

Stirring of the propagation and the absorption of light in complex nanophotonic systems



Oluwafemi Stephen Ojambati

Stirring of the propagation and the
absorption of light in complex
nanophotonic media

Het roeren van de propagatie en absorptie
van licht in complexe nanofotonische
materialen

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Cover: Foam at the sea shore. The ocean is a complex photonic medium with both scattering and absorption. Light can be stirred in such a medium to increase absorption. In the sea foam, scattering dominates.

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STIRRING OF THE PROPAGATION AND THE ABSORPTION OF LIGHT IN COMPLEX NANOPHOTONIC MEDIA

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To my parents

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CHAPTER 1

Introduction

1.1 Complex nanophotonic media

Since the earliest times, we human beings have been curious about nature and strived to understand it. Due to our curiosity, we want to collect as much information as we can about our surroundings. One of the major tools to achieve this aim is through observations with our eyes. With our eyes, we obtain an enormous amount of information such as the admirable beauty of nature, a lion approaching us, the shapes and colors of objects, when to safely cross the road, and even reading the letters in this book, amongst a host of other things. The eyes collect this vast information by detecting light that has interacted and scattered from the objects. Therefore to have a deeper understanding of nature, we must understand how light interacts with the materials around us.

Already the Platonic philosophers were aware of the rectilinear propagation of light, meaning that light travels in a straight line in a homogeneous medium, which has a constant refractive index such as air or a slab of glass [1, 2]. Light inside such a homogeneous medium can be described using the principles of geometrical optics [3]. Light, however, behaves differently inside materials that have rapid changes of the refractive index, for example, an ensemble of microscopic pieces of broken glass. Such composite dielectric materials, which have spatially varying refractive indices on length scales comparable to or even smaller than the wavelength of light, are referred to as complex nanophotonic media. The complexity means that light travels in a much different way from the rectilinear propagation, and that the optical properties of the composite material as a whole deviate strongly from each constituent material. Therefore several formidable challenges arise that researchers in this field must surmount to describe, understand, and ultimately predict light propagation. The research is within the realm of the young and blossoming field called nanophotonics, which takes a root from the physics of electrons and spins in condensed matter physics [4–6].

Complex nanophotonic media can be generally classified based on the arrangement of the constituents: disordered, *i.e.*, a random arrangement of the constituents materials, or ordered, *i.e.*, a periodic arrangement of the constituent materials. In addition, there are also the exquisite quasicrystals and aperiodic structures [7, 8]. Since the 1980s, the study of light propagation in complex nanophotonic materials has attracted a great attention because these materials are a suitable platform to study exciting fundamental phenomena in physics and

have sparked numerous applications. Disordered materials are suitable media to study Anderson localization [9], coherent backscattering [10, 11], random lasing [12], short-, long-, and even infinite-range correlations [13–20]. Disordered materials have also made it to many home appliances such as the rough texture on a solar cell, phosphor plates in a white LED [21], and the diffuser glass on a bathroom window. Ordered media such as photonic crystals have been widely studied for spontaneous emission control [22], photonic band gaps [23, 24], thresholdless laser action [25], and Anderson localization [26]. Photonic crystals are widely used in applications such as high-efficiency LEDs [27], light trapping in solar cells [28, 29], low threshold lasers [25], ultralow-loss waveguides [30, 31], and photonic integrated circuit [24].

This thesis focuses on the study and control of the propagation and absorption of light inside complex nanophotonic systems. In this chapter, we present the necessary background for the thesis. In Sections 1.2 and 1.4, we describe the physics of light propagation inside a disordered and ordered medium respectively. We introduce the concept of wavefront shaping and “stirring of light” in Sections 1.3 and 1.5 respectively and the outline of the thesis is given in Section 1.6.

1.2 Multiple light scattering in disordered media

1.2.1 Relevant length scales

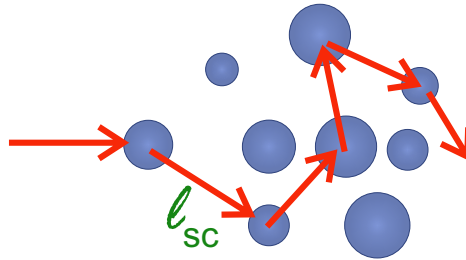


Figure 1.1: A cartoon illustrating the scattering mean free path ℓ_{sc} and the transport mean free path ℓ of scattered light random walking through an ensemble of disordered particles (blue circles). The incident light (red arrows) is scattered from particles to particle and the average step size of the scattering event is ℓ_{sc} . Light loses its sense of initial direction after traversing an averaged length of ℓ .

There are numerous scattering media¹ that are either of natural origin - such as biological tissue, fog, atmospheric and interstellar clouds, milk, and teeth - or man-made - such as paint, diffuser glass, and phosphor plates in white LEDs. When light is incident on a scattering medium, light is elastically scattered by the

¹We henceforth refer to a disordered complex nanophotonic system as a scattering medium. Other nomenclatures in literature include turbid, random, and opaque medium.



Figure 1.2: Light scattering in a suspension of milk with different concentrations inside four cuvettes. From left to right, the content of the cuvettes are water, milk suspension diluted 500 times, milk suspension diluted 50 times, and undiluted milk suspension. Behind each cuvette is the diffusion equation printed on a white paper.

randomly-distributed inhomogeneities (see Fig. 1.1). Multiple scattering occurs when light is scattered more than once, and if the wave nature of light is ignored, light performs a random walk through the medium. The mean distance between the scattering events is the scattering mean free path ℓ_{sc} [6, 32], see Fig. 1.1. Therefore, the condition for multiple scattering to occur is that the thickness of the sample L is larger than the scattering mean free path ℓ_{sc} , *i.e.*, $L > \ell_{sc}$. After several scattering events, the scattered light has lost the memory of its initial direction and has travelled through an average distance called the transport mean free path ℓ [6, 32]. A scattering medium with $L > \ell$ is therefore opaque since information in the incident light has been completely scrambled due to multiple scattering. In the presence of absorption, a relevant length scale is the absorption length ℓ_{abs} , which is the length after which light is absorbed to a fraction $1/e$. Interference effects (discussed in Subsection 1.2.2) are noticeable when the transport mean free path become comparable to the wavelength of light λ : $\ell \leq \lambda$.

The different regimes of multiple scattering are illustrated in Fig. 1.2. The diffusion equation is viewed through four cuvettes (length $L = 10 \text{ mm} \pm 1$) that are filled with water and different concentrations of milk (Albert Heijn Volle Melk). Undiluted milk has a transport mean free path $\ell \simeq 100 \mu\text{m}$ [33], $L/\ell \simeq 100$, and therefore the suspension is opaque. Taking a linear extrapolation of ℓ with respect to the dilution rate [33, 34], the second and third cuvettes with 500 and 50 times dilution have an estimated $\ell \simeq 50 \text{ mm}$ ($L/\ell \simeq 0.5$) and $\ell \simeq 5 \text{ mm}$ ($L/\ell \simeq 2$) respectively. The second cuvette is transparent because $L/\ell < 1$, nevertheless the few scattering events already result in a milky appearance, which is apparent in comparison to the first cuvette. In the third cuvette, the letters are already invisible due to multiple scattering, since $L > \ell$.

1.2.2 Interference of multiply scattered waves

One of the results of multiple scattering is speckle. Speckles are formed through the interference of multiple coherent waves, which are independent and have random amplitudes and phases [35]. In the early 1960s after the invention of the laser, speckles were first observed when light was reflected from a rough surface [36]. A ‘strange’ high-contrast, fine-scale granular pattern was observed and was later understood to be the interference of the scattered waves [37, 38]. Besides light, speckles occur in several other types of waves such as acoustic waves, microwaves, elastic waves, seismic waves and quantum waves [36].

Apart from speckle, interference of multiply scattered waves results in coherent backscattering, which was first observed for light in 1985 [10, 11]. Coherent backscattering means that even after multiple scattering the phase coherence of waves is preserved in the direction opposite to its incident direction, but less in other directions [4]. The experimental evidence of coherent backscattering is a bright intensity peak that is theoretically two times higher than the ensemble averaged diffuse background. Another effect of interference is Anderson localization and it was first proposed for inhibition of electron propagation in random lattices [9]. Due to strong multiple scattering, on average, waves are trapped inside the medium and the propagation is halted. One of the main foci of this thesis is about wavefront shaping, which is an interference effect as well and will be discussed in Section 1.3.

1.2.3 Diffusion of waves

Upon averaging over realizations of scatterers, speckles average out and the diffusion equation describes the resulting average energy density, which is the amount of energy in a wave field that is stored per unit volume [32, 39]. In the diffusion of waves, the phase information of the waves is neglected. The diffusion equation is a versatile tool to describe numerous complex problems in physics and several other fields of study. In 1905, Einstein first used the diffusion equation to describe the Brownian motion of particles undergoing a random walk in a liquid suspension [40]. It is now known that the diffusion equation also describes heat flow [41, 42], the propagation of electrons and neutrons in matter [43–45], the motion of solutes in a solvent [46], and the theory of stock exchanges [47]. The diffusion equation reads

$$\frac{\partial W(\mathbf{r}, t)}{\partial t} = D \nabla^2 W(\mathbf{r}, t), \quad (1.1)$$

where $W(\mathbf{r}, t)$ is the ensemble averaged energy density, $\mathbf{r}$ is the spatial coordinate, t is time, D is the diffusion constant. In this thesis, we find remarkably that the solutions to the diffusion equation describe the ensemble average energy densities of wavefront-shaped light and transmission channels, which are due to interference effects. An explanation for this remarkable finding is possibly that interference effects are still present even after ensemble averaging as is also the case in coherent backscattering and Anderson localization.

1.3 Control of light propagation through and inside a scattering medium

When light is incident on a scattering medium, it is decomposed in a set of N orthogonal modes, which have an amplitude A_n and phase ϕ_n , where n is the index of the input modes. The transmitted field E_m is related to the incident field E_n by a transmission matrix t_{mn}

$$E_m = \sum_n t_{mn} E_n = \sum_n A_n t_{mn} e^{i\phi_n} = \sum_n A_n |t_{mn}| e^{i(\phi_{mn} - \phi_n)}, \quad (1.2)$$

where m is the index of the output modes, the summation is over all input modes, and $|t_{mn}|$ and ϕ_{mn} are the amplitude and phase of the eigenvectors of t_{mn} . The transmission matrix t_{mn} contains the necessary information to describe uniquely the scattering events in a medium. In 1990, Freund proposed the possibility of using a scattering medium as a lens and other optical devices by controlling the incident field, since modifying the input field modifies also the transmitted field [48].

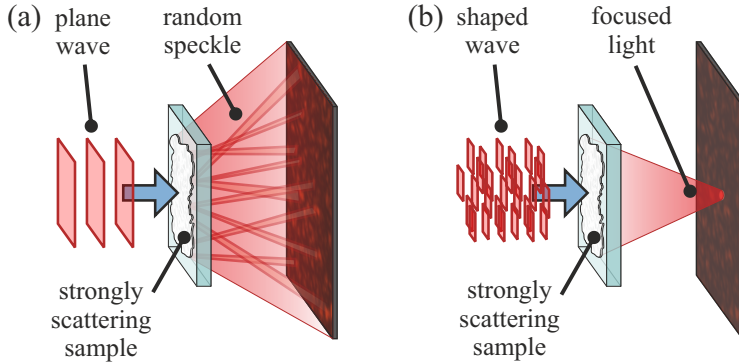


Figure 1.3: Cartoon illustrating light transport for plane incident waves and wavefront-shaped light. (a) A plane incident wave couples to all channels n that interfere to form a speckle pattern (b) The wavefront of light is shaped to have all transmitted channels interfere in a bright speckle spot. (Image from Ref. [49].)

Only in 2007, this prediction was confirmed in the pioneering work on wavefront shaping by Vellekoop and Mosk [49]. A scheme of a wavefront shaping experiment is shown in Fig. 1.3. For a plane incident wave, the transmitted field is a speckle pattern. In wavefront shaping, the input field is spatially shaped to obtain a bright transmitted spot, which is due to interference of all channels n . The amplitude of the field E_m is maximized if ϕ_n is exactly the opposite of ϕ_{mn} , *i.e.*, $\phi_n = -\phi_{mn}$. A spatial light modulator (SLM) such as a pixelated liquid crystal display controls the spatial phase of the incident light by applying a certain voltage to the terminals of each pixels of the SLM. The applied voltage

rotates the liquid crystal molecules of the SLM to give light a certain phase retardation thereby shaping the phase wavefront. An optimization algorithm uses the transmitted intensity in a speckle spot as a feedback signal and scans the phase of each SLM pixel to find the optimum wavefront that maximally couples light to the target [49, 50]. As shown in Ref. [49], the transmitted light can be enhanced up to 1000 times more than an ensemble average of the unoptimized light. With wavefront shaping, a scattering medium is therefore turned into a lens. Other methods have since been developed that also find $\phi_n = -\phi_{mn}$. These new methods are time reversal [51–53], phase conjugation [54–56], and transmission matrix-based control [57–59]. These wave shaping methods have opened up new optical applications such as non-invasive biomedical imaging [60–62], advanced optics [63–68], cryptography, and secure communication [69, 70].

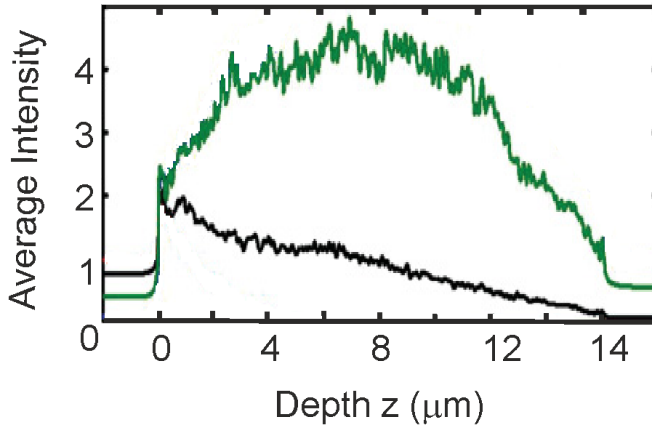


Figure 1.4: Average intensity versus depth obtained from numerical simulation of light inside a two-dimensional waveguide. The green and black curves are for wavefront-shaped and planar wavefront light [71].

Despite the uses of wavefront shaping, there is still an unresolved important question: how does light propagate *inside* the scattering medium when the incident wavefront is shaped? It is a challenge to “look” inside a scattering medium that is opaque. A first insight is available from numerical simulations that calculated the average intensity inside a two dimensional scattering medium [71]. The authors in Ref. [71] performed numerically a procedure similar to the optimization algorithm typically used in experiments. Fig. 1.4 shows the result of the numerical simulation for optimized light and a plane incident wave. The average intensity of wavefront-shaped light increases from the front side of the sample and peaks remarkably at a position around the center and then decreases towards the back side of the sample. The average intensity of the plane incident wave shows a linear decrease from the front side to the back side of the sample and this resembles the steady-state solution of the diffusion equation Eq. 1.1 (see Chapter 2). Fig. 1.4 shows that the total intensity is enhanced for optimized light compared to a plane incident wave. In this thesis, we investigate

experimentally and theoretically the energy density inside scattering media.

1.4 Light propagation in ordered media

Photonic crystals have attracted a lot of attention as highlighted in Section 1.1. Photonic crystals are periodic arrangements of dielectric materials with a period a in the order of the wavelength λ of light [24]. Similar to electrons in a semiconductor, a photonic crystal has a certain range of frequencies for which light is prohibited to propagate in the crystal and this range is called a stop gap. A stop gap originates from the interference of waves that are diffracted from periodic layers, which give rise to Bragg diffraction. The Bragg diffracted waves are associated with wavelength windows for which waves are forbidden to propagate: thus the stop band. In the case of visible light, the stop band gives a photonic crystal a shiny and colourful appearance, for instance, in the naturally occurring opals and in the wings of a butterfly [72–74].

Light with a frequency and wavevector outside the stop gap propagates through the photonic crystal as a Bloch wave, similar to electrons in atomic crystals [75]. Bloch waves are plane waves modulated by a spatially periodic function and they are the only allowed waves inside an infinite and perfect photonic crystal. However, the form of the waves in a real and finite photonic crystal differs from a Bloch wave as a result of the influence of the unavoidable disorder and finiteness of the crystal [76, 77]. We investigate this interplay between order and disorder in the Chapter 6.

An important parameter that defines how strongly light interacts with a complex photonic system is the photonic strength. The photonic strength S is defined as the dimensionless ratio of the polarizability² α of an average scatterer in a complex medium to the average volume per scatterer V [78].

$$S \equiv \frac{4\pi\alpha}{V}. \quad (1.3)$$

The photonic strength takes into account the refractive index contrast, in other words, it is the ratio of the “optical volume” that light experiences to the physical volume of each scatterer. For a photonic crystal, S can be determined experimentally from the relative width of the *first order* stop band [79, 80]. The width $\Delta\omega$ of the stop band is determined by the Bragg length L_B , which is the exponential decay length of the incident light at the center wavelength of the stop band. The two quantities S and L_B are related by

$$L_B = \frac{2d}{\pi S}, \quad (1.4)$$

where d is the spacing between the crystal planes. For a disordered medium, S

²Polarizability of a medium α is defined as the ratio of the induced dipole moment $\mathbf{p}$ of an atom to an external electric field $\mathbf{E}$ [39].

is defined in terms of ℓ and the wavevector k as

$$S = \frac{1}{k\ell}. \quad (1.5)$$

The wonderful feature of the photonic strength is that it gauges any nanophotonic medium, regardless of the structure of the building block [78].

1.5 “Stirring of light”

So far, we have described the principles of propagation and control of light through complex photonic systems. We have neglected the presence and the influence of absorption in these systems. As described by the Kramers-Kronig relations, absorption is inevitable in dielectric materials [3]. In optics, absorption is traditionally considered a nuisance since it reduces the optical signal. Absorption is however significant and is intertwined with scattering in several complex nanophotonic media. The ingenious idea of “Stirring of light” is to use absorption that is inherent in complex nanophotonic systems to our advantage and with a desired control³. This revolutionary idea is very relevant today in the enormous applications of nanophotonics, especially in those that uses absorption to harness optical energy. Examples of such applications are white LEDs, solar cells, and random lasers.

Our main tools to “stir” light are wavefront shaping and nanostructures. These tools “stir” light by optimally matching the optical impedance⁴ of the input light to modes of the system that are optimally absorbed. As described in Section 1.3, we want to explore wavefront shaping to enhance the total stored energy. Thereby, the total absorbed optical energy is enhanced, if absorption is present. How useful is this? An enhancement of the energy density inside a scattering medium is useful for systems that have absorption and scattering intertwined. First, for a white LED, which have a blue LED that illuminates phosphor particles, an enhanced energy density increases the output of the LED and therefore less amount of expensive phosphor can be utilized. Second, a solar cell has a scattering medium, (*e.g.*, etched titanium dioxide [82]) on top of the absorbing silicon layer scatters the incident light at large oblique angles to increase the optical path length inside the absorbing layer. An enhanced energy density can increase the output power of a solar cell, especially at the longer wavelengths where silicon has a lower absorption. Third, in a random laser that has an active scattering medium, an enhanced energy density serves to reduce the threshold level of the random laser [12].

The second tool to “stir” light is a complex nanostructure such as a photonic crystal, which has been used recently to enhance light trapping inside a solar cell in various architecture [28, 29]. An example is to utilize the photonic stop

³This work is part of a FOM program to that effect.

⁴Optical impedance Z_{opt} is an analogue of the impedance in an electronic circuit and is defined as $Z_{opt} = \sqrt{\mu/\epsilon}$, where μ and ϵ are the magnetic permeability and electric permittivity respectively [81].

gap of a photonic crystal in a back reflector of a solar cell to reflect the longer wavelengths. Another way to use a photonic crystal is to engineer a cavity inside a 3D photonic crystal and the cavity confines light to enhance the energy density. A solar cell placed inside the cavity experiences an enhanced absorption and hence the photocurrent increases.

1.6 Outline of this thesis

This thesis presents experimental studies on the fundamental concepts to “stir” light in complex nanophotonic materials. We interpret our experimental findings using theoretical and numerical models. This thesis is organized as follows:

In **Chapter 2**, we accurately determine the profile of the energy density inside a scattering medium. We compute numerically the energy density inside a quasi-1D waveguide with point scatterers. The computed energy density was reconstructed using diffusion eigenfunctions. We find that only a few diffusion eigenfunctions suffice to reconstruct the energy densities of shaped waves and transmission eigenchannels.

In **Chapter 3**, we present the theoretical background of probing the total stored energy and the depth-dependent energy density inside a 3D scattering medium using fluorescence microscopy.

