Light transport and localization in two-dimensional correlated disorder

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Multiple light scattering in disordered media plays a paramount role in the study of complex natural systems (e.g., biological tissues, porous materials, planetary atmospheres) [1] and wave phenomena (e.g., light localization, anomalous diffusion) [2–4]. In recent years there has been a growing interest in the use of photonic structures with controlled disorder, in particular within the context of mesoscopic transport effects [5–8], cavity quantum electrodynamics [9], photon management for energy efficiency [10–13] and even lab-on-chip spectroscopy [14]. Indeed structural correlations in the positions of scatterers are known to affect light propagation. Previous studies have shown that short-range correlations can either diminish or enhance the scattering strength of a disordered system [15–18] and lead to a modulation of the density of optical states [19], even in biological systems [20]. Such a modulation can be so large that a complete photonic bandgap is expected to form, even without long-range periodicity [19, 21–24]. The emerging concept of “disorder engineering” to manipulate light transport in random media is, however, still in its infancy and little is known so far on the occurrence of localization phenomena in correlated systems.

In this article, we theoretically investigate the transport of light and the occurrence of localization in two-dimensional (2D) photonic structures possessing short-range correlated disorder. A semi-analytical model describing the wave propagation in correlated-disordered systems allows us to investigate how key quantities, namely the transport mean free path, the scattering anisotropy factor and the localization length, evolve with the degree of correlation. In particular, short-range correlations are found to allow for the tuning of the localization length over several orders of magnitude and thus, make it possible to go from a quasi-extended to a strongly localized regime very easily, in sharp contrast with three-dimensional systems, where the localized regime is very difficult to reach [8, 23]. This trend is confirmed by numerical simulations.

The 2D photonic structures consist of disordered patterns of circular air holes ($n_i = 1$) with filling fraction $f = 20\%$ and diameter $\phi = 0.23 a$, where $a$ is the period of a hexagonal lattice of holes with the same $f$, in a medium with refractive index $n_o = 3.5$ [10, 19]. The short-range correlation in the disorder is controlled by imposing a minimum distance $d_{\text{min}}$ between the centers of the holes. This has been obtained [10, 20] by generating a disordered packing of hard disks with diameter $d_{\text{min}}$ at a packing fraction $p$, using a freely-available code [27] based on the Lubachevsky-Stillinger algorithm [28]. The centers of these disks have been used to generate the point patterns shown in Fig. 1(a) for three photonic structures with $p = 30, 50, 70\%$. The degree of local order evidently increases with increasing $p$, imposing an average distance between adjacent holes.

Fundamentally, the existence of a typical distance between neighboring scatterers implies a certain phase relation which, depending on direction and wavelength of scattered waves in the medium, gives rise to either constructive or destructive interference between them. Following this line of reasoning, structural correlations can be seen as a modification of the angular scattering pattern of the individual scatterers, and be taken into account (to a first approximation) by correcting the expression of the single scatterer differential scattering cross-section $d\sigma/d\theta$ by the static structure factor $S(q)$ as [15, 16]

\[
\frac{d\sigma^*}{d\theta} = \frac{d\sigma}{d\theta} S(q),
\]

where $q = (4\pi/\lambda_e) \sin(\theta/2)$, $\theta$ is the scattering angle and $\lambda_e = \lambda/n_e$ is the wavelength in a medium with effective refractive index $n_e$, which in our case equals 2.92 according to the two-dimensional Maxwell Garnett mix-
Structural correlations in 2D disordered media – (a) Disordered photonic structures with filling fraction $f = 20\%$ and different degrees of correlation ($p = 30, 50, 70\%$). (b) Structure factors $S(qd_{\text{min}})$ of the correlated patterns, evaluated numerically with Eq. 2 (hollow dots) and semi-analytically using the Baus-Colot model (solid lines).

Formally, the structure factor $S(q)$ is defined as

$$S(q) = \frac{1}{N} \sum_{i,j=1}^{N} e^{-iq(\mathbf{r}_i-\mathbf{r}_j)}$$

where $N$ is the number of scatterers, $\mathbf{r}_{i,j}$ the position of the scatterers labelled $i$ and $j$, and $(\cdots)$ denotes ensemble average. In previous works on 2D photonic structures with short-range correlated disorder (statistically isotropic), $S(q)$ was calculated numerically from the point patterns generated by a sphere packing protocol [29]. This approach is very time consuming and not suited to an exhaustive study of the effect of structural correlations on transport. An analytical expression of the structure factor of a correlated-disordered medium is often obtained by making use of the well-known Percus-Yevick model which, unfortunately, applies exclusively to systems with odd dimensionality ($d = 1, 3, \ldots$) [31]. In contrast, we adopt a semi-analytical approach, based on the Baus-Colot (BC) model for the structure factor of a fluid of hard disks [32], that is well-suited to systems of dimensionality $d = 2$. In Fig. 1(b), we compare the structure factor $S(qd_{\text{min}})$ of the BC model, using $p$ as the only input parameter. A very good agreement is observed, even for high $p$. As the degree of correlation is increased, the structure factor exhibits stronger oscillations, which indicate the emergence of a typical distance between neighbouring scatterers.

