nu$ centered around $f_0$) at all points on the lattice, we reconstruct movies of the propagating electromagnetic fields as a function of time for the selected bandwidth $\Delta \nu$. Individual frames of the movies are shown in Fig. 3(f-j) and the entire movies are included in the supplementary material (see Videos S6-12). From the velocity of the circular wave in the medium we can derive the effective refractive index of the samples and find $n_{\text{eff}} \sim 1.8$. Equally, counting the nodal lines of the modes and relating them to their frequencies, we obtain values of the effective refractive index of the metamaterial in the range $n_{\text{eff}} = 1.7 \pm 0.3$. The uncertainty is due to the fact that, for disordered systems, the cavity size is not well defined and moreover, we observe a slight increase of $n_{\text{eff}}$ from $\nu = 1 \to 3$ GHz. For comparison, the Maxwell-Garnett effective refractive index, which in 2D corresponds to the square of the surface averaged permittivity, is $n_{\text{MG}} = 2.05$.

Torquato and coworkers named their designer materials ‘stealthy’ hyperuniform because they predicted them to be fully transparent below a threshold frequency $\nu < \nu_c$ [14, 25]. The latter is tantamount to saying that the actual scattering mean free path $\ell$ becomes infinite, and thus $L/\ell = 0$, while $\ell_s < L$ can remain arbitrarily small as discussed earlier. In the first-order or single-scattering approximation this yields $\nu_c = \frac{\Delta \nu}{n_{\text{eff}}}$ [4]. For our system parameters, the theoretical $\nu_c$ range from $\sim 2.2$ GHz ($\chi = 0.15$) to $\sim 3.0$ GHz ($\chi = 0.4$) based on an effective refractive index of $n_{\text{eff}} \sim 1.8$. Leseur et al. demonstrated recently that stealthy transparency is also robust against recurrent multiple scattering [26]. They establish a criterion for transparency, $L/\ell_s \ll k\ell_s$, in a dense SHU disordered material composed of dipolar point scatterers. While transparency is retained under this condition it also implies that the transition at $\nu_c$ is not sharp but system size dependent. From a theoretical evaluation of $\sigma_s(\nu)$ for our $\varepsilon = 36$ cylinders in air, however, we find that only for $\nu < 1$ GHz the condition $L/\ell_s < k\ell_s$ is met, supplementary Fig. S1. The experimental results, however, suggest that the condition set by Leseur et al. is too restrictive and transparency remains a robust feature for $\nu < \nu_c$ in our dense, high index SHU materials, even for $k\ell_s \lesssim 1$ (see also supplementary Fig. S7).

For frequencies $\nu > \nu_c$ transparency is clearly lost and we observe scattering and wave diffusion. The modes become disordered, Fig. 3(b), and the propagating wavefronts in the time domain are highly distorted signaling mean free paths smaller than the system size, Fig. 3(g). A closer inspection of the propagating wavefronts, supplementary Fig. S7, illustrates how the onset of scat-
Next, we calculate the Thouless conductance $g_{Th} = \delta \nu / \Delta \nu$, which is a fundamental localization parameter \cite{27, 29}. Thouless argued that in the Anderson localization regime, the dimensionless ratio $g_{Th} = \delta \nu / \Delta \nu$ falls below unity. In this case, the spectral widths $\delta \nu$ of the modes is smaller than their spacing $\Delta \nu$, and the modes are isolated \cite{27}. In the opposite limit, for $g_{Th} \geq 1$ modes overlap and waves can propagate. By calculating the average width of the modes in each frequency bin, Fig. 2, we extract mean Thouless conductance for each frequency bin shown in Fig. 4. We have marked the data points directly at the band-edges by open circles in Fig. 4. Due to the discretization, their values can be affected by the zero’s in the DOS in the gap. Inside the bandgap there are no modes and $(g_{Th})$ is not defined. We find values of $(g_{Th}) \sim 1$ everywhere except in the vicinity of

![Diagram](image_url)

Figure 3. Electromagnetic field distribution of the eigenmodes and wave transport in the time domain for a sample with $\chi = 0.30$ ($\nu_c = 2.88$ GHz). (a-e): Signed amplitudes of selected eigenmodes at different characteristic frequencies. (a) cavity mode, (b) diffusive mode, (c) dielectric localized mode, (d) air localized mode and (e) diffusive mode. (f-j): Maps of the electric field for wave transport at different times $t_1, t_2, t_3$ and for different central frequencies $f_0$. The wave—a Gaussian pulse centered at $f_0$ and having a width of 0.5 GHz in the frequency domain—is emitted at the center of the maps, and its temporal representation is shown in the last line ($\Re[\tilde{F}_{f_0,\Delta \nu}(t)]$ is the real part of the Fourier transform of the Gaussian bandpass filter). The colored vertical lines indicate the time of each frame shown $t_1, t_2, t_3$. Entire videos are included in the supplementary material. The color scale is adjusted for each individual panels.
Figure 4. Thouless conductance for different degrees of stealthy hyperuniformity $\chi$ between 0.15 and 0.40. The curves are shifted by a factor 10 for clarity. The hatched areas show the width of the experimentally observed bandgaps for each value of $\chi$ using the same colors.