In **Chapter 4**, we report the first ever observation to show that wavefront shaping couples light to the fundamental diffusion eigenfunction inside a 3D scattering medium. The total stored energy in a scattering medium is probed by exciting fluorescent spheres that are randomly positioned in the medium. We find that the concomitant fluorescent power is enhanced for an optimized wavefront compared to a non-optimized incident wavefront. The observed enhancement increases with sample thickness. Our model based on fundamental eigenfunction agrees remarkably well with our experiments, notably since the model has no freely adjustable parameters.

In **Chapter 5**, we present an experiment to measure the depth-dependent energy density inside a 3D scattering medium. We probe the energy density at a specific depth inside the sample by using a single fluorescent particle positioned at a particular depth. The depth of the particle is determined by comparing the size of the diffuse fluorescent intensity to the solution of the diffusion equation. We find that the energy density is depth-dependent and increases at depths close to the back surface of the sample.

In **Chapter 6**, we study the nature of waves in real 2D and 3D photonic crystals, which have inevitable fabrication-induced disorder. We employ intensity statistics and spatial correlations to deduce the nature of the waves. We find that the waves in an artificial opal comprise of both Bloch waves, which are expected for a perfect crystal, and spherical waves that are scattered from the sites of disorder.

In **Chapter 7**, we experimentally study how the intensity enhancement on both interfaces of a scattering medium depends on the optimization area on the transmission side. We observe that as the optimization radius increases, the

enhancement of the total transmitted intensity increases. There is a concomitant decrease of the total reflected intensity, which implies an energy redistribution between transmission and reflection channels. In addition, we find a qualitative evidence of a long range reflection-transmission correlation.

In **Chapter 8**, we investigate the effect of spatial intensity fluctuation inside a laser beam on wavefront shaping. The spatial intensity fluctuations were suppressed using mode cleaning. We found that the fidelity of the optimization in wavefront shaping increases after mode cleaning.

In **Chapter 9**, we summarize the results in this thesis and present an outlook to possible future developments and applications.

CHAPTER 2

Mapping the energy density of shaped waves in scattering media onto a complete set of diffusion modes

2.1 Introduction

Recently, novel wave-shaping methods such as feedback-based wavefront shaping [48, 49, 83–87], time reversal [51–53], phase conjugation [54–56], and transmission matrix-based control [57–59] have demonstrated the control of transmission and reflection through scattering media. These wave-shaping methods remarkably enhance the intensity in a single speckle as well as the total reflected and transmitted intensity. The wave-shaping methods have led the way for exciting applications such as non-invasive biomedical imaging [60–62], advanced optics [63–68], and cryptography and secure communication [69, 70].

Although these methods are useful tools in controlling the intensity at the interfaces of a scattering medium, the energy density distribution of the shaped waves inside the scattering medium is still unknown. Here, we refer to shaped waves to mean perfect phase conjugation of the transmitted waves, which is equivalent to shaping to optimally focus light to a single speckle spot [83, 87]. In the case of light, the knowledge of the energy density distribution is important for applications such as enhanced energy conversion in white LEDs [88–92], efficient light harvesting in solar cells [82, 93, 94], high yield random laser [12, 95], and controlled illumination in biomedical imaging [96]. To date, only numerical calculations of scalar waves [71, 97, 98] and a single-realization elastic wave experiment [99] have addressed the energy density distribution of shaped waves inside two-dimensional (2D) scattering media. However, none of these studies provide an analytical model for the energy density distribution. For high-transmission channels, Davy *et al* [97] described the energy density using a parabolic function. In Ref. [100] and Chapter 4, our team reported the first measurement of the total energy density of light inside a three-dimensional (3D) scattering medium and reported an enhanced total energy density for shaped waves. In that paper, the experimental result was interpreted with a model that describes the total energy density of shaped waves using only the fundamental eigenfunction of the diffusion equation. The model agreed well with experimental

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observation. However, the much desired full description of the spatial energy density for shaped waves needs a more fundamental study.

In this chapter, we examine the spatial energy density of shaped waves inside a scattering medium. Our starting point is the mathematical theorem that the eigenfunctions of the diffusion equation form a complete set [101–103]. Therefore, this set can describe any function defined on the sample support, which is specified by the two boundaries. As an example, the complete set of diffusion eigenfunctions can accurately describe the spatial energy density as well as any speckle inside the sample. The fact that the diffusion eigenfunctions can describe any function inside the medium implies in no way a connection to the diffusion approximation. However, such a connection can be inferred if it turns out that the required number of eigenfunctions is small. In this chapter, we will demonstrate that expanding the spatial energy density using the complete set of diffusion eigenfunctions is enlightening because only a few diffusion eigenfunctions suffice to reconstruct the energy density.

We obtained the spatial energy density inside a quasi-1D disordered waveguide by numerical calculation of concatenated scattering matrices [104–106]. A quasi-1D disordered waveguide provides a tractable platform to study the essential physics. We calculated the spatial energy density $W(z)$ of all transmission channels. By calculating the transmission-averaged energy density for all transmission channels, we retrieved the energy density of shaped waves [83, 87]. We obtained the intensity transmission coefficients of all channels, and we henceforth refer to τ as the transmission of a specific channel.

We observe that only the first seven diffusion eigenfunctions suffice to reconstruct the spatial energy density distribution of the shaped waves. Taking into account only the fundamental eigenfunction of the diffusion equation, the total internal energy inside the sample is underestimated by only 2%. The spatial energy density is very similar to the fundamental eigenfunction, within a cosine distance of 0.01. Furthermore, we are able to reconstruct the energy density distribution of both high- and low- transmission channels, using a few M eigenfunctions of the diffusion equation, *e.g.*, $M = 16$ for channels with transmission $\tau = 0.1$.

It is fascinating to see that the energy densities of shaped waves and transmission channels, which are due to interference effects, can be truly described using a few diffusion eigensolutions. The reconstruction of the distribution of the energy densities that we report here can be extended to three dimensional (3D) scattering samples. The reconstruction also applies to all forms of waves such as light, sound, elastic waves and quantum waves.

2.2 Theory

2.2.1 Eigenfunctions of the diffusion equation

The diffusion equation describes the ensemble averaged energy density $W(\mathbf{r}, t)$ of multiply scattered waves as a function of position $\mathbf{r}$ and time t inside a scattering

medium [18, 32],

$$\frac{\partial W(\mathbf{r}, t)}{\partial t} = D \nabla^2 W(\mathbf{r}, t), \quad (2.1)$$

where D is the diffusion constant. Anticipating the fact that the directions (x, y) are decoupled from the z -direction we decompose the energy density as

$$W(\mathbf{r}, t) = W_{\perp}(x, y, t) W(z, t). \quad (2.2)$$

Since we are considering a slab that is a quasi-1D scattering medium, we assume translational invariance in the perpendicular directions (x, y) . The sample boundaries are at $z = 0$ and $z = L$, where L is the sample thickness. This symmetry allows us to solve $W_{\perp}(x, y, t)$ by using its two-dimensional spatial Fourier transform $W_{\perp}(\mathbf{q}_{\perp}, t)$. Solving the (x, y) -part of Eq. (2.1) and using the initial condition for $t = 0$ gives

$$W_{\perp}(\mathbf{q}_{\perp}, t) = W(\mathbf{q}_{\perp}, t = 0) e^{-Dq_{\perp}^2 t}. \quad (2.3)$$

Next we solve the remaining 1D equation

$$\frac{\partial W(z, t)}{\partial t} = D \frac{\partial^2 W(z, t)}{\partial z^2} \quad (2.4)$$

We will turn Eq. 2.4 into a Sturm-Liouville eigenvalue problem by introducing the Ansatz $W(z, t) = e^{-\gamma t} W(z)$

$$\frac{\partial^2 W(z)}{\partial z^2} + \Gamma W(z) = 0, \quad (2.5)$$

$$\Gamma \equiv \frac{\gamma}{D}. \quad (2.6)$$

As is well-known from linear algebra [101] solutions of Sturm-Liouville equations are orthogonal and form a complete set. To solve Eq. 2.5, we apply the von Neumann boundary conditions, which are given as

$$W(z = 0) - z_{e1} \frac{\partial W(z)}{\partial z} \Big|_{z=0} = 0 \quad (2.7)$$

and

$$W(z = L) + z_{e2} \frac{\partial W(z)}{\partial z} \Big|_{z=L} = 0, \quad (2.8)$$

where z_{e1} and z_{e2} are the well-known extrapolation lengths at the front and back surfaces of the sample respectively [107]. Applying the boundary conditions, Eqs. 2.7 and 2.8 lead to a discrete set of eigenvalues Γ_m . The general eigenfunction for eigenvalue Γ_m is cast in the form

$$W_m(z) = A_m^{-1/2} \sin(\kappa_m z + \eta_m). \quad (2.9)$$

As outlined in detail in Ref. [107] the wavevector κ_m and phase η_m are determined by solving Eq. 2.5 with the boundary conditions. The parameter A_m takes care of the normalization of the eigenfunctions, such that

$$\int_0^L W_m W_n dz = \delta_{mn}, \quad (2.10)$$

where δ_{mn} is the Kronecker delta. The relation between the eigenvalue and wavevector is $\Gamma_m = D\kappa_m^2$. The allowed wave vectors κ_m are obtained by solving the implicit equation

$$\tan(\kappa_m L) = \frac{(z_{e1} + z_{e2})\kappa_m}{z_{e1}z_{e2}\kappa_m^2 - 1}. \quad (2.11)$$

Following Ref. [107]*, the phase factor η_m fulfills the following equation

$$\tan \eta_m = z_{e1}\kappa_m, \quad (2.12)$$

and the normalization factor A_m is given by

$$A_m = \frac{1}{2}L - \frac{1}{2\kappa_m} \cos(\kappa_m L + 2\eta_m) \sin(\kappa_m L). \quad (2.13)$$

For the exact solution of Eq. 2.11, see Appendix 2.A. The first ten wavevectors are listed in Table 2.1 for a particular set of boundary conditions. In Fig. 2.1, we plot the first three eigenfunctions using Eq. 2.9 with the same boundary conditions. The fundamental diffusion eigenfunction $m = 1$ is a half-period sine function, which is positive value throughout the sample. Therefore, the $m = 1$ eigenfunction can describe a specific energy density inside a scattering medium, since the energy density is positive. On the other hand, the $m = 2, 3$ diffusion eigenfunctions (and other higher eigenfunctions not shown) are both positive and negative within the sample boundary. Since a negative energy density is non-physical, diffusion eigenfunctions $m = 2, 3$ cannot *individually* describe energy density in a scattering medium. However, a sum of these diffusion eigensolutions that is positive throughout the sample can describe a physical energy density inside the medium. The weight of the fundamental eigenfunction takes care of the sum to be positive everywhere.

2.2.2 Reconstructing the energy density with eigenfunctions of the diffusion equation

The diffusion equation is of the Sturm-Liouville type so its eigenfunctions form a complete and orthogonal set [101]. Therefore, any distribution of energy density within the domain of the sample boundaries can be decomposed into a sum over a finite number M of eigenfunctions for a specific set of coefficients. Such a

*It is noted that Eqs. 2.11 and 2.12 differs from the ones obtained in Ref. [107] because we consider here different extrapolation lengths for both surfaces, while in Ref. [107] they were taken to be the same.

Eigenfunction index index m	Wavevector κ_m
1	0.229566
2	0.460711
3	0.694591
4	0.931747
5	1.172199
6	1.415643
7	1.661649
8	1.909776
9	2.159633
10	2.410889

Table 2.1: Values of the wavevector κ_m for the first 10 eigenfunctions of the diffusion equation. The values were obtained by solving Eq. 2.11. The extrapolation lengths are $z_{e1} = z_{e2} = z_e \ell = (\pi/4)\ell$ (for a 2D sample [6]) and $L = 12.1\ell$.

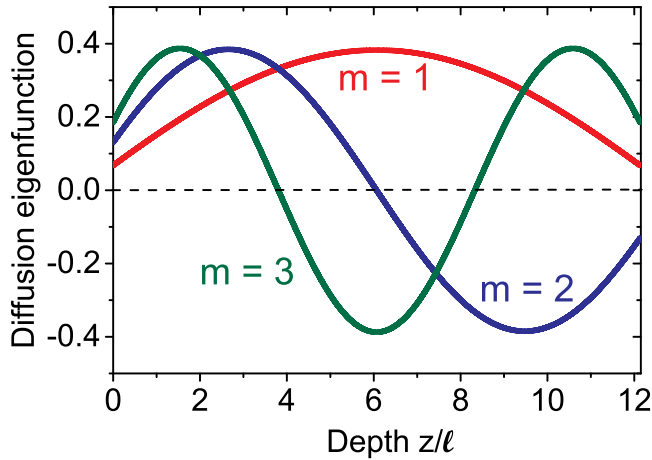


Figure 2.1: The first three eigenfunctions of the diffusion equation plotted using equation Eq. 2.9. The wave vectors κ_m are presented in Table 2.1 and the used parameters can be found in the caption of that table.

decomposition is only useful if the number of needed modes is small. We express a certain ensemble averaged energy density $W_s(z)$ inside the scattering medium in terms of the normalized eigenfunctions $W_m(z)$

$$W_s(z) = \sum_{m=1}^M d_m W_m(z), \quad (2.14)$$

where d_m are coefficients. We calculate the overlap integral I_p of the product of $W_s(z)$ and $W_p(z)$

$$I_p \equiv \int_{z=0}^{z=L} dz W_s(z) W_p(z), \quad (2.15)$$

and substitute Eq. 2.14 into Eq. 2.15 to obtain

$$I_p = \int_{z=0}^{z=L} dz \sum_{m=1}^M d_m W_m(z) W_p(z). \quad (2.16)$$

We use the orthonormality of functions $W_p(z)$ and $W_m(z)$, which is expressed as

$$\int_{z=0}^{z=L} dz W_p(z) W_m(z) = \delta_{mp}, \quad (2.17)$$

Therefore, Eq. 2.16 becomes

$$I_p = \sum_{m=1}^M d_m \delta_{mp} = d_p. \quad (2.18)$$

We now can write the decomposition (2.14) as

$$W_s(z) = \sum_{m=1}^M I_m W_m(z), \quad (2.19)$$

In this chapter, the spatial energy density $W_s(z)$ that we reconstruct is that of shaped waves $W_{\text{sw}}(z)$. We also reconstruct the spatial energy density of transmission eigenchannels $W_n(z, \tau)$. The spatial energy densities are obtained from numerical simulation and the details of the simulation are described in Section 2.3. The simulation results are compared to the reconstructed energy density $W_{\text{re}}(z)$ in Section 2.4.

In order to quantify the overlap between the reconstructed function $W_{\text{re}}(z)$ and the numerical data $W_s(z)$, we use the cosine distance COSD [108], which is defined as

$$\text{COSD} \equiv 1 - \frac{\sum_{i=1}^{N_s} W_{\text{re}}(z_i) W_s(z_i)}{\left[\sum_{i=1}^{N_s} W_{\text{re}}^2(z_i) \right]^{\frac{1}{2}} \left[\sum_{i=1}^{N_s} W_s^2(z_i) \right]^{\frac{1}{2}}}. \quad (2.20)$$

N_s is the number of points in the simulation data. COSD varies between 1 and 0 and tends towards 0 as the reconstructed function fully describes the simulation data. As a figure-of-merit of a good reconstruction, we choose the eigenfunction that has $\text{COSD} \leq 10^{-4}$. At this COSD, the reconstructed function deviates by only 2% from the actual function. This level of accuracy is within what is achievable in a measurement with a realistic level of experimental noise.

2.3 Numerical samples and setup

2.3.1 Numerical setup

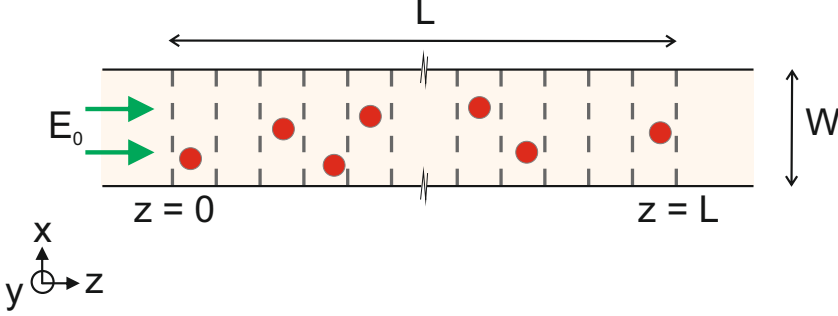


Figure 2.2: Schematic of the numerical sample: a quasi-1D waveguide. In the region $z = 0$ to $z = L$, scatterers (red circles) are randomly distributed in the waveguide. The width W and length L of the scattering part of the waveguide are such that $W \ll L$. The waveguide is illuminated with an incident field E_0 that is scattered in the waveguide. The waveguide is divided into slices (vertical dashed line) and in each slice there is at most one scattering particle. The scattering matrix of each of the slices is computed and concatenated to give the scattering matrix of the whole sample.

We perform simulations of transport of monochromatic scalar waves through a waveguide containing scatterers. The waveguide is modeled as a quasi-one-dimensional (quasi-1D) system possessing $N_m = 100$ transversal modes, with elastic scattering on bulk impurities. This is implemented as a 2D waveguide with 100 transversal modes, containing scatterers at random positions with a density of 0.2 scatterer per square wavelength. A schematic of the numerical sample is shown in Fig. 2.2. In order to calculate the energy density inside the waveguide we employ a recursive S -matrix formalism, where the waveguide is first divided into sub-wavelength slices, each containing a maximum of one scatterer. For each slice we define the S -matrix in a mode representation as follows [106],

$$S \equiv \begin{pmatrix} R_L & T^T \\ T & R_R \end{pmatrix}, \quad (2.21)$$

where R_L and R_R are the left and right reflection matrix respectively of dimension $N_m \times N_m$, T is the left-to-right transmission matrix. The right-to-left transmission matrix in this reciprocal system is the transpose of T . The S -matrix of each slice, of dimensions $2N_m \times 2N_m$, is calculated in the approximation that no recurrent scattering takes place within the thin slice. Next, the S -matrices of the slices are joined, including contributions of recurrent scattering, using the composition rule for S -matrices [104]. This numerically stable procedure yields the S -matrix of the entire waveguide and of subsections of it. From the

transmission matrix we extract the singular values and vectors corresponding to all channels by singular value decomposition. The energy density inside the waveguide, for a given incident field, is calculated as follows: For a given axial coordinate z_c the S matrices S_1, S_2 of the waveguide sections $0 < z < z_c$ and $z_c < z < L$ respectively are calculated. The field in the $z = z_c$ plane is then found by calculating recurrent scattering diagrams:

$$E_c = T_1 E_{in} + R_{L2} T_1 E_{in} + R_{R1} R_{L2} T_1 E_{in} + R_{L2} R_{R1} R_{L2} T_1 E_{in} + \dots \quad (2.22)$$

$$= (1 + R_{L2})(1 - R_{R1} R_{L2})^{-1} T_1 E_{in}. \quad (2.23)$$

Here, R_{L2} is the left reflection submatrix of S_2 , R_{R1} is the right reflection submatrix of S_1 , and T_1 is the left to right transmission submatrix of S_1 . Subsequently for every z_s the time-averaged energy density $\varepsilon_0 |E_c|^2$ is integrated over the cross section to obtain the projected energy density $W(z)$. The calculation is repeated for 8000 independently generated random waveguides from which the transmission channels are extracted. To obtain plots of the average energy density as a function of transmission, we average the energy densities of channels in a narrow band of transmission values.

2.3.2 Sample characterization

In order to characterize the numerical samples, we plot in Fig. 2.3(a) the probability distribution $P(\tau)$ as a function of the transmission τ . The probability distribution $P(\tau)$ obtained from the simulation is bi-modal: there is a high probability for transmitting channels with transmission τ close to zero and close to one. In Fig. 2.3(a), we compare the Dorokhov-Mello-Pereyra-Kumar (DMPK) distribution [106] with our numerical result. The probability distribution of transmission for a scattering medium is expected to follow the DMPK distribution. Indeed, the DMPK distribution agrees well with our numerical result. Furthermore, in order to confirm that our numerical samples are in the diffusive regime, we plot in Fig. 2.3(b) the equally-weighted ensemble averaged energy density versus reduced sample depth. The ensemble averaged energy density shows a linear decrease from the front surface towards the end surface of the sample, in agreement with the prediction of diffusion theory for a diffusive sample, see Fig. 2.3(b).