By making use of Eq. 1 we calculate the angularly and spectrally-resolved “effective” differential scattering cross-section $d\sigma^*/d\theta$ of holes in TE-polarization (electric field in the plane) as a function of the degree of short-range correlation (Fig. 2(a)). The single scatterer differential scattering cross-section $d\sigma/d\theta$ was calculated from Mie theory for circular cylinders [33]. As $p$ increments, $d\sigma^*/d\theta$ exhibits increasingly sharper features in frequency and angle due to the oscillations of $S(q)$, giving considerably different weights to the forward and backward scattering. Clearly, for low $p$ the scattering is primarily forward, whereas for strongly-correlated disorder the forward scattering is inhibited in a broad range of frequencies.

This redistribution of the scattered light is at the core of the modification of the transport properties in correlated disorder. To illustrate this point, we calculate the transport mean free path $\ell_t$ in the correlated system [31]

$$\ell_t = \left( \rho \int_0^\pi \frac{d\sigma^*}{d\theta} \left(1 - \cos \theta \right) d\theta \right)^{-1},$$

where $\rho$ is the number density of scatterers, and the scattering anisotropy factor $g$

$$g = \frac{1}{\sigma^*} \int_0^\pi \frac{d\sigma^*}{d\theta} \cos \theta d\theta,$$

which indicates the degree of anisotropy of the effective single scattering event. The results are shown in Fig. 2(b-c) as a function of $p$. First, as expected, correlations yield spectral ranges with either longer or shorter transport mean free paths, the latter occurring in particular when $\lambda_c = 2n d_{\text{avg}}$, with $n = 1, 2, \ldots$ (Bragg-like scattering) and $d_{\text{avg}}$ is the average distance between nearest-neighbour scatterers. Variations larger than a factor of 2 are observed. Second, the anisotropy factor $g$ for highly correlated structures becomes negative on broad frequency ranges, reaching values as low as -0.9, indicating a strong backward scattering [17]. Interestingly, this leads to a peculiar light transport in which the scattering mean free path $\ell_s = \ell_t (1 - g)$ is longer than the transport mean free path $\ell_t$. This scattering property is rare in systems of isolated particles, and it has been observed only in specific cases [35].

Gaining control over the transport mean free path provides an unprecedented control on light localization phenomena. In this respect, two-dimensional structures are very peculiar since the dependence of the localization length on the transport mean free path is critical, so that a small change in $\ell_t$ should yield dramatic changes of $\xi$. The localization length is indeed predicted to be given by [34]:

$$\xi \approx \ell_t \exp \left[ \pi^2 \frac{\ell_t}{\lambda_c} \right].$$

We therefore expect that a modification of the degree of correlation $p$ could lead to modifications of the localization length $\xi$ over orders of magnitude, making it possible...
to go from a quasi-extended to a localized regime easily in finite-size systems. In Fig 2(d), the localization length $\xi$ predicted semi-analytically is shown in semi-log scale. The strong modulation of $\xi$ as a function of frequencies is striking. In the 2D correlated system, one goes from a regime in which $\xi$ is much larger than any realistic system (low frequencies) to a regime in which the two can be comparable (at $a/\lambda \approx 0.2$). Although we do not expect Eq. 5 to be quantitatively accurate, this huge photonic dispersion (variation of orders of magnitude within $\Delta \omega/\omega_0 = 0.2$), suggests that we could truly observe a dramatic variation of $\xi$ in real systems.

To test these predictions, we investigated the transport properties of the 2D correlated systems through numerical 2D finite-difference time-domain (FDTD) simulations, using a freely available software package [37]. We considered finite systems with the same structural parameters and open boundaries (squares of side $L = 36a$ surrounded by perfectly matched layers, see Fig. 2(a)). The system was excited from a set of 225 randomly distributed dipole sources having an impulse with bandwidth of $0.02 \lambda/a$. Since the system is open, the energy density is expected to decay exponentially at long times as $U(t) \propto \exp(-\gamma t)$, where $\gamma$ is the decay constant. A change in the degree of correlation $p$ should yield a change in $\ell_t$ and thus in the average time needed for light to escape from the system. This is illustrated in Fig. 2(b), where, at a frequency $a/\lambda = 0.21$, increasing $p$ yields a diminution of $\gamma$. The multi-exponential decay at shorter times is due to the excitation of several modes in the structure which couple to the environment with different efficiency. The decay constants $\gamma$ were therefore obtained from exponential fits at sufficiently long times for various frequencies and degrees of correlation $p$, and an average was performed over 6 disorder realizations. Note that a proper statistical analysis of $\gamma$ would require more disorder realizations, which would be extremely time-consuming. We have observed, however, that 6 disorder realizations are sufficient to show the increase of the decay constant as the degree of correlation increases, as reported below (Fig. 2(d)).