The gap where $\langle g_{Th} \rangle$ drops by up to two orders of magnitude, signaling localization. This result is consistent with both the finite spatial extension of the modes we observe experimentally, see Fig. 3(c,d), and the localization of the propagating wave in the same frequency domain, Fig. 3(h). In the low-frequency regime, the Thouless conductance is close to one, and wave transport expands over the whole system size.

In conclusion, we show experimentally that disordered dielectric structures display different characteristic transport regimes such as transparency, photon diffusion, Anderson localization, as well as first and even second order bandgaps. We rationalize our findings by analyzing the mode structure and the propagation of waves in the time domain. We find evidence that transparency is robust against recurrent multiple scattering, and that the stealthy materials we study retain their low-frequency transparency even for the unusually strong refractive index mismatch between our scatterers and air $\sqrt{\varepsilon/\varepsilon_{\text{air}}} = 6$.

Our results lend support to recent numerical predictions and shed new light on the interplay between disorder and correlations [4]. We believe this will have significant consequences for the design of photonic materials, such as two-dimensional nanostructured materials for light harvesting in solar cells [30] or light guiding in all-optical circuit applications [31].

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Author contributions

G.A., L.S.F., F.S. and F.M. conceived the study. G.J.A. and F.M. carried out all experiments with contributions by U.K. and O.L. Data analysis was done by G.J.A. with input from all authors. Numerical simulations were carried out by L.S.F. The manuscript was written by G.J.A. and F.S. with contributions from all authors.

Methods

Mode analysis with a clustering algorithm. Ideally, resonances belonging to the same mode should all have the same frequency. In practice, the presence of the mobile antenna at every point \((x, y)\) shifts the resonant frequency by a small amount depending on the intensity of the electromagnetic field at the specific position \(\nu(x, y)\). To associate each resonant signal at position \((x, y)\) to a specific mode, we apply a semi-supervised clustering algorithm. This allows us to identify every single mode of the disordered cavity, associated with discrete resonance frequencies, as long as the mode amplitude is large enough to be detected by the VNA. We determine the frequencies \(\nu^i\), widths \(\gamma^i\) and complex amplitudes \(A^i\) of each resonance \(i = 1, \ldots, N\) using the harmonic inversion method described in ref. [18, 19]. In supplementary Fig. S3 we show a subset of the resonance frequencies for all points \((x, y)\) for two different frequency intervals. We see that the points form clusters in this \((x, y, \nu)\) space. We identify all data points belonging to a certain cluster by using a density-based clustering algorithm fulfilling the condition that two points having the same coordinate \((x, y)\) cannot be in the same cluster, see also supplementary materials section [11] and Fig. S3.

Pulsed wave propagation in the time domain. We obtain time domain propagation signals from the real part of the Fourier transform of the complex transmission spectra multiplied by a chosen bandpass filter centered at \(f_0\) with a standard deviation \(\Delta f\). We use a Gaussian bandpass filter to avoid window effects in the Fourier transform. The excitation in the time domain is therefore a Gaussian pulse with a temporal spread inversely proportional to \(1/\Delta f\) of the Gaussian bandpass filter. Section [1] provides links to the videos of the waves propagating inside the medium for different central frequencies.
SUPPLEMENTARY INFORMATION

I. BOLTZMANN SCATTERING MEAN FREE PATH.

In Fig. S1 we show the scattering efficiency $Q$ of an individual cylinder in TM polarization calculated using Mie theory [32] and the corresponding Boltzmann scattering mean free path $\ell_s(\nu)$ with a total scattering cross section $\ell_s(\nu) = [\sigma_s(\nu)\rho]^{-1}$ and $\sigma_s(\nu) = \pi r^2 \chi$.

II. POINT PATTERNS AND THEIR STRUCTURE FACTORS.

Figure S2(a) shows the point patterns of the samples studied in this study, and Fig. S2(b) the corresponding average structure factors of 1000 samples generated as the ones used in this study.

III. CLUSTERING PROCEDURE

To identify the modes, see Methods section and main text, we use a slightly modified version of the C-DBSCAN algorithm published in Ref. [21]. In our version, step 2 of the algorithm [21] either labels the points in the KD-tree leaf as noise ratio (if the density is too small), or we create a local cluster for each point in the leaf.