2.4 Results and discussions

2.4.1 Energy density of shaped waves

We obtained the energy density of shaped waves W_{sw} as the transmission-weighted ensemble average of the energy density of transmission channels $W_n(z, \tau)$, as shown in Ref. [83, 87]:

$$W_{sw}(z) = \sum_n^N \tau_n W_n(z, \tau), \quad (2.24)$$

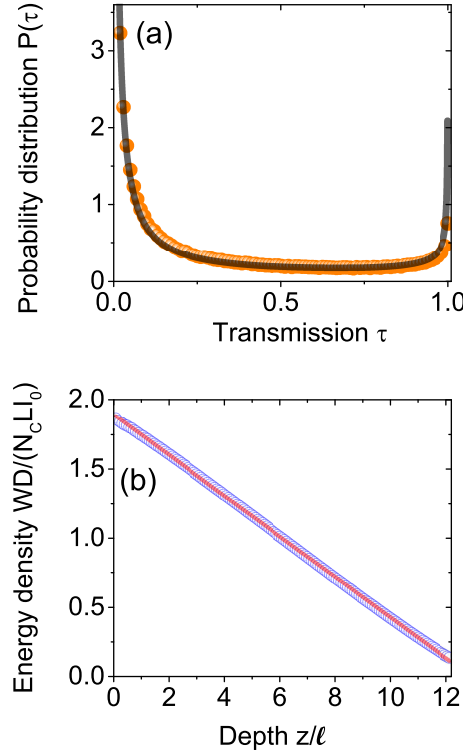


Figure 2.3: (a) Probability distribution of the transmission from the simulation (blue dots) and the expected Dorokhov-Mello-Pereyra-Kumar (DMPK) distribution (red curve). (b) An equally-weighted summation over the energy density of all transmission channels (blue dots) and the expectation from diffusion theory (red line) as a function of the normalized depth z/ℓ . In (a) and (b), we ensemble averaged over 8000 waveguides and the sample thickness is $L = 12.1\ell$.

where N is the number of transmission channels. Eq. 2.24 implies that wavefront shaping couples an energy fraction τ_n into a transmission eigenchannel n . Using Eq. 2.24, we obtain the energy density of the shaped waves from the transmission-weighted average of the energy density for all transmission channels. We show in Fig. 2.4(a) the ensemble averaged energy density distribution of shaped waves. The energy density distribution of the shaped waves increases from the interfaces of the sample and is maximal at $z = 4.9\ell$, about 20% off the center of the sample ($z = 6.1\ell$). The ensemble averaged energy density of wavefront-shaped light was first obtained in Ref. [71] by solving the Maxwell equation using the finite-difference time-domain (FDTD) method. Despite the different numerical calculation methods, our numerical result is closely similar to the one obtained by Choi *et al* [71]. The exact peak position of the energy density obtained in Ref. [71] is difficult to estimate due to the noise in the data, however, the peak

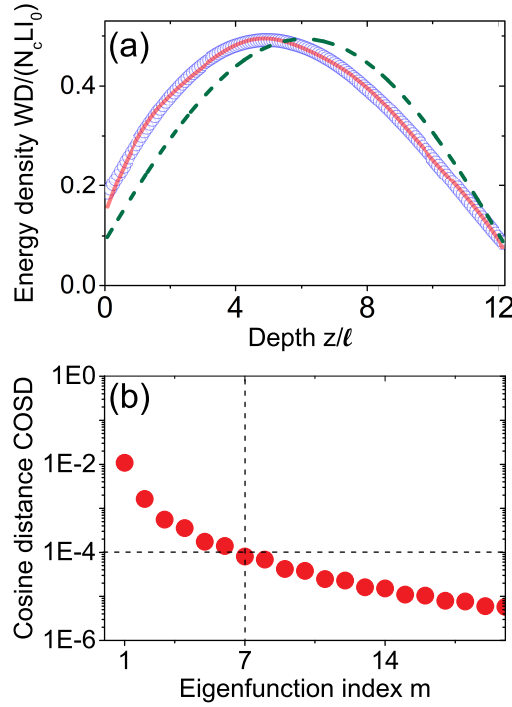


Figure 2.4: (a) Transmission-weighted averaged energy density versus normalized sample depth z/ℓ . The transmission-weighted averaged energy density is equivalent to wavefront-shaped or phase-conjugated light [83]. The blue circles are the energy density obtained from simulation, the green dashed curve is the diffusion $m = 1$ eigenfunction, and the red curve is the summation over the first seven eigenfunctions. (b) Cosine distance COSD versus eigenfunction index m . The vertical and horizontal dashed-lines show our figure-of-merit $COSD \approx 10^{-4}$ and the eigenfunctions that fulfill the criterion $m = 7$ respectively. In (a) and (b), the sample is the same as in Fig. 2.3.

position is close to the center of the sample.

We reconstructed the distribution of the energy density of shaped waves with eigenfunctions of the diffusion equation. From the reconstruction, we obtain the cosine distance COSD that we plot in Fig. 2.4. In case of shaped waves, a summation over the first $M = 7$ eigenfunctions performs remarkably well ($COSD \leq 10^{-4}$) to reconstruct the energy density, see Fig. 2.4(a). Only the fundamental diffusion eigenfunction $m = 1$ has $COSD \approx 10^{-2}$ and is not sufficient to describe the energy density profile. The fundamental eigensolution peaks at exactly the center of the sample, in contrast to the energy density of shaped waves. To put the number $M = 7$ in perspective, we performed the same reconstruction on the profile of 50 speckle spots within the same sample domain. We find that over 300 eigenfunctions are required to obtain a $COSD \leq 10^{-4}$,

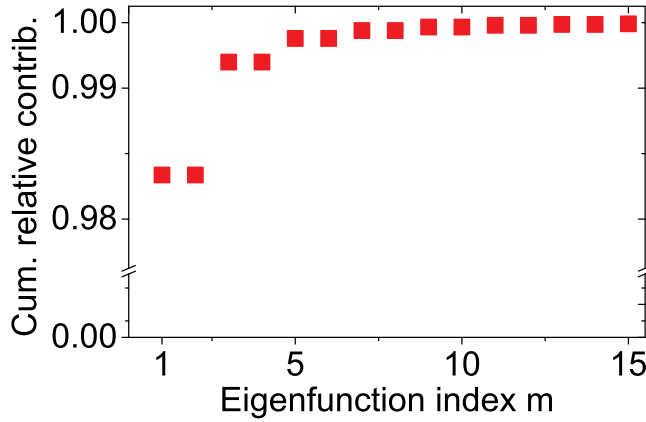


Figure 2.5: The cumulative contribution of the eigenfunctions of the diffusion equation to the total internal energy density of shaped waves relative to a sum over the first 100 eigenfunctions versus the eigenfunction index m .

indicating that the diffusion eigenfunctions are not a suitable basis for a random speckle. In contrast, the low number of eigenfunctions required to represent the shaped waves implies that the solution of the diffusion equation is close to the ensemble averaged energy density of shaped waves.

We further investigated the contribution of the eigenfunctions to the *total* energy density integrated along the depth of the sample relative to the contribution of the first 100 eigenfunctions. As a reference, we choose the first 100 eigenfunctions because the higher order eigenfunctions have a very negligible contribution. In Fig. 2.5, we plot the contributions. For shaped waves, the $m = 1$ contributes 98% of the *total* energy density inside the sample. Our finding about the strong contribution of $m = 1$ to the total internal energy density agrees remarkably well with the heuristic model in Ref. [100] that wavefront shaping *predominantly* couples light to the fundamental diffusion eigensolution. The contribution of $m = 2$ is negligible compared to $m = 1$ since $m = 2$ has negative energy density in almost half of the sample depth, which cancels the positive energy density (see Fig. 2.1). A similar effect also happens for all other even index eigenfunction.

2.4.2 Energy density of transmission channels

The ensemble averaged energy density of open transmitting eigenchannels with a transmission τ in the range $0.99 < \tau < 1$ is shown in Fig. 2.6(a). The energy density shows a symmetric profile, which peaks in the middle of the sample. Our numerical result is similar to the one obtained by Davy *et al* [97]. Interestingly, only the fundamental diffusion eigenfunction $m = 1$ is sufficient to reconstruct the energy density of the open channel, see Fig. 2.4(a). The fundamental diffusion eigenfunction $m = 1$ is expected to describe the open eigenchannels for the following reasons: Firstly, as the open eigenchannel has the highest individual

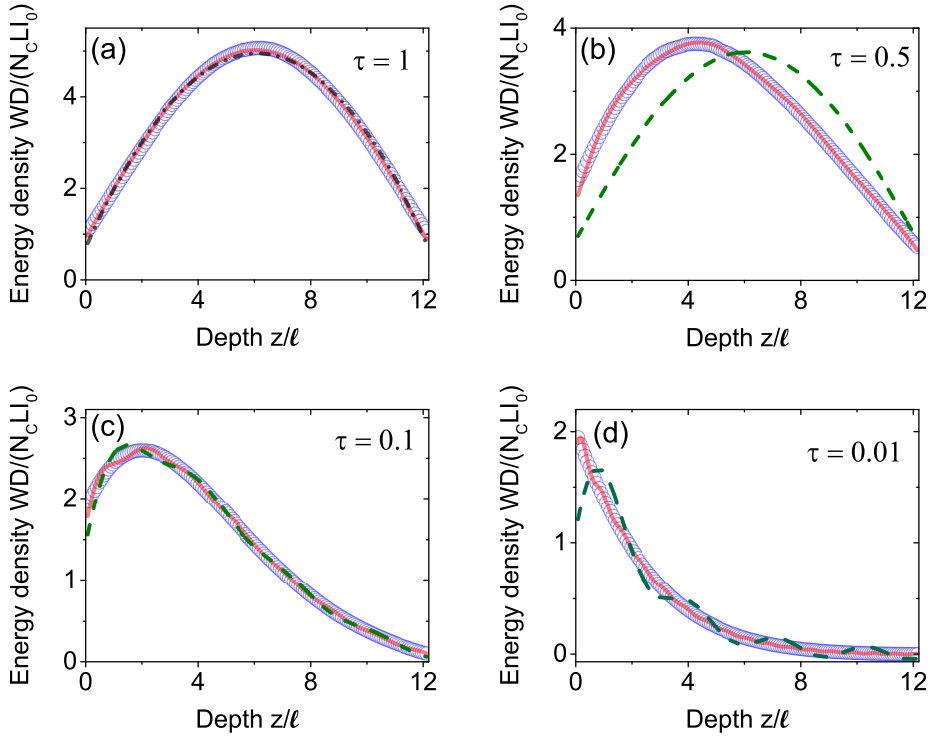


Figure 2.6: (a) Ensemble averaged energy density of an open channel in a scattering medium versus normalized depth z/l_{tr} with transmission τ in the range $0.99 < \tau < 1$. The blue circles are the energy density data obtained from simulation, and the red line is the fundamental diffusion eigenfunction $m = 1$. The black dashed-dotted curve is a parabolic fit. (b) Ensemble averaged energy density of transmission channels in a scattering medium versus the normalized depth z/l_{tr} for transmission τ in the range $0.49 < \tau < 0.51$. The blue circles are the energy density data obtained from simulation, the green line is the diffusion $m = 1$ eigenfunction, and the red line is a $M = 7$ summation of diffusion eigenfunctions. (c) The blue circles are obtained as in (a) and the transmission τ is in the range of $0.09 < \tau < 0.11$. The green and the red lines are summations over $M = 8$ and $M = 16$ diffusion eigenfunctions respectively. (d) The blue circles are obtained as in (a) and the transmission τ is in the range: $0 < \tau < 0.02$, which signifies closed channels. The green and the red lines are summations over $M = 8$ and $M = 33$ diffusion eigenfunction respectively.

transmission, the fundamental eigenfunction ($m = 1$) as well contributes most to the total transmission as shown in Ref. [100]. Secondly, the fundamental eigenfunction is the only physical solution with a positive energy density along the sample depth z , see Fig. 2.1 and therefore this eigenfunction must have the

largest weight in the expansion. Interestingly, a simplified mathematical model by Davy *et al* [97] described the energy density of open channels as a parabolic function. We note that the deviations from a parabolic shape are obvious only near the edges, and the shape of the energy density curve agrees better with the cosine shape of the fundamental eigenfunction, see Fig. 2.6(a). Indeed, the COSD for the parabola is 10^{-3} compared to 10^{-5} for the fundamental eigenfunction.

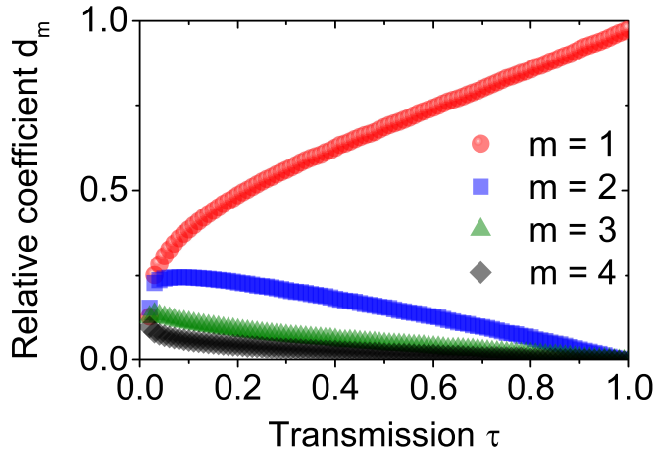


Figure 2.7: Contribution of eigenfunctions of the diffusion equation relative to a sum of the first 100 eigenfunctions versus transmission τ for eigenfunctions $m = 1, 2, 3$, and 4 plotted with red circles, blue squares, green triangles, and black diamonds respectively.

In order to describe the distribution of energy density of low-transmission channels, we also use the eigenfunctions of the diffusion equation to reconstruct the ensemble averaged energy densities of low-transmission channels obtained from simulation. In Figs. 2.6 (b - d), we show the ensemble averaged energy densities for transmission eigenchannels with transmissions $\tau = 0.5, 0.10$ and 0.01 . For the eigenchannel with transmission $\tau = 0.5$, only $M = 7$ eigenfunctions of the diffusion equation are sufficient to reconstruct its energy density, $M = 16$ eigenfunctions for $\tau = 0.1$, and $M = 33$ is required for a closed channel $\tau = 0.01$. As the transmission τ decreases, the number of eigenfunctions sufficient to reconstruct the energy density increases. The increase in the number M of sufficient eigenfunctions occurs since the asymmetry of the distribution of energy density increases as the transmission τ reduces, and likewise, the asymmetry of the eigenfunctions increases with the eigensolution index. Therefore, the higher index eigenfunctions contribute more to an asymmetric function. The contribution of the eigenfunctions with index $m = 1, 2, 3$ and 4 relative to a summation over the first 100 eigenfunctions is shown in Fig. 2.7. The relative contribution of the fundamental eigenfunction of the diffusion equation $m = 1$, which is a symmetric function, increases as the transmission increases, while the relative contributions of the greater index eigenfunctions, which are asymmetric

functions, are higher for low-transmission channels.

2.4.3 Effect of sample thickness

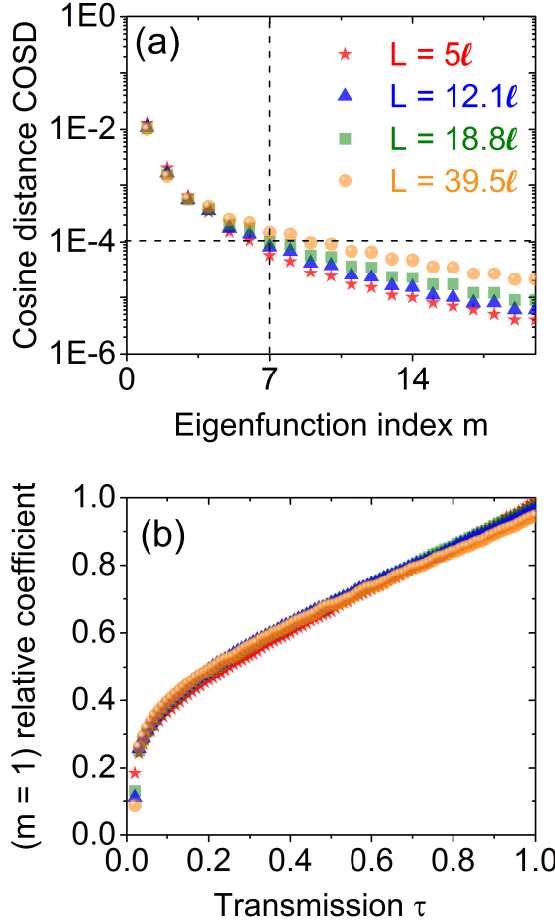


Figure 2.8: (a) Cosine distance COSD versus eigenfunction index m for 4 samples with different thickness: $L = 5\ell, 12.1\ell, 18.8\ell$ and 39.5ℓ , which are plotted as pink stars, green squares, blue triangles, and orange circles respectively. The vertical and horizontal dashed-lines shows position of our figure-of-merit $\text{COSD} \approx 10^{-4}$ and the eigenfunction, which fulfills the criterion $m = 7$ respectively. (b) Contribution of the fundamental diffusion mode $m = 1$ relative to the sum of the first 100 eigenfunctions as a function of transmission τ .

Here, we investigate the effect of the sample thickness on the number of eigenfunctions sufficient to reconstruct the energy density of shaped waves. We therefore perform simulations of the energy density on different samples with a

range of sample thicknesses: $L = 5\ell, 12.1\ell, 18.8\ell$ and 39.5ℓ . We show in Fig. 2.8 the COSD versus the eigensolution index for the 4 samples. Interestingly, all samples have a convergence to $\text{COSD} \leq 10^{-4}$ at the seventh eigensolution. There is a slight deviation for the thickest sample $L = 39.5\ell$ that is probably due to artifacts from the simulation due to the large number of waveguide slices. This convergence of the COSD shows that the summation over the first 7 eigenfunctions of the diffusion equation is sufficient to describe the energy density of shaped waves, *irrespective* of the sample thickness.

In the previous sections, we have seen that the diffusion fundamental eigenfunction of the diffusion equation $m = 1$ dominates the energy density of high-transmission channels. We investigate if this result holds for the different samples thicknesses. In Fig. 2.8, we show the coefficient of the $m = 1$ mode relative to the sum of the first 100 eigenfunctions versus the transmission for the 4 different thicknesses. The relative contribution of the $m = 1$ mode is similar for all samples, irrespective of the thickness and this is quite remarkable. The fundamental diffusion eigenfunction dominates the energy density of high-transmission channels for all sample thicknesses.

2.5 Conclusions

We have shown that only a few eigenfunctions of the diffusion equation suffice to accurately reconstruct the distribution of the shaped energy density inside a quasi-1D scattering waveguides. In particular, the fundamental eigenfunction of the diffusion equation is very similar to the distribution of energy density of shaped waves. To reconstruct the distribution of open channels only the diffusion fundamental eigenfunction $m = 1$ is sufficient. In addition, we have shown that a few diffusion eigenfunctions reconstruct the energy density of low-transmission eigenchannels, to a great precision. The diffusion eigenfunctions are very efficient to reconstruct the energy densities within the boundaries of the sample. This reconstruction is enlightening because even energy densities that are the result of wave phenomena – shaped waves and transmission eigenchannels – can be described efficiently with diffusion eigenfunctions. The diffusion eigenfunctions are capable of this reconstruction since they are a complete set of functions. Our results are relevant for applications that require the precise knowledge of distribution of the energy density inside scattering media. Such applications include efficient light harvesting in solar cells especially in near infrared where silicon has a low absorption [82, 93, 94]; enhanced energy conversion in white LEDs, which serves to reduce the quantity of expensive phosphor [88–92]; low threshold and higher output yield of random lasers [12, 95]; as well as in homogeneous excitation of probes in biological tissues [96].

2.A The wave vectors of diffusion eigenfunctions

In Eq. 2.11, we show an implicit equation, which defines the allowed wave vectors of the diffusion eigenfunctions. The equation is again stated here:

$$\tan(\kappa_m L) = \frac{(z_{e1} + z_{e2})\kappa_m}{z_{e1}z_{e2}\kappa_m^2 - 1}. \quad (2.25)$$

We define

$$f(\kappa_m) = f_1(\kappa_m) - f_2(\kappa_m), \quad (2.26)$$

where

$$f_1(\kappa_m) \equiv \tan(\kappa_m L) \quad (2.27)$$

and

$$f_2(\kappa_m) \equiv \frac{(z_{e1} + z_{e2})\kappa_m}{z_{e1}z_{e2}\kappa_m^2 - 1}. \quad (2.28)$$

Our goal is to find the wave vectors κ_m^r that fulfills the condition

$$f(\kappa_m^r) = 0. \quad (2.29)$$

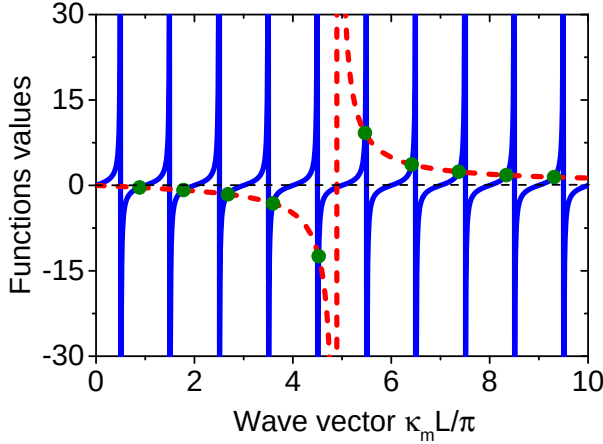


Figure 2.9: The values of functions f_1 (blue line) and f_2 (red dashed line) in Eqs. 2.27 and 2.28. as a function of reduced diffusion wave vector $\kappa_m L/\pi$. The green circles are the roots of Eq. 2.25.