In Fig. 2(c), we show the average decay constants of the photonic structures with $p = 30\%$, $50\%$ and $70\%$ as a function of the pulse excitation frequency, estimated from the numerical FDTD simulations. The effect of correlations on light transport is particularly clear. At frequencies close to $a/\lambda \approx 0.2$, $\gamma$ is strongly diminished due to a reduction of $\ell_t$ and at lower frequencies ($a/\lambda < 0.17$) one observes an increase of $\gamma$, in accordance with the increase of $\ell_t$ (see Fig. 2(b)). Note that $\gamma$ drops over 2 orders of magnitude within a relative bandwidth of $\Delta \omega/\omega_0 = 0.2$.

It is also interesting to compare the values obtained numerically with those expected from the semi-analytical approach within the diffusion approximation. The decay constant for diffuse light in a 2D system open along two directions is given by

$$\gamma = \frac{2\pi^2 D}{(L + 2z_c)^2},$$

where $z_c = \frac{\pi}{2} \ell_t$ is the so-called extrapolated length [38, 39] (internal reflections are neglected), and $D = v_c \ell_t/2$ is the diffusion constant with $v_c = c/n_e$ the energy velocity. According to Eq. 5 the optical thickness $L/\ell_t$ of our systems can be extremely small at very low frequencies, so that even less than a single scattering event can occur. In such a regime, Eq. 5 is not accurate [40]. Hence, we limit our analysis to frequency domains (inset in Fig. 2(c)) for which $L/\ell_t \geq 6$, so that the accuracy of diffusion theory is still reasonably good. While the trends of the decay constant as a function of frequency
and degree of correlation are in good agreement, only a fair agreement is found quantitatively. The deviation for $p = 70\%$ in particular is marked (more than one order of magnitude). This discrepancy can be explained by considering that (i) the expression used to evaluate the transport mean free path (Eq. 5) neglects completely recurrent scattering and near-field phenomena which are likely to occur in such dense systems (here, the filling fraction is $f = 20\%$), and (ii), more importantly for the strongly correlated system, the diffusion approximation disregards completely light localization phenomena, due to interference between multiply-scattered waves, which are expected to yield a reduction of the diffusion constant.

To better appreciate the occurrence of light localization in these systems, we retrieve the average lifetime $\gamma^{-1}$ from the numerical data at frequency $a/\lambda = 0.21$ for different $p$, as shown in Fig. 3(d). The clear, weakly fluctuating trend with varying $p$ indicates that 6 disorder realizations already provide reasonably converged results. The red line represents the prediction of diffusion theory according to Eqs. 3 and 6. A clear deviation between numerical calculations and diffusion theory occurs as $p$ increases. In particular, a dramatic increase of $\gamma^{-1}$ over an order of magnitude is observed for $p = 70\%$. Such a pronounced effect cannot be attributed solely to a reduction of $\ell_f$, since it would be captured by Eq. 6, but rather to Anderson localization of light. Localized modes are, on average, characterized by an exponentially decaying intensity distribution. This implies, due to weak coupling between these modes and the environment, lifetimes on average that are much longer than those of quasi-extended modes. Here, the reduction of $\ell_f$ with increasing $p$ yields a considerable variation of the localization length $\xi$, which eventually becomes shorter than the sample size $L$. This is, indeed, supported by the intensity maps shown in the insets of Fig. 3(d), calculated for a single steady-state source placed in the center of the system. Our interpretation is also corroborated by the observation of large variations of lifetimes for different realization of disorder, as shown by the pronounced errorbars in Fig. 3(d) for high $p$, which is expected in the localized regime. A transition from a quasi-extended regime ($\xi > L$) to a localized regime ($\xi \leq L$) has therefore been achieved by merely adding short-range correlations in the disordered system, keeping $f$ and $\phi$ unchanged. From these considerations, we can estimate that $\xi \leq L$, which is 1 order of magnitude smaller with respect to the prediction of Eq. 5.

To conclude, we have investigated how short-range correlations lead to considerable modifications of light transport and localization phenomena in 2D disordered photonic structures. Using a semi-analytical approach for the structure factor of the correlated systems (due to Baus and Colot), and a modified independent scattering approximation, we have investigated how key transport quantities are affected by short-range structural correlations. We have found in particular that short-range correlations make it possible to increase and/or decrease the localization length by several orders of magnitude. This shows that it is possible to design structures that are very weakly scattering and strongly localizing at nearby frequencies. Two-dimensional disordered systems in which the light transport and localization is finely-controlled may find interest in the fundamental study of localization phenomena, the conception of planar random lasers, thin-film photovoltaic and lighting technologies, or even help to reach the strong coupling...
regime with quantum dots or molecules [43, 44].

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