Depending on the frequency range, we run our modified version of C-DBSCAN either in the $(x, y, \nu)$, $(x, y, \nu, \gamma)$ or $(x, y, \nu, \gamma,\ln A)$ space to reach the best clustering results. An example of the result is shown in Fig. S3 where the different clusters, or modes, found by the algorithm are plotted using different colors.

IV. NUMERICAL SIMULATIONS OF THE DOS

Figure S4 shows the normalized density of states (nDOS) of the stealthy hyperuniform samples obtained numerically for a large statistical ensemble of point pattern and using periodic boundary conditions. The properties of the dielectric cylinders and their density are identical to the system studied in the experiment. The nDOS was calculated using the MIT Photonic Bands [33] software using the supercell method [3] as described earlier in ref. [3]. This dataset was obtained by calculating 500 different samples for each $\chi$-value (between 0.1 and 0.5, every 0.05).

Figure S5 shows the average and the standard deviation of the gap central frequency and width found for the samples used in Fig. S4. The statistical variations are large at low and intermediate $\chi$-values (between 0.10 and 0.35). At large $\chi$-values ($\geq 0.4$), the standard devi-
V. TIME DOMAIN PROPAGATION VIDEOS

A. Low frequency wave transport: transition from the stealth to the scattering regime

Videos S6-1, 2 and 3 show the propagation of the wave in the low frequency regime (well below the gap frequency $\nu_G \approx 5$ GHz. We observe that for frequencies $\nu < \nu_c$ and at early times, the spherical wave structure is well preserved, indicating the absence of scattering. This boundary between the stealth regime and the diffusive regime is also shown in more detail in Fig. S7. The panels in the green shaded polygon indicate that the Gaussian pulse central frequency $f_0$ is below the critical stealth frequency $\nu_c = \frac{c}{\pi n \sqrt{\rho \chi}}$, and above $\nu_c$ elsewhere. By eye, we see a clear correlation between the wave front smoothness and the transition from the stealth regime to the diffusive regime for frequencies $\nu > \nu_c$. Since $\nu_c \propto \sqrt{\chi}$ the transition is shifted to higher frequencies when increasing the degree of stealthiness $\chi$. Note that the wave distortion at later times (in the videos) is explained by reflections of the wave on the non-ideal absorbing foam walls.

Video S6-4 (respectively S6-6) shows the electromagnetic field for a Gaussian pulse centered 0.25 GHz below (resp. above) the band gap and having a width $\Delta \nu = 0.25$ GHz. Video S6-7 shows the propagation of the wave in the high frequency regime, well above the first band gap. As in the low frequency regime for frequen-
cies above $\nu_c$, we observe a strong scattering and wave diffusion.

Finally, video S6.5 shows the electromagnetic field in the band gap. For this video, the bandpass filter was chosen to be a square filter fitting exactly the bandgaps as extracted from Fig. 2. This explains the windowing effect seen in the input signal.
1. Stealth regime (Gaussian bandpass filter, \( f_0 = 1.75 \text{ GHz}, \Delta \nu = 0.25 \text{ GHz} \))
2. Stealth regime (Gaussian bandpass filter, \( f_0 = 2.25 \text{ GHz}, \Delta \nu = 0.25 \text{ GHz} \))
3. Wave diffusion (Gaussian bandpass filter, \( f_0 = 3.5 \text{ GHz}, \Delta \nu = 0.25 \text{ GHz} \))
4. Dielectric Anderson localized modes just below the bandgap (Gaussian bandpass filter, \( \Delta \nu = 0.25 \text{ GHz} \))
5. Square filter in the bandgaps
6. Air Anderson localized modes just above the bandgap (Gaussian bandpass filter, \( \Delta \nu = 0.25 \text{ GHz} \))
7. Wave diffusion (Gaussian bandpass filter, \( f_0 = 6.5 \text{ GHz}, \Delta \nu = 0.25 \text{ GHz} \))

Figure S6. (Supplementary information) Videos links

Figure S7. (Supplementary information) Maps of the electric field amplitude for the propagation of a pulse of spectral width \( \Delta \nu = 0.125 \text{ GHz} \) at different central frequencies \( f_0 \) (for details see text and Fig. 3), and first half of the Gaussian pulse used for the excitation. The frames shown in the figure are taken at the time marked by the blue vertical line. The panels in the green polygon indicate frequencies below \( \nu_c(\chi) \). The radius of the dashed circles indicate the place where a wave emitted at the time marked by the red vertical line should be at the time marked by the blue vertical line, for a homogeneous medium with \( n_{\text{eff}} = 1.8 \). The color scale is adjusted for each individual panels.