In Fig. 2.9, we plot the two functions f_1 and f_2 . The condition in Eq. 2.29 is fulfilled by the wave vectors κ_m^r at the intersection of the two functions f_1 and f_2 , as indicated in Fig. 2.9. We found the wave vectors κ_m^r by dividing function f into domains with size π/L . In most of the domains, only one root is expected, except the domain where the function f_2 has a divergence, where two roots appear. In each domain, we use a Simple Bisection method to search the domain for wave vectors that fulfill the desired condition. Each domain is

divided into two equal parts. The function f is first evaluated for the left part of the domain and then checked if there is root present. If there is no root present, the left domain is further divided into smaller subdomains and each sub-domain is searched for the root. We divide the domains until the size of the sub-domain is 10^{-12} . If there is no root in all the sub-domains of the left domain, then the right domain is searched. This way, all the desired roots are found iteratively.

CHAPTER 3

Theory of probing energy density inside a 3D scattering medium

3.1 Introduction

Due to multiple scattering, a scattering medium is necessarily opaque and therefore it is impossible to image the light distribution inside a three-dimensional (3D) medium by using standard imaging techniques. It is of great interest to explore the possibility of ‘looking’ inside a scattering medium. Of particular relevance to diffusing light is the energy density that is defined as the amount of energy in a light field that is stored in a given volume [32]. In particular, we are interested in studying the energy density inside a scattering media, when the incident wavefront is shaped to obtain a desired transmitted intensity [49, 52, 53, 56–58, 86, 87].

In Chapter 4 and 5, we introduce an experimental method to probe the energy density of shaped waves inside a scattering medium. As probes, we positioned randomly fluorescent particles inside a scattering medium and they are excited by the incident shaped wave, which we henceforth refer to as optimized light. In this Chapter, we present an elaborate theoretical description of the method of probing the total and depth-dependent energy densities of optimized light. We model the enhancement of the fluorescent intensity exiting the back surface of the sample for incident optimized light compared to unoptimized light. We find that the total energy density is proportional to the total fluorescent intensity, which validates that measuring the fluorescent intensity gives insight to the total energy density inside the scattering medium.

3.2 Assumptions

We first state the three general assumptions that we have made. Firstly, we assume that the scattering medium can be described by a slab geometry with infinite lateral dimensions in the x and y directions. There are however boundaries at $z = 0$ and $z = L$, since light propagates through a very small area on the sample compared to the whole extent of the sample. For a 10 μm -thick sample, the diameter of the diffuse spot at the back surface of the sample is about

A part of this chapter has been published in the appendix of: O. S. Ojambati, H. Yılmaz, A. Lagendijk, A. P. Mosk, and W. L. Vos, New J. Phys. 18, 043032: 1-13 (2016).

20 μm , much smaller than the 40 mm extent of our whole sample. Therefore, we can safely assume that the boundaries in both x and y directions have negligible effect on light diffusion inside the medium. The boundaries along z , however, have an effect on light propagation. Therefore, the three-dimensional diffusion equation becomes effectively a one-dimensional equation that depends on only the z direction [109].

Secondly, we assume that absorption is negligible in the samples. This assumption is valid since the albedo a of our sample with the highest absorption is $a \approx 1$ (see Chapter 4.2.1). However, weak absorption is present in our samples due to the presence of the low density of probe spheres. Therefore, the assumption that there is no absorption in the medium is an extremely mild limitation to our model. If necessary, the diffusion equation can be extended to account for absorption, see Ref. [110].

Thirdly, we assume that multiple scattering events occur inside the medium before light exits the sample, such that the ensemble-average energy density in our samples can be modelled using diffusion theory. Therefore, we assume that the sample thickness L is much larger than the transport mean free path ℓ

$$L \gg \ell \quad (3.1)$$

and much larger than the extrapolation lengths on the front and back side of the sample z_{e1} and z_{e2} respectively

$$L \gg z_{e1}, z_{e2}. \quad (3.2)$$

The extrapolation lengths account for reflections on each side of the sample and are proportional to the transport mean free path

$$z_{en} = \tau_n \ell, \quad (3.3)$$

where the index $n = 1, 2$ and τ_i accounts for the average reflectance at the interfaces [107]. The inequality 3.1 is valid for our samples with thickness L ranging from 3ℓ to 37ℓ , where $\ell = 0.6 \mu\text{m}$. We note that in Ref. [111], it has been shown that diffusion theory is robust to describe thin samples of only $L = 2\ell$ to an accuracy of 2%.

3.3 Modelling the enhancement of the total stored energy

3.3.1 Concept of probing the total stored energy

In Fig. 3.1, we show the concept of probing the total stored energy inside a scattering medium. Fluorescent particles are randomly positioned inside the scattering medium and are reporters of the energy density inside the medium. The incident light excites the fluorescent particles and they emit light with a total power proportional to the energy density at the location of the particles.

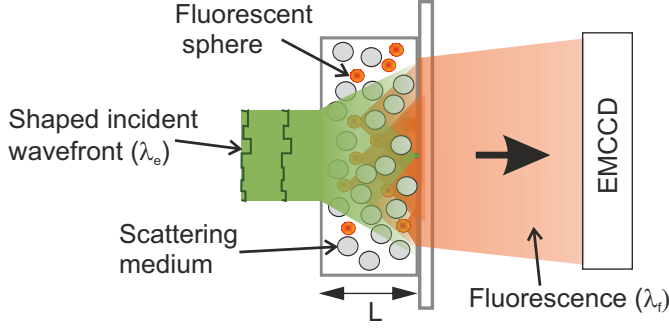


Figure 3.1: Setup to probe the total stored energy inside a scattering medium. The scattering medium is lightly doped with randomly positioned fluorescent spheres to probe the energy density inside the sample. The fluorescent particles are excited by the incident shaped wavefront λ_i (green intensity). The total fluorescent power emitted by the fluorescent spheres at λ_f (red intensity) is detected by a sensitive camera.

The total fluorescent flux exiting the back surface of the sample is measured by an electron multiplying charge coupled device (EMCCD) camera. We note that the scattering medium is lightly doped so that the effect of absorption is only mild, in accordance to our second assumption. Absorption starts to play a significant role in perturbing the field distribution inside a scattering medium when the $\ell \sim 10^{-2}\ell_a$, where ℓ_a is the absorption mean free path, *i.e.*, albedo $a \approx 0.99$ [98, 112]. More experimental details about this method of probing the total energy density are given in Chapter 4.

3.3.2 Spatial distribution of the energy density of wavefront-shaped light

In Chapter 2, we show that the total stored energy of optimized light is underestimated by only 2%, when only the fundamental diffusion eigenfunction is considered. To obtain the total energy density, we therefore model the spatial distribution of optimized light using the fundamental diffusion eigenfunction, which we obtain by solving the one-dimensional diffusion equation

$$\frac{\partial W(z, t)}{\partial t} = D \frac{\partial^2 W(z, t)}{\partial z^2}, \quad (3.4)$$

where $W(z, t)$ is the ensemble-averaged energy density and D is the diffusion constant that is equal to $D = v_E \ell / 3$ with v_E the energy velocity [113]. We use

the Dirichlet boundary condition* [18]

$$W(z, t) = 0 \quad \text{at} \quad \begin{cases} z = -z_{e1} \\ z = L + z_{e2} \end{cases} \quad (3.5)$$

The source is taken to be the result of the exponential decay of the scattered incident coherent beam

$$W(z, t = 0) = \frac{I_0}{z_{\text{inj}}} \exp\left(-\frac{z}{z_{\text{inj}}}\right), \quad (3.6)$$

where I_0 is the incident intensity and z_{inj} is the averaged injection depth at which the incident light becomes diffuse that accounts for the angular distribution of the incident shaped wavefront [114, 115]. We solve Eq. 3.4 using the method of separation of variables [42], to obtain the energy density

$$W(z, t) = \sum_{m=1}^{\infty} A_m \exp\left[\frac{-\pi^2 m^2 D t}{L_{\text{ex}}^2}\right] \sin\left(\pi m \frac{z + z_{e1}}{L_{\text{ex}}}\right) \sin\left(\pi m \frac{z_{\text{inj}} + z_{e1}}{L_{\text{ex}}}\right). \quad (3.7)$$

A_m is a coefficient given by

$$A_m \equiv \frac{2I_0 \left[\pi m z_{\text{inj}} \cos\left(\pi m \frac{z_{e1}}{L_{\text{ex}}}\right) - \zeta_m e^{-\frac{L_{\text{ex}}}{z_{\text{inj}}}} + L_{\text{ex}} \sin\left(\pi m \frac{z_{e1}}{L_{\text{ex}}}\right) \right]}{L_{\text{ex}}^2 + \pi^2 m^2 z_{\text{inj}}^2}, \quad (3.8)$$

where the parameter ζ_m is defined as

$$\zeta_m \equiv \pi m z_{\text{inj}} \cos\left(\pi m \frac{L'_{\text{ex}}}{L_{\text{ex}}}\right) + L_{\text{ex}} \sin\left(\pi m \frac{L'_{\text{ex}}}{L_{\text{ex}}}\right) \quad (3.9)$$

m is the index of the eigenfunctions, $L_{\text{ex}} = L + z_{e1} + z_{e2}$ is the effective sample thickness and $L'_{\text{ex}} \equiv L_{\text{ex}} + z_{e1}$. Since the excitation is a continuous wave source, we integrate out the time-dependent part of Eq. 3.7 to obtain

$$\begin{aligned} W_d(z) &= \sum_{m=1}^{\infty} \frac{A_m L_{\text{ex}}^2}{\pi^2 m^2 D} \sin\left(\pi m \frac{z_{\text{inj}} + z_{e1}}{L_{\text{ex}}}\right) \sin\left(\pi m \frac{z + z_{e1}}{L_{\text{ex}}}\right) \\ &\equiv \sum_{m=1}^{\infty} C_m \sin\left(\pi m \frac{z + z_{e1}}{L_{\text{ex}}}\right), \end{aligned} \quad (3.10)$$

where we define the coefficient

$$C_m \equiv \frac{A_m L_{\text{ex}}^2}{\pi^2 m^2 D} \sin\left(\pi m \frac{z_{\text{inj}} + z_{e1}}{L_{\text{ex}}}\right). \quad (3.11)$$

*The Dirichlet boundary condition is only valid to describe functions that are approximately linear at the boundaries. For arbitrary functions, the mixed boundary condition used in Chapter 2 is more accurate.

The series in Eq. 3.10 can be analytically evaluated [116] to obtain the energy density of unoptimized light

$$W_d(z) = \frac{I_0}{D} \left[\frac{z_{\text{inj}} + z_{e1}}{L_{\text{ex}}} (L + z_{e2} - z) - z_{\text{inj}} \exp\left(-\frac{z}{z_{\text{inj}}}\right) \right]. \quad (3.12)$$

In Fig. 4.1, we plot the unoptimized energy density W_d and it first increases within one transport mean free path and linearly decrease afterwards.

It has been shown experimentally [83] and theoretically [106] that the total transmission of perfectly optimized light is equal to $T_o = 2/3$. We therefore scale the energy density of the fundamental diffusion eigenfunction $W_{m=1}(z)$ such that the total transmission is equal to $T_o = 2/3$. We define a scaling factor α and a scaled energy density of optimized light $W_o(z)$ (corresponding to T_o) such that

$$W_o(z) \equiv \alpha W_{m=1}(z). \quad (3.13)$$

From Eq. 3.10, the fundamental diffusion eigenfunction $W_{m=1}(z)$ is obtained as

$$W_{m=1}(z) = \frac{A_1 L_{\text{ex}}^2}{\pi^2 D} \sin\left(\pi \frac{z + z_{e1}}{L_{\text{ex}}}\right) \sin\left(\pi \frac{z_{\text{inj}} + z_{e1}}{L_{\text{ex}}}\right). \quad (3.14)$$

The total transmission T_{tot} is the ratio of the total flux at the back side of the sample ($z = L$) to the incident flux [32] and it can be expressed as

$$T_{\text{tot}} = -\frac{D}{I_0} \frac{\partial W(z)}{\partial z} \Big|_{z=L}. \quad (3.15)$$

Substituting Eq. 3.13 into 3.15, we obtain the optimal total transmission

$$T_o = \frac{2}{3} = -\alpha \frac{D}{I_0} \frac{\partial W_{m=1}(z)}{\partial z} \Big|_{z=L}. \quad (3.16)$$

Substituting 3.14 for $W_{m=1}$, we then determine the scaling factor

$$\alpha = -\frac{2}{3 T_{m=1}}, \quad (3.17)$$

where $T_{m=1}$ is the contribution of the $m = 1$ mode to the total transmission, and it is given as

$$T_{m=1} = -\frac{A_1 L_{\text{ex}}}{\pi I_0} \cos\left(\pi \frac{L + z_{e1}}{L_{\text{ex}}}\right) \sin\left(\pi \frac{z_{\text{inj}} + z_{e1}}{L_{\text{ex}}}\right). \quad (3.18)$$

Using the trigonometry identity $\cos(\pi - \theta) = -\cos \theta$, we obtain

$$T_{m=1} = \frac{A_1 L_{\text{ex}}}{\pi D} \cos\left(\pi \frac{z_{e2}}{L_{\text{ex}}}\right) \sin\left(\pi \frac{z_{\text{inj}} + z_{e1}}{L_{\text{ex}}}\right). \quad (3.19)$$

Substituting α into Eq. 3.13, we find the energy density of optimized light to be

$$W_o(z) = \frac{2I_0 L_{\text{ex}}}{3\pi D} \frac{\sin\left(\pi \frac{z+z_{e1}}{L_{\text{ex}}}\right)}{\cos\left(\pi \frac{z_{e2}}{L_{\text{ex}}}\right)}. \quad (3.20)$$

In Fig. 4.1, we plot the scaled energy density of optimized light W_o and it is a nearly symmetric function that is peaked around the center of the sample.

3.3.3 Enhancement of total stored energy and fluorescent power

We derive the enhancement of the total stored energy η_{ed} inside the sample, for optimized light compared to unoptimized light. We define the enhancement as

$$\eta_{\text{ed}}(L) \equiv \frac{W'_o}{W'_d}, \quad (3.21)$$

where W'_o and W'_d are the total stored energies of optimized light and unoptimized light respectively. Integrating Eqs. 3.12 and 3.20, we obtain

$$\eta_{\text{ed}}(L) = \frac{2L_{\text{ex}}^2 \sec\left(\pi \frac{z_{e2}}{L_{\text{ex}}}\right) [\cos\left(\pi \frac{z_{e1}}{L_{\text{ex}}}\right) - \cos\left(\pi \frac{L'}{L_{\text{ex}}}\right)]}{3\pi^2 \left[\frac{L z'_{\text{inj}} (L + 2z_{e2})}{2L_{\text{ex}}} - z_{\text{inj}}^2 (1 - e^{-\frac{L}{z_{\text{inj}}}}) \right]} \quad (3.22)$$

and we plot η_{ed} as a function of sample thickness L in Fig. 3.2. For thick samples, η_{ed} is linearly dependent on L .

In our experiments, we probe the total stored energy by using fluorescent spheres positioned inside the sample as reporters. We now proceed to model the enhancement of the fluorescent power measured at the back surface of the sample. We consider the small fluorescent spheres as point emitters located at a depth z_f inside the scattering medium. The diffuse emission from the fluorescent spheres is also described by the diffusion equation and with a Dirac delta function as the source term [117, 118]. At steady-state, the solution is

$$W_f(z) = \frac{P_{\text{ex}}(z_f)}{D} \begin{cases} \frac{L - z_f + z_{e2}}{L + z_{e1} + z_{e1}} (z + z_{e1}) & 0 \leq z \leq z_f \\ \frac{z_f + z_{e1}}{L_{\text{ex}}} (L + z_{e2} - z) & z_f \leq z \leq L \end{cases}, \quad (3.23)$$

where $W_f(z)$ is the energy density of fluorescent light and $P_{\text{ex}}(z)$ is the excitation power, which is obtained from Eqs. 3.20 and 3.12 for optimized light and unoptimized light respectively. From Eq. 3.23, we obtain the fluorescent energy flux exiting the back surface of the sample and it is given as

$$F_f(\lambda_i, \lambda_f) = \frac{P_{\text{ex}}(z_f, \lambda_i)}{D(\lambda_f) L_{\text{ex}}(\lambda_f)} [z_f + z_{e1}(\lambda_f)]. \quad (3.24)$$

Since the parameters in Eq. 3.24 are wavelength dependent, we evaluate the parameters at the appropriate wavelengths.

The fluorescent energy flux $F(z = L)$ exiting the sample is the integral of the fluorescence energy density reaching the back of the sample from all probes located inside the medium and is equal to

$$F(z = L, \lambda_i, \lambda_f) = \int_{z_f=0}^{z_f=L} \frac{P_{\text{ex}}(z_f, \lambda_i)}{D(\lambda_f)L_{\text{ex}}(\lambda_f)} [z_f + z_{\text{e1}}(\lambda_f)] dz_f. \quad (3.25)$$

We define the enhancement in the fluorescent power η_f^p as

$$\eta_f^p(L, \lambda_i, \lambda_f) \equiv \frac{F_o(L, \lambda_i, \lambda_f)}{F_d(L, \lambda_i, \lambda_f)}, \quad (3.26)$$

where F_o and F_d are the total fluorescence energy flux at the back surface of the sample for optimized and unoptimized light respectively. To calculate F_o , we obtain the excitation power $P_{\text{ex}}(z_f, \lambda_i)$ from the optimized energy density (in Eq. 3.20), which is proportional to power from its dimension. We then substitute $P_{\text{ex}}(z_f, \lambda_i)$ into Eq. 3.25 to obtain

$$F_o(L, \lambda_i, \lambda_f) = \int_{z_f=0}^{z_f=L} \frac{S_o(\lambda_i, \lambda_f)}{\cos\left(\pi \frac{z_{\text{e2}}(\lambda_i)}{L_{\text{ex}}(\lambda_i)}\right)} [z_f + z_{\text{e1}}(\lambda_f)] \sin\left(\pi \frac{z_f + z_{\text{e1}}(\lambda_i)}{L_{\text{ex}}(\lambda_i)}\right) dz_f, \quad (3.27)$$

where the parameter

$$S_o(\lambda_i, \lambda_f) \equiv \frac{2I_0 L_{\text{ex}}(\lambda_i)}{3\pi D(\lambda_f) D(\lambda_i) L_{\text{ex}}(\lambda_f)}. \quad (3.28)$$

Similarly, we obtain F_d by using the unoptimized energy density in equation 3.12 as $P_{\text{ex}}(z_f, \lambda_i)$ in Eq. 3.25 to obtain

$$F_d(z = L, \lambda_i, \lambda_f) = \int_{z_f=0}^{z_f=L} S_d(\lambda_i, \lambda_f) [(L + z_{\text{e2}}(\lambda_i) - z_f) - z_{\text{inj}} \exp(-\frac{z_f}{z_{\text{inj}}})] [z_f + z_{\text{e1}}(\lambda_f)] dz_f, \quad (3.29)$$

where

$$S_d(\lambda_i, \lambda_f) \equiv \frac{I_0 [z_{\text{inj}} + z_{\text{e1}}(\lambda_i)]}{D(\lambda_f) D(\lambda_i) L_{\text{ex}}(\lambda_f) L_{\text{ex}}(\lambda_i)}. \quad (3.30)$$

Evaluating the integrals in Eq. 3.27 and Eq. 3.29 and substituting into Eq. 3.26,

we obtain

$$\eta_f^p(L, \lambda_i, \lambda_f) = \frac{K \left[\pi z_{e1}(\lambda_f) \cos\left(\frac{\pi z_{e1}(\lambda_i)}{L_{\text{ex}}}\right) - L_{\text{ex}} \left[\sin\left(\frac{\pi z_{e1}(\lambda_i)}{L_{\text{ex}}}\right) - \sin\left(\frac{\pi L'_i}{L_{\text{ex}}}\right) \right] - \pi L'_f \cos\left(\frac{\pi L'_i}{L_{\text{ex}}}\right) \right]}{\Gamma(\lambda_i, \lambda_f) + z_{\text{inj}}^2 [L'_f + z_{\text{inj}}] e^{-\frac{L}{z_{\text{inj}}}} - z_{\text{inj}}^2 [z_{\text{inj}} + z_{e1}(\lambda_f)]}, \quad (3.31)$$

where

$$K = \frac{2L_{\text{ex}}^2 \sec\left(\frac{\pi z_{e2}(\lambda_i)}{L_{\text{ex}}}\right)}{3\pi^3}, \quad (3.32)$$

$$\Gamma(\lambda_i, \lambda_f) = \frac{L[z_{\text{inj}} + z_{e1}(\lambda_i)]}{6L_{\text{ex}}} [L^2 + 3L(z_{e1}(\lambda_f) + z_{e2}(\lambda_i)) + 6z_{e1}(\lambda_f)z_{e2}(\lambda_i)], \quad (3.33)$$

and $L'_i = L + z_{e1}(\lambda_i)$, $L'_f = L + z_{e1}(\lambda_f)$, and $L_{\text{ex}} = L + z_{e1}(\lambda_i) + z_{e2}(\lambda_i)$. The dependency of η_f^p on the sample thickness is shown in Fig. 3.2 and for thick samples, η_f^p linearly depends on L .

In order to relate the enhancement in fluorescent power $\eta_f(L)$ to the enhancement in energy density η_{ed} in Eq. 3.22, we find the Taylor expansion of $\eta_{\text{ed}}(L)$ and $\eta_f(L)$ which holds only when $L \gg \ell$. We derive

$$\eta_{\text{ed}}(L, \lambda_i, \lambda_f) = \frac{2}{3} \eta_f^p(L, \lambda_i, \lambda_f) + O(\ell/L), \quad (3.34)$$

where $O(\ell/L)$ includes higher orders of the series expansion in terms of ℓ/L . From 3.34, we learn that the increase of the detected fluorescence is a measure of the increase of the energy density. We plot in Fig. 3.2, the total energy density η_{ed} using Eq. 3.22 and the fluorescent intensity enhancement η_f^p multiplied by a factor $2/3$ using Eq. 3.31. The purpose of the multiplying factor $2/3$ is to compare η_f^p with η_{ed} , according to Eq. 3.34. The scaled η_f^p differs from η_{ed} by about 30% and this is a result contribution of the higher order terms in the series expansion in Eq. 3.34. The close relationship between the two curves implies that Eq. 3.34 is valid and the total fluorescent intensity is truly a measure of the total stored energy.

3.4 Modelling the three-dimensional energy density

The 3D energy density of light optimized to focus to a point by wavefront shaping is denoted $W_o(x, y, z)$. Due to the cylindrical symmetry around the optical axis z , $W_o(x, y, z)$ is denoted as $W(\boldsymbol{\rho}, z)$, where $\boldsymbol{\rho} \equiv (x, y)$. Following the maximal fluctuation theorem [119, 120], we assume that the transmission channel in the sample consists of only open and closed channels. This theorem implies that there are only ‘on’ and ‘off’ states and the intermediate states are neglected.

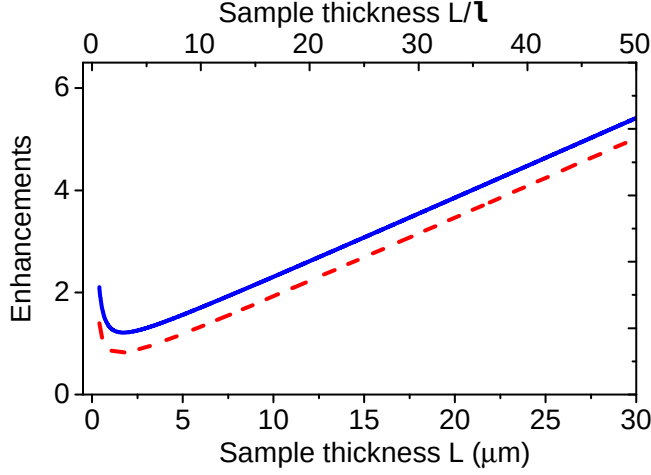


Figure 3.2: Enhancements of the total stored energy η_{ed} (blue solid line) and total fluorescent intensity η_f^p (red dashed line) as a function of sample thickness L . We scaled η_f^p by a factor of $2/3$, in order to compare with η_{ed} , according to Eq. 3.34. η_{ed} and η_f^p were plotted using Eqs. 3.22 and 3.31 using the sample parameters given in Table 4.1.

Therefore we make the simplifying assumption that optimized light couples fully to only open channels. For a fully open channel we know from Chapter 2 that the ρ -integrated energy density $W_o(z)$ is well described using only the fundamental diffusion eigenfunction, and therefore

$$W_o(z) = A W_{m=1}(z) = 2\pi \int_0^\infty \rho d\rho W_o(\rho, z), \quad (3.35)$$

where A is a proportionality constant.

The optimized focus can be seen as the time reverse (phase conjugate) of a focused beam entering the sample from the back surface [83, 87] (see Fig. 3.3), whose ensemble average energy density $W_{tr}(\rho, z)$ is described by the 3D diffusion equation (see Eq. 5.2). We model the energy density of optimized light as a sum of three components:

$$W_o(\rho, z) \equiv W_{fo}(\rho, z) + W_{bg}(\rho, z) + W_{dif}(\rho, z, |\gamma|^2). \quad (3.36)$$

(The functions $W_{fo}(\rho, z)$ and $W_{bg}(\rho, z)$ are illustrated in Fig. 3.3). W_{fo} is the energy density of light propagating from the front surface to the back surface of the sample to obtain the optimized focus, W_{bg} is a background contribution due to incomplete time reversal, since the closed channels are not reversed, and diffuse light W_{dif} is a result of imperfect wavefront shaping and experimental noise. Here, we assume that the wavefront shaping experiment is perfect, *i.e.*, $\gamma = 1$, therefore diffuse energy density vanishes: $W_{dif}(\rho, z, |\gamma|^2 = 1) = 0$. Corrections

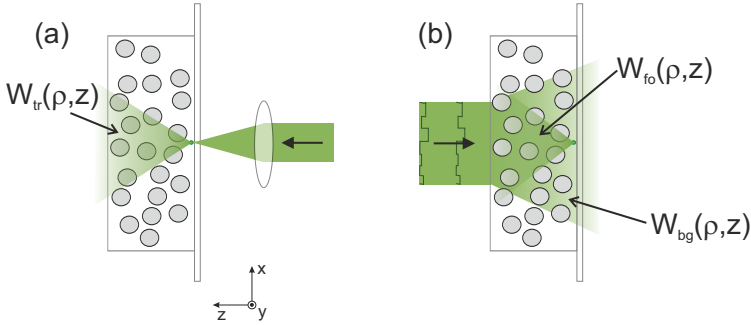


Figure 3.3: An illustration of the functions to describe optimized light in 3D inside a scattering medium. (a) A laser beam is focused at the back surface of the sample and the energy density $W_{tr}(\rho, z)$ of diffuse light is described using the diffusion equation. The focused beam is the time reverse of the component of optimized light that contributes to the optimized focus. (b) Optimized light propagating from left to right focuses light at the back surface of the sample. The energy density $W_o(\rho, z)$ of optimized light has two contributions: light that goes to the optimized focus $W_{fo}(\rho, z)$ and the background $W_{bg}(\rho, z)$. The cartoon shows wavefront shaping in the absence of experiment imperfections, therefore $W_{dif} = 0$.

for imperfect shaping can be made through the extrapolation method used in Chapters 4 and 5.

At first sight, one might think that the energy density W_{fo} is equal to W_{tr} . However, this reasoning is not plausible as W_{fo} contains only open channels, therefore its profile along z must follow $W_{m=1}(z)$. Therefore, to obtain W_{fo} , W_{tr} is normalized and mapped onto the profile of $W_{m=1}(z)$ using the equation

$$W_{fo}(\rho, z) = A_1 \frac{W_{tr}(\rho, z)}{2\pi \int_0^\infty \rho' d\rho' W_{tr}(z, \rho')} W_{m=1}(z), \quad (3.37)$$

where A_1 is an amplitude factor. The background term W_{bg} does not contribute to the optimized focus, and since we are looking at perfect shaping in the maximal fluctuation approximation, W_{bg} consists of only open channels. We describe the background term W_{bg} by mapping $W_{m=1}$ onto a Gaussian profile with a constant width ρ_0 along z

$$W_{bg}(\rho, z) = A_2 W_{m=1}(z) \exp[-\rho^2/\rho_0^2], \quad (3.38)$$

where A_2 is a constant indicating the level of background intensity. The constants $A_{1,2}$ are chosen such that the intensity pattern of the optimized light at the back surface is obtained. We determine A_1 from the fact that when $|\gamma|^2 = 1$, the total intensity in the optimized focus is proportional to the total transmission T_{tot} , therefore

$$A_1 = T_{tot} I_0, \quad (3.39)$$

where I_0 is the incident intensity. The intensity in the background is therefore the total transmitted intensity of an open channel minus the intensity in the focus,

$$A_2 = (T_o - T_{\text{tot}})I_0, \quad (3.40)$$

where T_o is the the total transmission of an open channel equal to $T_o = 1$. Similar to Eq. 3.38, we obtain the 3D energy density of unoptimized light $W_{uo}(\boldsymbol{\rho}, z)$ to be

$$W_{uo}(\boldsymbol{\rho}, z) = W_d(z) \exp[-\boldsymbol{\rho}^2/\rho_0^2], \quad (3.41)$$

where $W_d(z)$ is the solution of the 1D diffusion equation and is obtained from Eq. 3.12. We have neglected the diffuse ‘fanning’ out of light, since the increase in the beam width is small compared to the full width of the beam. The model can be improved to account for this effect.

The procedure described above guarantees that the radially integrated energy density equals the one for open channels: $W_o(z) = W_{m=1}(z)$. To check, we compared the total stored energy using $W_o(z)$ and $W_{m=1}(z)$ and we obtained 1.95 and 1.92 using $W_o(z)$ and $W_{m=1}(z)$ respectively for a sample with $L = 13 \mu\text{m}$. The two values agree very well and this confirms that the procedure works well. In Fig. 3.4, we show the calculated energy densities at various depths inside a scattering medium. For optimized light in Fig. 3.4(a), the energy density spreads and converges at the back surface of the sample. For unoptimized light in Fig. 3.4(b), the energy density decreases as the depth approaches the back surface of the sample, in agreement with the 1D solution of the diffusion equation. To the best of our knowledge, this is the first description of the energy density of light in 3D.

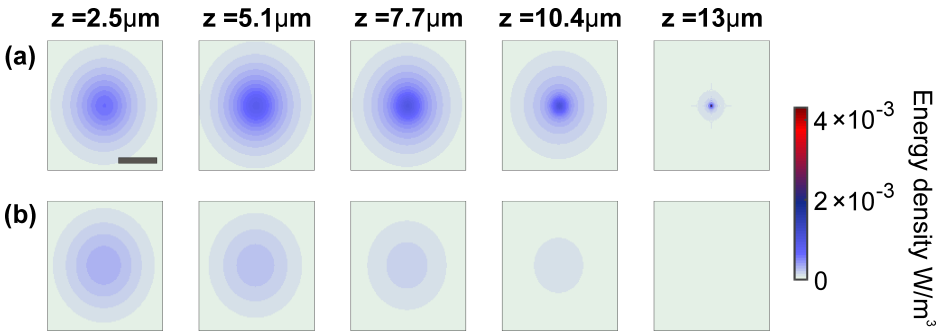


Figure 3.4: Images of calculated energy densities at various depths inside a scattering medium for optimized (a) and unoptimized light (b) using Eqs. 3.36 and 3.41 respectively. The sample thickness is $L = 13 \mu\text{m}$. The scale bar is $4 \mu\text{m}$.

3.5 Enhancement of the depth-dependent fluorescent intensity

In the experiment to measure the depth-dependent energy density in Chapter 5, we measure the fluorescent intensity from probes located at specific depth z_f and a lateral position $\boldsymbol{\rho}_f = (x_f, y_f)$ inside a 3D scattering medium. Using the 3D energy densities obtained in Section 3.4, we model the enhancement of the fluorescent power η_f^s from a fluorescent particle. We define η_f^s as

$$\eta_f^s \equiv \frac{F_o^s(\boldsymbol{\rho}_f, z_f; \lambda_i, \lambda_f)}{F_d^s(\boldsymbol{\rho}_f, z_f; \lambda_i, \lambda_f)}, \quad (3.42)$$

where the fluorescence energy fluxes F_o^s and F_d^s correspond to optimized W_o and unoptimized light W_{uo} , respectively, and are obtained from Eq. 3.24. The flux due to unoptimized light is

$$F_o^s(\boldsymbol{\rho}_f, z_f; \lambda_i, \lambda_f) = 2\pi \int_{\boldsymbol{\rho}_f - r}^{\boldsymbol{\rho}_f + r} \int_{z_f - r}^{z_f + r} \boldsymbol{\rho}'_f d\boldsymbol{\rho}'_f dz'_f cW_o(\boldsymbol{\rho}'_f, z'_f) \frac{z'_f + z_{e1}(\lambda_f)}{D(\lambda_f)L_{ex}(\lambda_f)}, \quad (3.43)$$

where the finite integrals account for the finite volume of the probe particle, c is a proportionality constant relating power to energy density, $\boldsymbol{\rho}'_f$ and z'_f are integration variables, and r is the radius of the particle. Similarly, the flux due to unoptimized light is

$$F_d^s(\boldsymbol{\rho}_f, z_f; \lambda_i, \lambda_f) = 2\pi \int_{\boldsymbol{\rho}_f - r}^{\boldsymbol{\rho}_f + r} \int_{z_f - r}^{z_f + r} \boldsymbol{\rho}'_f d\boldsymbol{\rho}'_f dz'_f cW_{uo}(\boldsymbol{\rho}'_f, z'_f) \frac{z'_f + z_{e1}(\lambda_f)}{D(\lambda_f)L_{ex}(\lambda_f)}. \quad (3.44)$$

Substituting Eqs. 3.43 and 3.44 into Eq. 3.42, we obtain the depth-dependent fluorescence enhancement η_f^s .

In Fig. 3.5, we plot η_f^s as a function of the reduced depth of the fluorescent particle z_f/ℓ using Eq. 3.42 for particles at different $\boldsymbol{\rho}_f$: η_f^s increases from the front surface of the sample and peaks at the back surface for all $\boldsymbol{\rho}_f$. The enhancement η_f^s for a particle located at the center of the illuminated area is about 2 to 4 times higher than probing fluorescent particles displaced by $\boldsymbol{\rho}_f = 3\mu\text{m}$ and $6\mu\text{m}$ from the center, respectively. The plots suggest that the optimized focus has a significant effect on the fluorescent intensity from the particles.

3.6 Summary

We have derived a theory to predict the energy density inside a three-dimensional scattering medium. We derived an expression for the enhancement of the total stored energy and total fluorescent intensity from fluorescent particles randomly positioned inside the sample. We find that the total stored energy inside the sample is proportional to the total fluorescent intensity. We have also presented a model that describes the 3D energy density and used the result to derive the

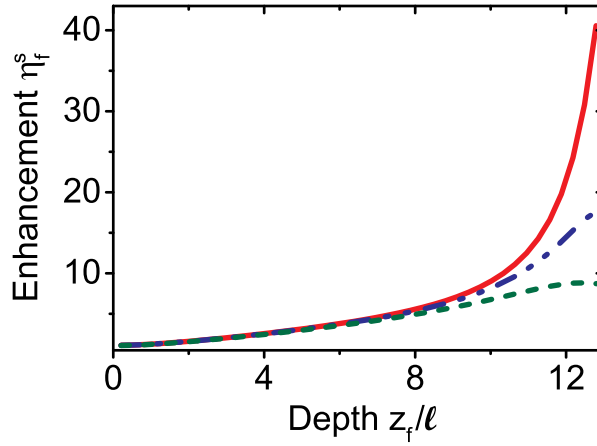


Figure 3.5: Depth-dependent fluorescent enhancement η_f^s as a function of reduced depth z_f/ℓ using Eq. 3.42. The red solid, blue dash-dot, and green dash curves are with positions of the probing particle $\boldsymbol{\rho}_f = (x_f, y_f) = (x_c, y_c) = (6.5 \mu\text{m}, 6.5 \mu\text{m})$, $(x_c + 3 \mu\text{m}, y_c)$, and $(x_c + 6 \mu\text{m}, y_c)$ respectively. (x_c, y_c) is the center of the illuminated area. The sample thickness $L = 13 \mu\text{m}$ and the other sample parameters used are given in Table 4.1.

depth-dependent fluorescent enhancement. The results of our model is compared to experimental result in Chapters 4 and 5.

CHAPTER 4

Coupling of energy into the fundamental diffusion mode of a complex nanophotonic medium

4.1 Introduction

Diffusion is a process that leads to a uniform spreading of matter or energy as a result of randomness. Numerous physical phenomena are described by diffusion, ranging from colloidal particles to currencies to animal motion [6, 32, 40, 46, 121–124]. Many wave phenomena are also well described by diffusion: upon averaging over the disorder, interference is washed out [6, 125, 126]. Hence, waves become diffuse after a distance of the order of one transport mean free path ℓ , i.e., the distance over which the memory of the incident light direction is lost [6]. Of particular relevance to diffusing waves is the energy density that is defined as the amount of energy in a wave field that is stored in a unit volume [32, 39]. Due to diffusion, the ensemble-averaged energy density of the wave W_d has a characteristic shape shown in Fig. 4.1(a) [18, 32, 123, 127–129]. The gradient of the energy density at the exit surface of the scattering medium is related to the transport of energy, which yields the wave-equivalent of the well-known Ohm’s law $T \approx \ell/L$, with L the thickness of the medium.

A fundamental question addressed here is whether it is feasible to couple energy into a diffusion eigenfunction, which has a greater energy density than that of the usual non-optimized energy density W_d , as shown in Fig. 4.1(a). In case of light, an enhanced energy density inside the scattering medium is important for applications such in white LEDs [88–91], solar cells [82, 93, 94], random lasers [12, 95, 130], and biomedical optics [96].

The ensemble-averaged energy density W is described by the diffusion equation as

$$\frac{\partial W(\mathbf{r}, t)}{\partial t} = D \nabla^2 W(\mathbf{r}, t), \quad (4.1)$$

where D is the diffusion constant. For a real and finite medium, such as the widely studied slab geometry, there is translational invariance in the $\rho = (x, y)$ plane [18]. The steady-state solution $W_d(z)$ along the direction of light propagation z is given

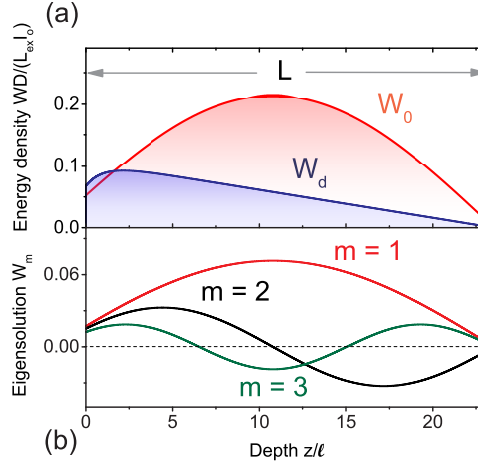


Figure 4.1: (Color) (a) The energy density of optimized light W_o and non-optimized light W_d are shown as the red and blue curves respectively. W_o and W_d are plotted using equations in Chapter 3. The energy density is reduced with the diffusion constant D , the incident intensity I_o , and the effective thickness L_{ex} . (b) The first three eigenfunctions W_m of the diffusion equation in equation Chapter 3. The parameters used in these plots are: $z_{\text{inj}} = 0.86\ell$, $z_{e1}=1.96\ell$, $z_{e2}=0.68\ell$.

by [42],

$$W_d(z) = \sum_{m=1}^{\infty} W_m(z) = \sum_{m=1}^{\infty} C_m \sin\left(\pi m \frac{z + z_{e1}}{L_{\text{ex}}}\right), \quad (4.2)$$

where m is the eigenfunction index, C_m is the relevant coefficient (see Chapter 3), and $L_{\text{ex}} = L + z_{e1} + z_{e2}$ is the effective thickness of the sample, where z_{e1} and z_{e2} are the extrapolation lengths at the front and back surfaces of the sample, respectively. Fig. 4.1(b) shows the first three eigenfunctions. The fundamental eigenfunction $m = 1$ is the only physical solution with a positive energy density along the sample depth z , as shown in Fig. 4.1b. When a plane wave is incident on a scattering medium, energy is coupled into all eigenfunctions that add up to the usual energy density W_d shown in Fig. 4.1(a) [131].

Recently, it has been demonstrated that waves in complex media can be exquisitely controlled by shaping the spatial phase of the incident waves, notably by feedback-based wavefront shaping [48, 49, 83, 84, 86, 87, 132], time reversal [52, 53], phase conjugation [54, 56], and transmission matrix-based control [57–59]. In wavefront shaping, the spatial phase of the field incident on the scattering medium is controlled to enhance the intensity several hundred times in a single speckle spot (area $A = \lambda^2/2\pi$) at the back surface of the sample [133]. With this method, the total transmission T through a scattering medium can be made to differ from Ohm's law [83, 84]. Therefore, it is relevant to investigate whether the energy density inside a scattering medium can be controlled. In particular, we wonder if wavefront shaping couples energy into the diffusion fundamental mode, which has a higher energy density. To date, only numerical

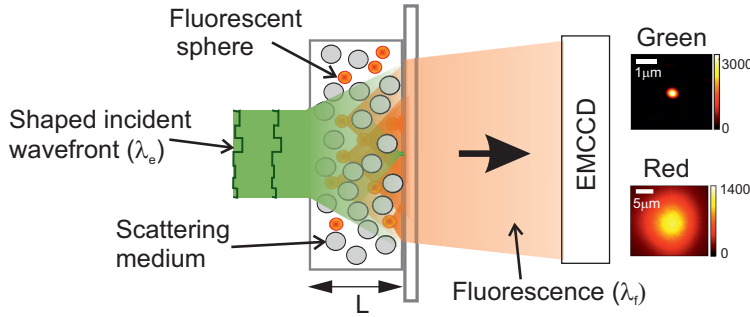


Figure 4.2: (Color) Setup to probe the total energy density inside a scattering medium. The scattering medium is a slab of disordered ZnO particles in air. The medium is illuminated with a shaped incident wavefront such that incident light at λ_i (green intensity) is optimized in a speckle spot at the back surface of the sample (see inset). The scattering medium is lightly doped with randomly positioned fluorescent spheres to probe the energy density inside the sample. The total fluorescent power emitted by the fluorescent spheres at λ_f (red intensity) is detected by the EMCCD (see inset).

calculations [71, 97, 98] and a single-realization elastic wave experiment [99] have addressed the distribution of the energy density inside two-dimensional (2D) scattering media. Control of the optical energy density inside a three-dimensional (3D) scattering medium has so far not been explored.

In this chapter, we experimentally demonstrate the coupling of light predominately into the fundamental diffusion mode by wavefront shaping. Since it is difficult to probe waves inside a 3D material, we use embedded fluorescent nanospheres as reporters. The incident shaped waves excite the fluorescent spheres, whose fluorescent intensity is proportional to the local energy density. By averaging over many fluorescent spheres distributed throughout the samples, we probe the total internal energy of the 3D scattering medium. We observe that the total optical energy is enhanced when the wavefront of the incident light is optimized. The enhancement increases with sample thickness. To interpret our results, we propose a parameter-free model wherein the energy density of wavefront-shaped light is described by the fundamental eigenfunction of the diffusion equation, which agrees well with the experimental observations.

4.2 Experimental details

4.2.1 Samples and preparation

In our experiments, we studied scattering media that are slabs of zinc oxide (ZnO) nanoparticles (Sigma-Aldrich Zinc Oxide 205532, average grain size of 200 nm). Using a professional airbrush (Harder & Steenbeck Airbrush Evolution Silverline, 123003), we sprayed an aqueous suspension of 2.5 g of the ZnO nanoparticles and a suspension of the fluorescent particles, which are dye-doped polystyrene

spheres (Thermo Scientific R50 Fluoro-Max Dyed Red, diameter $d = 50$ nm). The concentration of the fluorescent spheres is $1.5 \mu\text{m}^{-3}$ in 7.5 ml of deionized water. The two suspensions were thoroughly mixed and then spray painted on a cleaned microscope glass slide of thickness 0.17 mm with lateral dimensions of $25 \text{ mm} \times 50 \text{ mm}$. In order to obtain a well-defined thickness of our samples, we automated the spray-painting process by using a LEGO mindstorm kit. The samples have thicknesses L ranging from $2 \mu\text{m}$ to $22 \mu\text{m}$, as a result of varying the spraying time. Inside the samples, the fluorescent spheres are randomly dispersed (see Fig. 4.2) to probe the total energy density inside the ZnO scattering medium. The fluorescent spheres are excited by incident light with wavelength $\lambda_i = 561$ nm and emit fluorescent light at a different wavelength $\lambda_f = 612$ nm.

In order to ensure that the distribution of the energy density at λ_i inside the scattering medium is not perturbed by the absorption from the probing fluorescent spheres [98, 112], we use fluorescent spheres with low absorption probability of $2 \times 10^{-4}\%$ [134]. The absorption probability is the ratio of the absorption cross section to the physical cross section of the fluorescent spheres. Moreover, the concentration of the fluorescent spheres in the samples is as low as $0.35 \mu\text{m}^{-3}$, which results into an albedo $a \approx 1 - 10^{-6}$ averaged over all the fluorescent spheres [135]. The total fluorescent power emitted from the sample fluctuates by 10% from different positions on the sample, which shows that the fluorescent particles are fairly homogeneously distributed.

4.2.2 Experimental set-up

The experimental set-up is shown in Fig. 4.3. The coherent light source is a diode-pumped solid state laser (Cobolt Jive, 100 mW continuous wave output at 561 nm). We control the laser power using a combination of a half-wave plate ($\lambda/2$) and a polarizing beam splitter (PBS). A beam expander with magnification of 3 expands the beam and lens L1 ($f = 11$ mm) focuses the beam onto the front surface of a single mode fiber (SMF) (Thorlabs P3-460B-FC-1). The single mode fiber spatially filters the laser beam in order to prevent pointing instability of the beam. A fiber collimator C (Kirchoff 60FC-L-0-M75-26) collimates the spatially-cleaned beam and it passes through an aperture A1 to illuminate a phase-only liquid-crystal spatial light modulator (SLM) (Holoeye PLUTO-VIS-014-C). The diameter of the beam on the SLM is 8 mm. We used the piecewise sequential algorithm described in Ref. [49] to find an optimized incident wavefront. The image of the SLM is demagnified and imaged to the pupil of a microscope objective MO1 (Nikon: Infinity corrected, $100\times$, $\text{NA} = 0.9$) through a telescope consisting of lenses L2 ($f = 200$ mm) and L3 ($f = 100$ mm). An aperture A2 filters out the higher diffraction orders of the SLM's pixels and transmits only the 0^{th} order. The demagnified image of the SLM is focused onto the surface of the sample by the microscope objective MO1 to a diffraction-limited spot of 620 nm, when wavefront of light is flat. The back surface of the sample is imaged onto a charge-coupled device (CCD) camera (Allied Vision Technologies Stingray F-145) and an electron multiplying CCD (EMCCD) camera (Andor iXon Ultra 897) by an oil-immersion microscope objective MO2 (Nikon: Infinity corrected, $60\times$,

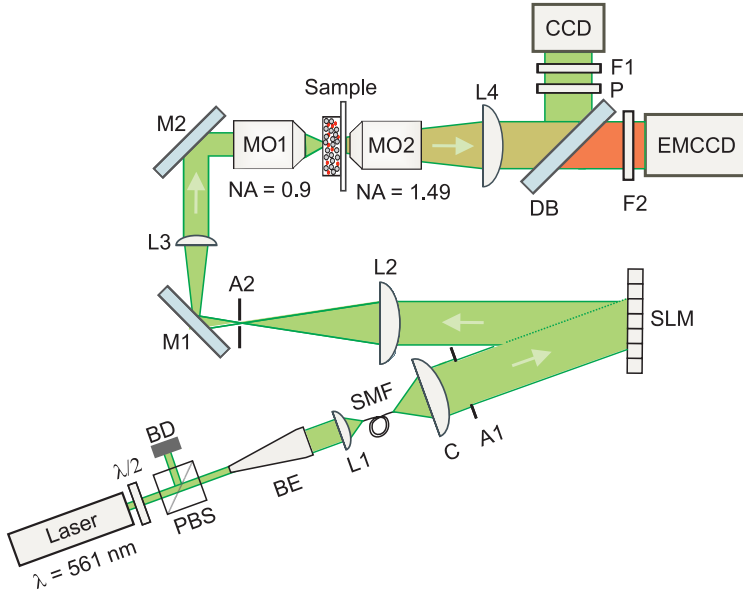


Figure 4.3: (Color online) **Experimental setup:** A spatially-filtered and expanded CW laser at 561 nm illuminates a spatial light modulator (SLM). The beam size on the SLM is 8 mm. A demagnified image of the surface of the SLM is focused onto a scattering sample of ZnO nanoparticles doped with fluorescent spheres. The back surface of the sample is imaged onto the EMCCD camera, which detects the fluorescent intensity and a CCD camera, which detects the excitation green laser light. The dichroic beam splitter separates the fluorescent intensity from the excitation beam. $\lambda/2$ is a half-wave plate, PBS is a polarizing beam splitter, BD is a beam dump, BE is a beam expander, L1, L2, L3, and L4 are lenses with a focal distances of 11 mm, 200 mm, 100 mm, and 500 mm respectively, SMF is a single mode fiber, C is a collimator, A1 and A2 are apertures, M1 and M2 are mirrors, MO1 and MO2 are microscope objectives, DM is a dichroic beam splitter, F1 and F2 are filters, and P is a polarizer.

NA = 1.49) and an achromatic lens L4 ($f = 500$ mm). A dichroic beam splitter DB (Semrock Di02-R561-25 $\times$ 36) reflects the excitation green beam and transmits the emitted fluorescent intensity from the probes. The dichroic beam splitter has a transmission of 93% from 578.4 nm to 1200 nm. A combination of a notch filter (Semrock NF03-561E-25) and a single-band bandpass filter F2 (Semrock FF01-620/52-25) blocks the remnant excitation green light. The green light is detected by the CCD camera. A polarizer P blocks the orthogonal polarization that is not controlled by the SLM. The intensity of the laser beam is reduced by a neutral density filter F1 with an optical density of 2 to prevent saturating the CCD camera.

4.2.3 Measurement procedure

To obtain ensemble-averaged data that can be interpreted with theory, we performed wavefront shaping experiments at 100 to 130 different positions on each sample. At each position, we determine the fluorescent power enhancement η_f that is defined as $\eta_f \equiv P_f^o/P_f^n$, where P_f^o is the total fluorescent power with an optimized phase pattern on the SLM, and P_f^n is the total fluorescent power for a reference incident wavefront. As a reference we use a phase pattern stored from an optimization on a very different part of the sample. This reference field experiences the same diffraction efficiency and surface reflection as our optimized field, in contrast to a plane wave.

4.2.4 Fidelity of optimization

The incident optimized field in any wavefront shaping experiment is a superposition of the ideally optimized incident field, and a noise field that is by definition orthogonal to the ideal field [83]. The fidelity $|\gamma|^2$ quantifies the overlap between the experimentally obtained incident field and the ideal incident field that optimally couples light to the target spot [83]. A perfectly optimized field has $|\gamma|^2 = 1$, while all imperfections such as limited spatial control, temporal decoherence or modulation noise, inevitably reduce $|\gamma|^2$. Here we neglect a correction of the order of $1/N$, where N is the number of transmitting channels $N \approx 10^5$, see [117]. The measured fluorescent power also consists of a linear combination of the ideal incident field term and a noise term. Since the noise field is unrelated to the sample properties, its response is on average equal to that of an unoptimized incident field with the same power. Thus the fluorescent power enhancement η_f necessarily depends on fidelity $|\gamma|^2$ as

$$\eta_f = \eta_f^p |\gamma|^2 + (1 - |\gamma|^2). \quad (4.3)$$

Here η_f^p is the fluorescent power enhancement at perfect fidelity ($|\gamma|^2 = 1$) due to a perfectly shaped incident field. The second term on the right hand side of 4.3 is the contribution from the noise fields. To measure the fidelity of each generated field, we exploit the fact that $|\gamma|^2$ is well approximated by the ratio of the power in the target spot (Fig. 4.2, inset) to the average total transmitted power for an unoptimized incident field [83]. This approximation holds up to the level of fluctuation of the total transmittance, which is about 2% [15]. In practice, the determination of fidelity is limited by experimental noise, which is estimated to be about $\pm 7\%$. Factors such as inhomogeneous sample thickness, experimental noise, and instability in environmental conditions limit $|\gamma|^2$, and result in variations thereof [50, 136]. Although variations are not desired, they have the advantage of yielding a range of $|\gamma|^2$ to investigate. In each experiment at a different position on the sample, we determined both the fidelity $|\gamma|^2$ and the fluorescent power enhancement η_f .

4.3 Experimental results

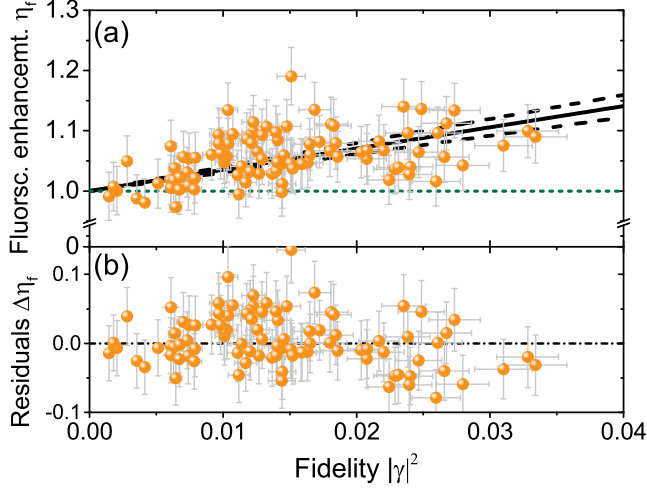


Figure 4.4: (Color online) (a) Measured fluorescent power enhancement η_f versus the fidelity $|\gamma|^2$ for an $L = 22.8 \pm 0.95 \mu\text{m}$ thick ZnO sample. The symbols are 100 experimental data points obtained at different positions on the sample. The vertical and horizontal error bars are 4% and 7% respectively (Appendix 4.A contains error estimation). The solid line is a linear regression through the data and the dashed curves delimit the 95% confidence interval. The green dotted line: $\eta_f = 1$ (b) Residuals of the experimental data, that are randomly distributed about the linear model.

Fig. 4.4(a) shows the fluorescent power enhancement η_f versus fidelity $|\gamma|^2$ for a sample with thickness $L = 22.8 \pm 0.95 \mu\text{m}$. We see an enhancement in the fluorescent power η_f by about 10% when $|\gamma|^2$ has increased to about 0.035. (For the $23\text{-}\mu\text{m}$ thick sample, the fidelity is limited due to the large number of transmitting channels, of the order of 10^5 , that is much greater than the number of degrees of control on the SLM, of the order of 10^3). This fluorescent enhancement implies that the total energy density for optimized incident wavefronts is higher than for unoptimized incident wavefronts. According to 4.3, η_f is directly proportional to $|\gamma|^2$ with a slope $\alpha = \eta_f^p - 1$ and an intercept of 1. Physically, the intercept means that for an unoptimized wavefront ($|\gamma|^2 = 0$), there is no enhancement ($\eta_f = 1$). Using linear least-squares, we therefore make a linear regression of the data with only the slope as adjustable parameter. For this sample, we obtain a slope $\alpha = 3.6 \pm 0.48$, where the error margin gives the 95% confidence level. The residuals of the data are randomly distributed about the model, see Fig. 4.4(b), with more than 60% of the data within one standard deviation as expected for normally distributed errors [137]. From the slope, we find an ideal enhancement of $\eta_f^p = \alpha + 1 = 4.6 \pm 0.48$.

In Fig. 4.5, we plot results as a function of sample thickness. The fluorescent

power enhancement at perfect fidelity η_f^p increases with sample thickness L , which means that wavefront shaping causes more energy to be stored in the bulk of the medium. The horizontal error bars denote the standard deviation of the measurement of the sample thickness at different positions on the sample, while the vertical error bars denote uncertainty in the determination of η_f^p . For perfect fidelity, the total fluorescent power inside a $22.8\text{-}\mu\text{m}$ thick sample is 4.6 times greater than for non-optimized light.

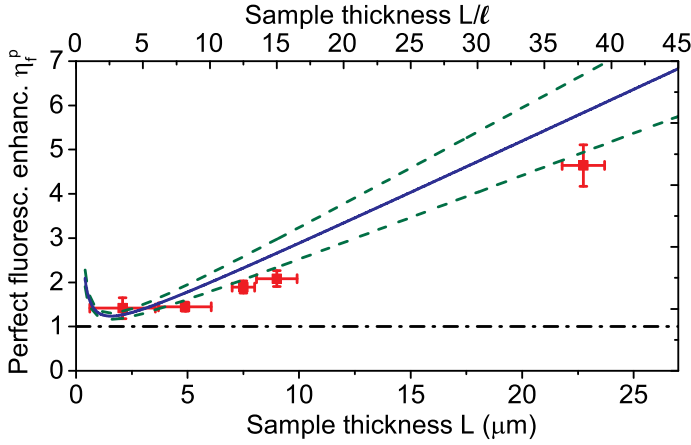


Figure 4.5: (Color online) Fluorescent power enhancement η_f^p in ZnO scattering samples versus sample thickness. The red squares are the measurement-derived fluorescent power enhancement at perfect intensity control. The blue solid line is the calculated fluorescent power enhancement η_f^p from 4.4. The uncertainty in the fluorescent power enhancement $\delta\eta_f^p$ is plotted as the two green dashed curves, which were calculated using equation 4.7. The black dash-dotted curve is for an invariant distribution of energy density along the sample depth. Parameters to obtain the theoretical curves are given in Table 4.1.

4.4 Interpretation and discussion

To interpret our experimental results, we employ diffusion theory (see Chapter 3). For light with an optimized incident wavefront, the distribution of the energy density W_o inside the medium is *a priori* unknown. The energy density within the boundary domain of the sample must be a linear combination of the diffusion eigenfunctions, since the diffusion eigenfunctions form a complete set. In the case of optimized light, the energy density W_o must also be a linear combination of diffusion eigenfunctions. The diffusion eigenfunctions that describe optimized light are expected to have the highest positive contribution to the total transmission, since wavefront shaping is known to increase the total transmission [83, 84]. Here, we model the energy density of optimized light W_o as

the fundamental eigenfunction ($m = 1$) of the diffusion equation for three reasons. Firstly, the contribution to the total transmission* of the first six eigenfunctions in Fig. 4.6 shows that the fundamental eigenfunction ($m = 1$) contributes most to the total transmission. The $m = 1$ contribution is even greater than the total transmission itself, which is a summation of all eigenfunctions. Secondly, the fundamental eigenfunction is the only physical solution with a positive energy density along the sample depth z , as shown in Fig. 4.1b. Thirdly, a symmetric function peaked near the sample center, similar to the fundamental eigenfunction, has been obtained from numerical calculation of wave-front shaped light [71], theoretical and numerical calculations [97, 98], as well as a single-realization experiment [99] of high transmitting channels. Hence we neglect small corrections due to coupling to other modes than the fundamental eigenfunction.

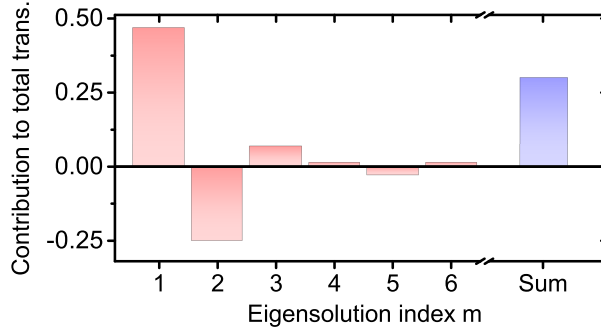


Figure 4.6: (Color online) The contribution to the total transmission of the first six eigenfunctions are represented by the red bars. The total transmission, that is the sum of the transmissions of all individual eigenfunctions, is shown as the blue bar. Parameters: $L = 8.33\ell$, $z_{e1} = 1.96\ell$, $z_{e2} = 0.68\ell$.

The prefactor of the energy density of wavefront-shaped light in our model is determined by the known total transmission of $2/3$ for perfectly wavefront-shaped light [83, 106]. The energy density of optimized light W_o thus obtained is much larger than the diffusive energy density W_d , as shown in Fig. 4.1(a). Since fluorescent light emitted by the nanospheres in our samples propagates diffusively to the detector, we also take the diffusion of fluorescent light into account in the analytical model of the fluorescence enhancement $\eta_f^p(L)$, leading to

$$\eta_f^p(L, \lambda_i, \lambda_f) =$$

$$\frac{A \left[\pi z_{e1}(\lambda_f) \cos\left(\frac{\pi z_{e1}(\lambda_i)}{L_{\text{ex}}}\right) - L_{\text{ex}} \left[\sin\left(\frac{\pi z_{e1}(\lambda_i)}{L_{\text{ex}}}\right) - \sin\left(\frac{\pi L'_i}{L_{\text{ex}}}\right) \right] - \pi L'_f \cos\left(\frac{\pi L'_i}{L_{\text{ex}}}\right) \right]}{\Gamma(\lambda_i, \lambda_f) + z_{\text{inj}}^2 [L'_f + z_{\text{inj}}] e^{-\frac{L}{z_{\text{inj}}}} - z_{\text{inj}}^2 [z_{\text{inj}} + z_{e1}(\lambda_f)]}, \quad (4.4)$$

*The total transmission is the ratio of the total flux at the back surface of the sample ($z = L$) to the incident flux and is equal to $T_{\text{tot}} = -\frac{D}{I_0} \frac{\partial W}{\partial z} \Big|_{z=L}$ [32].

	Incident intensity @ $\lambda_i = 561\text{nm}$	Fluorescent intensity @ $\lambda_f = 612\text{nm}$
ℓ	$0.58^{+0.16}_{-0.10}$	$0.6^{+0.17}_{-0.10}$
z_{e1}	$2.19^{+0.57}_{-0.51}$	$1.80^{+0.60}_{-0.50}$
z_{e2}	$0.68^{+0.10}_{-0.02}$	$0.69^{+0.02}_{-0.03}$
n_{eff}	$1.45^{+0.12}_{-0.12}$	$1.36^{+0.14}_{-0.13}$

Table 4.1: Values of the transport mean free path ℓ , extrapolation lengths z_{e1} and z_{e2} for wavelengths of the incident and fluorescent intensities. The transport mean free path was determined from total transmission measurement of similar samples (see Ref. [138]). Following [16], we derived the extrapolation lengths from the average reflectivities at the interfaces for a scattering medium with an effective refractive index n_{eff} . The effective refractive index n_{eff} was determined from measurement of the angle-resolved escape function on similar samples [138].

where

$$A = \frac{2L_{\text{ex}}^2 \sec(\frac{\pi z_{e2}(\lambda_i)}{L_{\text{ex}}})}{3\pi^3}, \quad (4.5)$$

$$\Gamma(\lambda_i, \lambda_f) = \frac{L[z_{\text{inj}} + z_{e1}(\lambda_i)]}{6L_{\text{ex}}} [L^2 + 3L(z_{e1}(\lambda_f) + z_{e2}(\lambda_i)) + 6z_{e1}(\lambda_f)z_{e2}(\lambda_i)], \quad (4.6)$$

and $L'_i = L + z_{e1}(\lambda_i)$, $L'_f = L + z_{e1}(\lambda_f)$, $L_{\text{ex}} = L + z_{e1}(\lambda_i) + z_{e2}(\lambda_i)$, z_{inj} is the injection depth at which the incident light becomes diffuse, which accounts for the angular distribution of the incident shaped wavefront [114]. With an incident numerical aperture of 0.9, we find $z_{\text{inj}} = 0.86\ell(\lambda_i)$. The fluorescent power enhancement η_f^{p} is determined by factors such as transport mean free path ℓ and the extrapolation lengths z_{e1} and z_{e2} , which are determined experimentally (see Table 4.1). It therefore follows that the evaluation of η_f^{p} also has an uncertainty $\delta\eta_f^{\text{p}}$. We calculate $\delta\eta_f^{\text{p}}$ using

$$\delta\eta_f^{\text{p}}(L, \lambda_i, \lambda_f) = \frac{\partial\eta_f^{\text{p}}}{\partial\ell}\Delta\ell(\lambda_i, \lambda_f) + \frac{\partial\eta_f^{\text{p}}}{\partial z_{e1}}\Delta z_{e1}(\lambda_i, \lambda_f) + \frac{\partial\eta_f^{\text{p}}}{\partial z_{e2}}\Delta z_{e2}(\lambda_i, \lambda_f), \quad (4.7)$$

where ∂ signifies a partial derivative, $\Delta\ell(\lambda_i, \lambda_f)$ and $\Delta z_{e1,2}(\lambda_i, \lambda_f)$ are the errors in the determination of the transport mean free path and the extrapolation lengths respectively.

To compare our model to our experimental results, we plot in Fig. 4.5 the analytic model for η_f^{p} versus sample thickness L . Within the uncertainty of the samples' parameters, our model agrees remarkably well with our experimental results, especially in view of the fact that the model has no adjustable parameters. The excellent agreement between our model and our experimental results confirms that energy has been mostly coupled into the fundamental ($m = 1$) diffusion mode.

In our experiments we obtain the fluorescent power enhancement η_f^p rather than the energy density enhancement η_{ed} that is defined as $\eta_{ed} \equiv \int dz W_o / \int dz W_d$. We derive

$$\eta_{ed}(L) = \frac{2}{3} \eta_f(L) + O(l/L), \quad (4.8)$$

where $O(l/L)$ represents higher orders, see Chapter 3. The relation in 4.8 shows that the total fluorescent power depends on the total energy density inside the medium. Hence, the observed increase of the fluorescence is indeed a probe of the increased energy density.

4.5 Summary

We have measured the increase of the total energy density inside a scattering medium that is the result of a shaped incident wavefront. The results agree quantitatively with a model that considers energy to be dominantly coupled to the fundamental mode of the diffusion equation. Our results are of broad relevance since they apply to other wave control methods in scattering media, such as time reversal, phase conjugation, and transmission matrix-based control, as well as to other types of waves such as microwaves, acoustic waves, elastic waves, surface waves, and electron waves. We expect our results to be relevant for applications that require enhanced total optical energy density, such as efficient light harvesting in solar cells especially in near infrared where silicon has low absorption; for enhanced energy conversion in white LEDs, to reduce the quantity of expensive phosphor; for low threshold and higher output of random lasers; and in homogeneous excitation of probes in biological tissues. Last but not least, it will be fruitful to investigate possible relationships between the fundamental diffusion eigenfunction and the universal diffusion time as obtained in [123, 139, 140].

4.A Estimation of experimental errors

4.A.1 Error in the estimation of the fluorescent power enhancement

The fluorescent power enhancement is defined as $\eta_f \equiv P_f^o / P_f^n$, where P_f^o is the total fluorescent power with an optimized phase pattern on the SLM, and P_f^n is the total fluorescent power for a reference incident wavefront. Therefore, the error in the fluorescent power enhancement $\sigma(\eta_f)$ equals

$$\sigma(\eta_f) = \sqrt{\sigma^2(P_f^o) + \sigma^2(P_f^n)}, \quad (4.9)$$

where $\sigma(P_f^o)$ and $\sigma(P_f^n)$ are the independent errors in determining the fluorescent intensity of optimized and non-optimized light respectively. Contributions to $\sigma(P_f^o)$ are from the laser and the camera noise σ_{lc} , and independently from the mesoscopic intensity fluctuations that illuminate the randomly positioned spheres

σ_m ,

$$\sigma(P_f^o) = \sqrt{\sigma_{lc}^2 + \sigma_m^2}. \quad (4.10)$$

In order to determine σ_{lc} , we measured the total fluorescent power emanating from the sample for 12 hours. We determine the laser and camera noise σ_{lc} from the standard deviation of the fluorescent power with respect to the mean power and we obtain $\sigma_{lc} = 0.03$.

The mesoscopic intensity fluctuation σ_m is given by the fluctuation of the overlap integral between the illuminating speckle intensity and the spatial distribution of the fluorescent spheres in the scattering medium. We estimated σ_m using a simple model. For a given number of spheres N_{sphere} we generated a random array with N_{sphere} non-zero values which represent our spheres embedded inside the scattering medium. To simulate the speckle intensity inside the scattering medium, we generated a second random array consisting of numbers with a Rayleigh distribution. We multiply the two random arrays and take the summation of the resulting array, which gives the simulated total fluorescence power P_f^s . We repeated the procedure to simulate 100 realizations and calculated the standard deviation of the normalized total fluorescence power $P_f^s / \langle P_f^s \rangle$, which corresponds to σ_m for a given number of spheres in a scattering medium. Here $\langle \rangle$ represents ensemble average over several realizations of the simulation. There are $N_{\text{sphere}} = 1.6 \times 10^4$ inside the 23 μm -thick sample and we obtain $\sigma_m = 0.005$ from the simulation. We then find $\sigma(P_f^o) = (0.03^2 + 0.005^2)^{\frac{1}{2}} = 0.03$. We assume that $\sigma(P_f^o) = \sigma(P_f^n)$. Therefore, we obtain $\sigma(\eta_f) = (0.03^2 + 0.03^2)^{\frac{1}{2}} = 0.04$, as mentioned in the main text.

4.A.2 Error in the estimation of the fidelity $|\gamma|^2$

In the main manuscript, we determine the fidelity $|\gamma|^2 = \frac{P_o}{\langle P_n \rangle}$, where P_o is the power in the optimized focus and $\langle P_n \rangle$ is the ensemble-averaged power transmitted before optimization. Therefore, the error in estimating the fidelity $\sigma(|\gamma|^2)$ is

$$\sigma(|\gamma|^2) = \sqrt{\sigma(P_o)^2 + \sigma(\langle P_n \rangle)^2}, \quad (4.11)$$

where $\sigma(P_o)$ and $\sigma(\langle P_n \rangle)$ are the independent errors in estimating P_o and $\sigma(\langle P_n \rangle)$ respectively. We determine $\sigma(P_o)$ from the laser and the camera noise, and then have $\sigma(P_o) = 0.03$. We write $\sigma(\langle P_n \rangle)^2$ as

$$\sigma(\langle P_n \rangle)^2 = \sqrt{\sigma_{lc}^2 + \sigma_{C2}^2 + \sigma_s^2 + \sigma_c^2}, \quad (4.12)$$

where σ_{C2} is due to expected fluctuation in the total transmitted intensity, *i.e.* the C_2 intensity correlation [18], σ_s is due to the roughness of the sample, and σ_c is the fraction of intensity cropped out by the optics. We estimated σ_{C2} as

$$\sigma_{C2} = \frac{1}{2NT_{\text{tot}}}, \quad (4.13)$$

where N is the number of transmitting channels and T_{tot} is the total transmission [114]. From the size of the diffuse spot in the sample, we estimate $N \approx 5.1 \times 10^4$ and the total transmission is estimate as $T_{\text{tot}} \approx \ell/L = 0.03$. Therefore, $\sigma_{C2} = 0.018$. To estimate σ_s , we measured the sample thickness at several positions on the sample and we obtain a fluctuation of $\sigma_s = 0.04$. From the size of the diffuse spot at the back of the sample, we estimate $\sigma_c \approx 0.02$. We find $\sigma(\langle P_n \rangle) = 0.06$ and $\sigma(|\gamma|^2) = 0.07$.

We use the estimated errors: $\sigma(|\gamma|^2) = 0.07$ and $\sigma(\eta_f) = 0.04$ as the horizontal and vertical error bars respectively in Figs 3(a) and (b) in the main manuscript. From the plot of the residuals in Fig. 3(b), about 60% of the data points are within one standard deviation from the model, which means is expected since the data points are Gaussian distributed.

CHAPTER 5

Probing the spatial energy density inside a three dimensional scattering medium

5.1 Introduction

A conventional optical microscope can image intensity in a homogeneous medium with a resolution that is diffraction-limited. Using a microscope, it is impossible, however to produce a diffraction-limited image of the average light distribution inside a scattering medium, which has inhomogeneous spatial variations of the refractive index. The microscope ‘sees’ a diffuse intensity blob due to multiple scattering of light by the inhomogeneities in the medium [18, 32]. This is the reason why scattering media, which are part of our everyday life are necessarily opaque. Examples of a scattering medium are biological tissue, fog, cloud, teeth, paper, paint, and diffuser glass. It is of a great interest to find a suitable method to image the light distribution inside a scattering medium, since they are widely studied for both fundamental research and applications.

The energy density inside a two dimensional (2D) scattering medium has been studied using methods such as out-of-plane measurement of scattered fields [129] and near-field measurements [141]. These methods are limited to 2D samples because the imaging is through the third dimension, which is typically an accessible homogeneous medium but this is not available in a three dimensional (3D) sample. It is therefore especially challenging to image inside a 3D scattering medium. Of particular relevance is the light distribution of shaped waves, which focus the intensity transmitted through a scattering medium [49, 86]. Recently, our team reported the first measurement of the total energy density inside a 3D scattering medium for shaped incident waves (see Chapter 4). We probed the *total* stored energy using fluorescent particles. However, spatially resolving the energy density distribution of shaped light, which has been predicted in Refs. [71, 100, 142] remains an open challenge.

In this chapter, we present an experimental study to spatially resolve the depth-dependent energy density inside a 3D scattering medium for shaped incident wavefronts. We probe the energy density using a similar fluorescence microscopy method used in Ref. [100] and Chapter 4. We measured the intensity emitted by isolated fluorescent probes particles that are located at various depths inside the sample (see Fig. 5.1). We observe that the energy density is higher for particles

The experiments in this chapter were done in close collaboration with dr. Peilong Hong (COPS, FOM Stirring of light!).

at depths close to the back surface of the sample. We interpret the results with a theory describing the 3D energy density of optimized light in Chapter 3.

5.2 Experimental details

5.2.1 Sample, preparation, and experimental setup

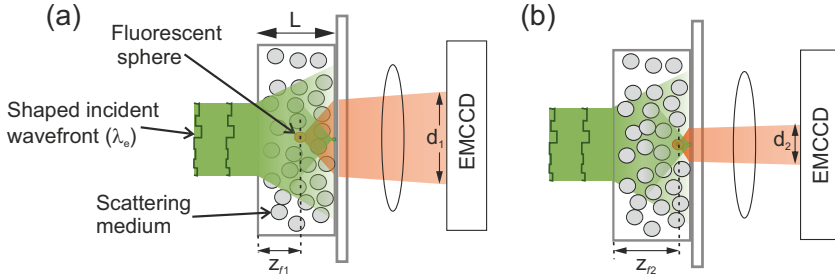


Figure 5.1: Set-up to probe the depth-dependent energy density. Two fluorescent particles are located at different depths z_{01} and z_{02} in (a) and (b) inside a scattering medium, respectively. The fluorescent particles are excited by the incident pump intensity at wavelength λ_e . The emission at wavelength λ_f from the particles probes the energy density at the position z_f . The emitted fluorescent intensity is imaged onto the chip of an electron multiplying charged-coupled device (EMCCD).

As a scattering medium, the sample is a slab of zinc oxide similar to the ones used in Chapter 4. The main difference is that here, there is only a single fluorescent particle in the illuminated volume and the particle probes energy density at a specific depth (see Fig. 5.1). In addition, to achieve sufficient fluorescent signal from the single particle, we use fluorescent particles with diameter $d = 300$ nm rather than $d = 50$ nm, which were used to measure the total energy density in Chapter 4. The sample preparation method by spraying painting is described in Section 4.2.1. The concentration of the fluorescent particles in the suspension is $0.34 \mu\text{m}^{-6}$, which is $1000\times$ lower than the samples studied in Chapter 4. With the low concentration of the probe particles, there is a higher probability of finding isolated single probe particles in the illuminated volume.

The experimental set-up is exactly the same as in Chapter 4. To detect the low fluorescent intensity from a single sphere, the EMCCD camera was operated in the EM gain mode with a gain of 300 and an integration time of 5 seconds. The chip of the EMCCD was cooled to -70°C using a Peltier element, in order to reduce noise due to thermally-generated photoelectrons.

5.2.2 Phase perturbation to control fidelity $|\gamma|^2$

Since we want to measure the energy density from a single particle at a fixed position, a new method is required to control the fidelity of the optimization at the sphere position. In order to achieve this control, we first obtain an optimized pattern from a wavefront shaping experiment. Then, we systematically perturbed the optimized pattern by adding random phase noise to it. The perturbed optimized pattern $\phi_{i,j}^p$ is expressed as

$$\phi_{i,j}^p = \phi_{i,j}^o + \delta\phi_{i,j} \quad (5.1)$$

where (i, j) are the indices of the segments on the spatial light modulator (SLM), $\phi_{i,j}^o$ is the optimized phase pattern, and $\delta\phi_{i,j}$ is the phase noise, which takes random values between zero and a maximum value $\delta\phi_m$. We varied $\delta\phi_m$ from 0 to 2π in 52 steps. For each phase perturbation, we measured the fidelity. We expect that the phase noise gradually reduces the fidelity and at $\delta\phi_m = 2\pi$, the fidelity has completely vanished.

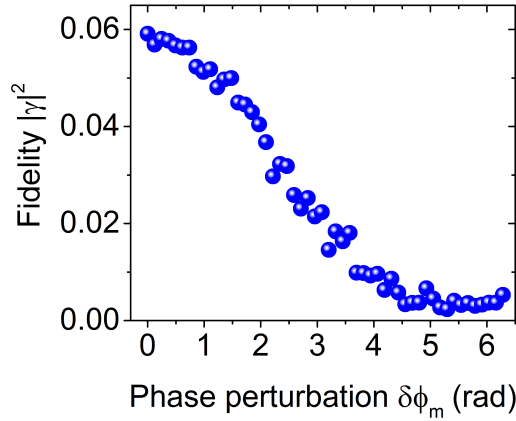


Figure 5.2: Measured fidelity $|\gamma|^2$ versus maximum phase perturbation $\delta\phi_m$ of the optimized phase pattern.

In Fig. 5.2, we plot the measured fidelity $|\gamma|^2$ versus the maximum phase perturbation $\delta\phi_m$ of the optimized wavefront. The fidelity is maximal ($|\gamma|^2 = 0.06$) at $\delta\phi_m = 0$ and vanishes gradually as $\delta\phi_m$ increases. The fidelity decreases because the incident waves that are excited by the perturbed wavefront no longer interfere at the target spot but rather contribute to the background speckle. Using the experimental data, we can control the fidelity at a fixed position of a fluorescent sphere.

5.2.3 Measurement procedure

Below are the procedures to measure the depth-dependent energy density inside a scattering medium.

1. We scanned the sample in the transversal (x, y) plane and measured the fluorescent intensity exiting the back surface of the sample. The purpose of the scan is to find the (x, y) positions of an isolated fluorescent particle that is illuminated by the incident light. A typical signature of a fluorescent particle is a diffuse blob of intensity and an example is shown in Fig. 5.3(a).
2. Before performing measurements on a selected fluorescent particle, the position of the sample was adjusted such that the fluorescent particle is positioned at the center of the transmitted speckle pattern. This ensures that each fluorescent particle probes the same position on the spatial profile of the diffuse light inside the sample. The position (x_f, y_f) centered on the speckle pattern of each available fluorescent particle was then saved.
3. The sample was translated to a saved position (x_f, y_f) where we performed a wavefront shaping experiment to focus light to a speckle spot. We used the piecewise sequential algorithm for the optimization and the number of segments on the spatial light modulator (SLM) is 900.
4. After the wavefront shaping experiment, we determined the fidelity $|\gamma|^2$ and the enhancement η_f of the fluorescent intensity from the particle. The fluorescent enhancement η_f is defined in Section 4.2.3.
5. We measured the dependence of the fluorescent enhancement η_f on the fidelity $|\gamma|^2$. We controlled the fidelity using the phase perturbation method described in Section 5.2.2. During each phase perturbation step, we measured both $|\gamma|^2$ and the corresponding η_f . The unoptimize phase pattern that we used to determine η_f is a phase pattern optimized for an uncorrelated position on the sample. This phase pattern is also perturbed during the phase perturbation procedure to have sufficient statistical averaging.
6. We repeated the procedures 3 to 5 for 100 times at all saved positions (x_f, y_f) of the fluorescent particles.

5.3 Estimating the depth of a fluorescent sphere inside a scattering medium

In this section, we describe a method to estimate the depth of the fluorescent particles located deep inside a scattering medium. During the sample preparation, the particles end up at random depths inside the scattering medium, and the depths are unknown *a priori*. We estimate the depth of the particles using a method that is similar to one use previously in Refs. [20, 143, 144]. The method compares the size of the diffuse fluorescent intensity at the back surface of the sample with the solution of the three dimensional (3D) diffusion equation for a point emitter inside a scattering medium. The depth of the particle and an intensity prefactor are the only adjustable parameter in the model. The accuracy of the method is limited to about one transport mean free path [143, 145].

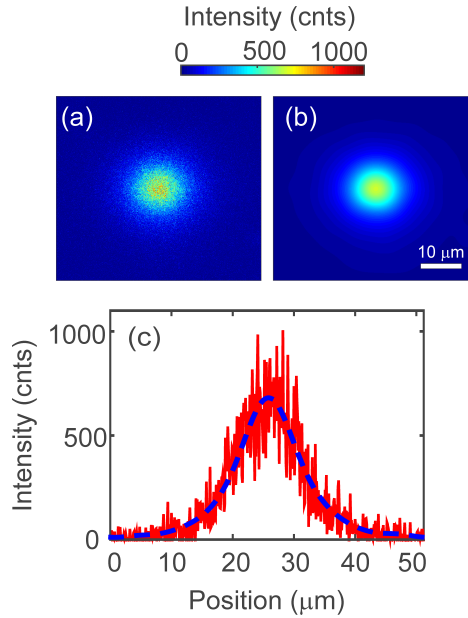


Figure 5.3: Fluorescent intensity image of a single fluorescent particle before (a) and after (b) Fourier filtering. Fourier filtering is used to remove high frequency intensity fluctuation. (c) Cross section through the center of the intensity image before (red solid curve) and after (blue dashed line) filtering versus position.

We show in Fig. 5.3(a) an image of the fluorescent intensity from one sphere located inside the scattering medium. The image appears “grainy” and noisy, which is more apparent in a cross section through the center of the image (see Fig. 5.3(c)). The noise shows up as high spatial frequencies and is attributed to insufficient ensemble averaging, shot noise, and camera noise. We attenuate the noise in the image by performing Fourier filtering, which reduces the amplitude of the high spatial frequencies. The Fourier-filtered image and a cross section through its center are shown in Figs. 5.3(b) and (c) respectively. The Fourier filtering significantly reduces the spatial frequencies in the image and we then determine the size of the Fourier-filtered image.

To quantify the size of the fluorescent intensity, we use the full width at half maximum (FWHM) w_f . The Fourier-filtered image in Fig. 5.3(b) has width that is determined to be $w_f = 6.2 \pm 0.2 \mu\text{m}$, where the error is from the camera and laser noise. Another measure of the size of the fluorescent intensity is the second moment, however, it is largely influenced by the background level of the intensity. For a background increase of 20%, the second moment of an image drastically increases by approximately 80% compared to an increase of approximately 10% for the full width at half maximum. Therefore, the full width at half maximum is a more robust measure of the size of the fluorescent intensity and has an error

depending on the peak intensity to background ratio (2% in our case), which is negligible. We now proceed to estimate the depth of the fluorescent particle from diffusion theory.

The energy density from a point emitter at a depth z_f is described by the diffusion equation [18, 32]. The solution of the 3D diffusion equation in the Fourier transformed transversal coordinates $(q_{\perp x}, q_{\perp y})$ is

$$W_d(q_{\perp}; z, z_f) = \frac{P_{\text{in}}}{D} \frac{\sinh(q_{\perp} [L_{\text{ex}} - z - z_{e1}]) \sinh(q_{\perp} [z_f + z_{e1}])}{q_{\perp} \sinh(q_{\perp} L_{\text{ex}})} \quad (5.2)$$

where W_d is the energy density at a specific depth z , $q_{\perp} \equiv (q_{\perp x}^2 + q_{\perp y}^2)^{\frac{1}{2}}$, P_{in} is the incident power, D is the diffusion constant, and $L_{\text{ex}} = L + z_{e1} + z_{e2}$ [144]. The solution in Eq. 5.2 only holds for a condition $z > z_f$ (see the full solution in Ref. [144]). We obtain the flux $F_d(q_{\perp}, z)$ exiting the back surface of the sample at $(z = L)$ by taking a derivative of W_d with respect to z to get

$$F_d(q_{\perp}; z = L, z_f) = C \frac{\cosh(q_{\perp} [L_{\text{ex}} - L - z_{e1}]) \sinh(q_{\perp} [z_f + z_{e1}])}{\sinh(q_{\perp} L_{\text{ex}})} \quad (5.3)$$

where C is a constant prefactor.

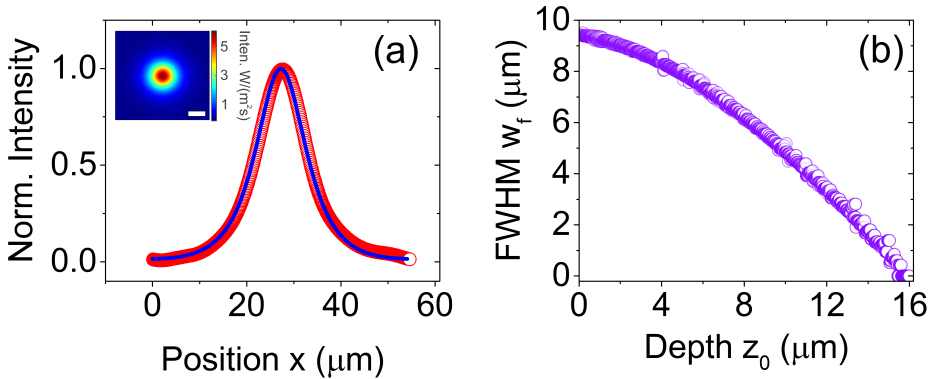


Figure 5.4: (a) Fluorescent intensity normalized to its maximum versus position. The blue curve is a cross section through the center of the image (see inset), which is a Fourier transformation of the diffusion equation solution in Eq. 5.3. The solution applies to a point emitter inside a scattering medium and in this case, the depth of the emitter is $z_f = 8.0 \pm 0.6 \mu\text{m}$. The full-width at half maximum of the image in the inset is determined to be $w_f = 6.2 \pm 0.2 \mu\text{m}$. The red circles are the Fourier-filtered fluorescent intensity experimental data, already shown in Fig. 5.3(c). The scale bar is $10 \mu\text{m}$. (b) The full-width at half maximum w_f of the diffusion equation versus depth z_f . The sample thickness L in both (a) and (b) is $L = 14.0 \pm 2 \mu\text{m}$.

We performed a numerical Fourier transformation of $F_d(q_{\perp}, z_f)$ to obtain the

flux $F_d(x, y)$ in real space (x, y) that can be compared to experimental data. In the inset of Fig. 5.4, we show the result of the real space intensity flux of the numerical Fourier transformation. The particle is at a depth $z_f = 8.0 \pm 0.6 \mu\text{m}$. The curve is a symmetric function, which is similar to the experimental data shown in Fig. 5.3(a) and the two results are compared in Fig. 5.4. There is an excellent match between the solution of the diffusion equation and the Fourier-filtered fluorescent intensity. Furthermore, we determined $F_d(x, y)$ for 2000 points between $z_f = 0$ to $z_f = L = 16 \mu\text{m}$, which is the upper bound of the measured thickness of the sample. For each z_f , we determined w_f of $F_d(x, y)$ and we plot w_f versus z_f in Fig. 5.4(b). The full width at half maximum w_f has the largest values when the particle is close to the front surface of the sample, *i.e.*, $z_f = 0$, and w_f steadily reduces to zero when $z_f = L$. This means that a particle close to the front surface has larger spread of the fluorescent intensity than a particle close to the back surface. The reason for this behaviour is because fluorescent light from a particle closer to the front surface experiences more scattering events and therefore diffuses to a larger space in the plane (x, y) . We created a look-up table using Fig. 5.4(b) and then determine the depth of the fluorescent particles in our experiment from the full width at half maximum of the measured intensity image. For the fluorescent particle in Fig. 5.3(a), the depth is determined to be $8.0 \pm 0.8 \mu\text{m}^*$ using $w_f = 6.2 \pm 0.2 \mu\text{m}$ from the image of the fluorescent intensity.

5.4 Results and discussion

5.4.1 Controlling the fidelity $|\gamma|^2$

To start with, we test the new method to control fidelity at a fixed position. We performed experiments to measure the total energy density inside a scattering medium and compare with the result obtained in Chapter 4. The sample is one of the samples used in Chapter 4 that has a thickness $L = 9.0 \pm 1 \mu\text{m}$. The sample has fluorescent particles randomly distributed throughout the sample volume and they probe the total energy density. In Fig. 5.5, we plot the measured fluorescence enhancement η_f versus fidelity $|\gamma|^2$ and η_f increases as $|\gamma|^2$ increases. Using linear least-squares, we make a linear regression of the data following Eq. 4.3. From the fit, we found a slope $\alpha = 1.12 \pm 0.06$ that agrees amazingly well with the slope $\alpha = 1.08 \pm 0.08$, which was obtained by the “free-style” method of varying the fidelity that was used in Chapter 4.

5.4.2 Depth-dependent energy density

In Fig. 5.6(a), we show the measured fluorescence enhancement η_f versus fidelity $|\gamma|^2$ for a particle that has at a depth $z_f = 10.4 \pm 0.6 \mu\text{m}$. There is an increase of

*The uncertainty of $0.8 \mu\text{m}$ is from w_f and the accuracy of the depth determination method. An addition error to determine z_f arises from the uncertainty of the sample thickness and the error on z_f is asymmetric inclining towards the negative side. The asymmetry is because we have used the maximum measured L to determine z_f . Therefore, $0.8 \mu\text{m}$ is an underestimation of the actual error on z_f , which needs to be calculated.

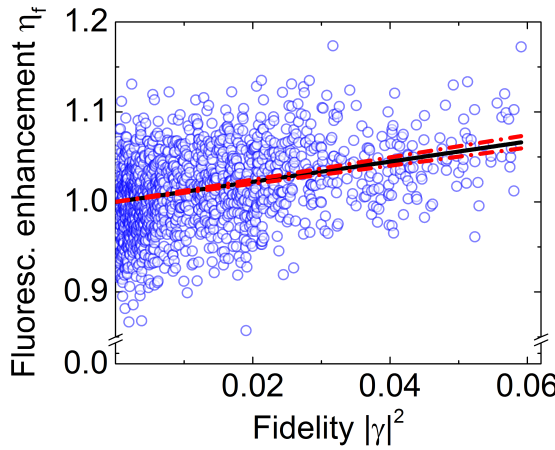


Figure 5.5: Measured fluorescent power enhancement η_f versus the fidelity $|\gamma|^2$ for an $L = 9.0 \pm 1\mu\text{m}$ thick ZnO sample, with randomly positioned fluorescent spheres. The blue circles are 1560 experimental data points taken at 10 different positions on the sample, 3 measurements per positions, and 52 phase perturbations per measurement. The black solid line is a linear regression through the data and the red dashed curves delimit the 95% confidence interval.

η_f as $|\gamma|^2$ increases because the internal energy density approaches the optimized light as the fidelity increases. This observation is similar to what we have observed for the total energy density. The fluctuation in the data is attributed to factors such as insufficient ensemble averaging by a single particle, laser noise, and camera noise as we have discussed in Appendix 4A. Using a similar procedure as in Chapter 4, we compared the experimental data to Eq. 4.8, which is a linear curve passing through an intercept at $\eta_f = 1$. We determined the slope α of the linear curve to be $\alpha = 11.3 \pm 0.8$, where the error margin gives the 95% confidence level. From the slope, we derive the fluorescent enhancement at perfect fidelity $\eta_f^p = 12.3 \pm 0.8$.

We determined η_f^p for 8 other fluorescent particles located at different depths inside the sample and plot η_f^p versus depth z_f in Fig. 5.6(b). Amongst the fluorescent particles detected, the closest to the front surface is located at a depth $z_f = 2.7\mu\text{m}$. There is a low probability of detecting particles at lower depth because the measured fluorescent intensity from these particles are very low due to scattering. At the lowest measured depth $z_f = 2.7\mu\text{m}$, $\eta_f^p = 3.6$ and steadily increase as z_f increases. At depths closer to the back surface, η_f^p interestingly increases drastically and has a maximum value $\eta_f^p = 25.0$ for a particle at a depth $z_f = 12\mu\text{m}$. This is the first measurement of the depth-dependent fluorescent enhancement, which corresponds to the internal energy density inside a 3D sample.

To understand the experimental data, we compared in Fig. 5.6 a theory that

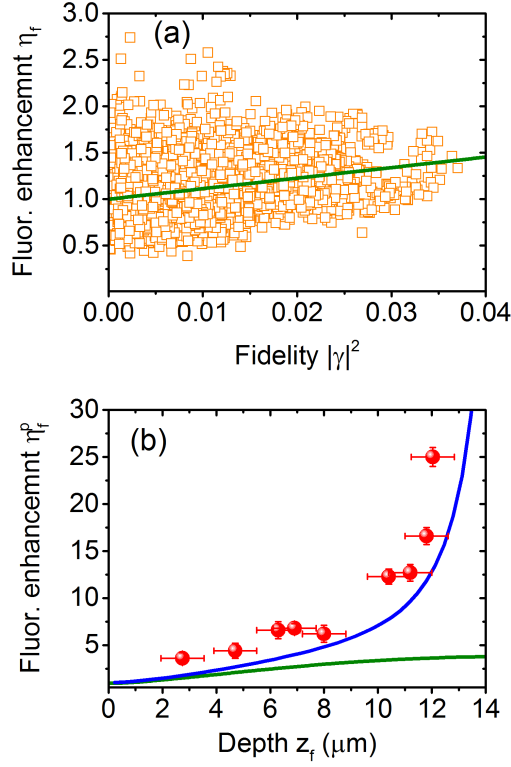


Figure 5.6: (a) Measured fluorescent enhancement η_f versus the fidelity $|\gamma|^2$ for a particle located at depth $z_f = 10.38 \pm 0.8 \mu\text{m}$ inside a $L = 14.0 \pm 2 \mu\text{m}$ thick ZnO sample. The symbols are 100 wavefront shaping experiments and 41 phase perturbations for each experiment. The solid line is a linear regression through the data and 95% confidence interval is comparable to the thickness of the solid line. (b) Fluorescent enhancement η_f^p at fidelity $|\gamma|^2 = 1$ versus depth z_f . The symbols are the experimental data extracted from the dependence of η_f on $|\gamma|^2$, which is shown in (a) for one particle. The vertical error bars denote uncertainty in the determination of η_f^p , while the horizontal error bar is one transport mean free path ℓ , which is the accuracy of the method to determine the depth. The blue and green curves are the theoretical fluorescence enhancement as a function of depth calculated using the 3D and 1D energy densities respectively.

is based on the 3D energy density inside a scattering medium (see Chapter 3). Without any adjustable parameter, there is a good agreement between the 3D-based theory and the experimental data. This agreement confirms that the depth-dependent energy density can be model using only the fundamental diffusion eigenfunction as we have considered in Chapter 3. A 1D-based theory deviates obviously from the experimental data and this implies that a 1D-based theory is insufficient to describe our data. In Chapter 4, a 1D-based theory describes the

total energy density inside a scattering medium and this is because the probe fluorescent particles are distributed throughout the sample and therefore, there is translational invariance in the (x, y) plane.

5.5 Summary and outlook

We have presented experimental details on the measurement of the depth-dependent energy density inside a 3D scattering medium for incident shaped wavefront. We probe the energy density using a isolated fluorescent particles located at various depths. the depth of the particles was determined by comparing the prediction of diffusion theory. with the full width at half maximum of the transmitted fluorescent intensity. We observe that the energy density is higher for particles at depths close to the back surface of the sample. A theory that describes the 3D energy density agrees well with the experimental data without adjustable parameters.

CHAPTER 6

Interplay between Bloch and scattered waves in a real photonic crystal

6.1 Introduction

Photonic crystals are composite materials that greatly influence the propagation and emission of light. Photonic crystals are periodic arrangements of dielectric materials with a period in the order of the wavelength of light [24]. In an infinite and perfect photonic crystal, any allowed wave can be decomposed in a basis of propagating Bloch waves, due to the translational symmetry of the photonic crystal [75, 146]. Bloch waves are plane waves modulated by a spatially periodic function. The spatial structure of the waves inside an infinite and perfect photonic crystal is therefore well-defined.

The structure of a real photonic crystal deviates significantly from that of an infinite and perfect crystal in two major ways. Firstly, a real photonic crystal is necessarily finite in extent [147, 148]. Secondly, imperfections in the fabrication processes are inevitable, due to limitations in the fabrication processes of any crystal [74, 149–153]. Such imperfections include size and position variations of the constituents, impurities with a different dielectric constant, and crystal mosaic spread that occurs in artificial opals grown using self-assembly [154, 155]. These imperfections result in deviations from the periodic nature of the photonic crystal, thereby introducing disorder. Disorder influences the spatial structure of waves that propagate inside the crystal, since the disorder breaks the discrete translational symmetry of the crystal [76]. Therefore, inside a photonic crystal with disorder, the structure of waves is different from Bloch waves. It is relevant to study the complex nature of waves in real photonic crystals, which are suitable media to study fundamental concepts such as Anderson localization [22] and control of spontaneous emission [26]. Moreover, for the purpose of application, an understanding of the nature of the waves is crucial since disorder adversely affects the applications of photonic crystal devices such as high-efficiency light emitting diodes [27], low threshold lasers [25], ultra-low waveguides, and disrupts the functionality of photonic integrated circuits [151, 156].

It is a challenge to experimentally study the structure of waves inside a three-dimensional (3D) photonic crystal. Attempts have been made using methods such as near-field measurements [157–159] and emission spectra from infiltrated fluorophores [160]. These methods extract the optical properties inside a 3D photonic crystal by using probes such as a glass tip that perturbs the

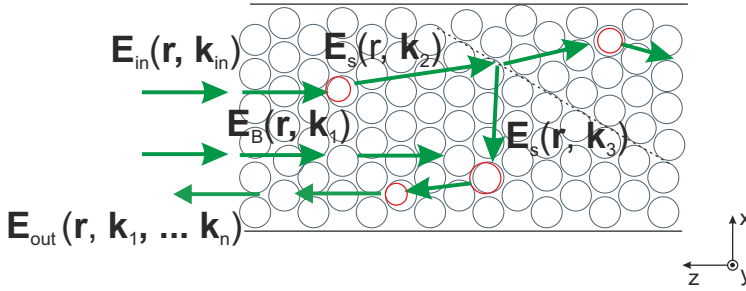


Figure 6.1: Schematic representation of light propagation inside a real photonic crystal. The red circles show the size and position variations, which are deviations from a perfect crystal. The black dashed line represents a crystal grain boundary. The incident light E_{in} is coupled to Bloch waves E_B typical of propagation in a perfect crystal. The imperfections break the translational symmetry of the structure and therefore the Bloch waves are scattered to generate E_s at different wavevectors. The total reflected intensity E_{out} therefore is a contribution from different fields with different wavevectors.

detected field or infiltrated fluorophore that probes the local field. Reflection and transmission microscopy analyze the flux exiting the interfaces of real photonic crystal, without probing the internal field. Using these methods, experimental investigations of the influence of disorder on light propagation have been carried out on real photonic crystals [161–166]. From these previous studies, no information has been extracted on the structure of the waves inside a real photonic crystal.

In this chapter, we introduce a method to study how waves are affected by disorder inside a real photonic crystal. We study positional correlations of the reflected wave from opal and two-dimensional (2D) Si photonic crystals. As depicted in Fig. 6.1, we assume that light exiting a real photonic crystal is a superposition of Bloch waves E_B and scattered waves E_s , which are the result of scattering from disorder such as grain boundaries, position and size variations, and a different dielectric material. The Bloch waves E_B are expected to reveal periodic spatial correlations due to the long-range periodic order of the photonic crystal, while the scattered waves E_s are expected to be uncorrelated due to multiple scattering [18]. We therefore investigate the correlation in the reflected intensities by comparing the intensity distribution of the reflected light with the Rayleigh distribution, which is expected for uncorrelated intensities [167]. We observe that an opal with a large crystal domain shows a high correlation compared to an opal with small crystal domains and more imperfections. In addition, using spatial intensity correlation, we investigate the similarity between light reflected from different positions. From the spatial correlation, we observe regions of high and low correlations, which are as a result of different contributions of Bloch and scattered waves. Structural features such as the periodicity of the crystal and the grain boundaries are revealed by the spatial correlation and are in

agreement with structural characterization using scanning electron microscope. We derive a simple and phenomenological analytical model to describe the intensity correlations from a photonic crystal. The only adjustable parameter in our model is the ratio β of the amplitude of the scattered waves to the Bloch waves. With $|\beta|^2 = 0.36$, the model agrees well with our experimental result. Our results remarkably reveal that the reflected intensity from an opal photonic crystal has a comparable contribution from both Bloch and scattered waves.

6.2 Experimental setup and sample

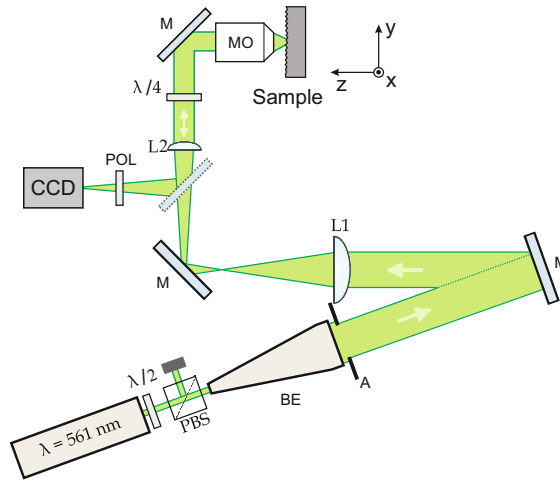


Figure 6.2: (Color online) **Experimental setup:** The light source is a continuous wave laser, which emits at 561 nm. The laser beam passes through a half wave plate ($\lambda/2$) and a polarizing beam splitter (*PBS*) to obtain a linearly polarized light. The laser beam is expanded using the beam expander ($\times 20$) and aperture *A* controls the beam diameter to be 8 mm. A combination of lenses *L1* ($f = 100$ mm) and *L2* ($f = 100$ mm) demagnifies the beam two times in order to completely fill the back aperture of the microscope objective (*MO*). The microscope objective focuses light onto the sample surface. The sample is mounted on a three-axis piezoelectric translational stage. With a combination of the microscope objective and lens *L2*, the reflected light exiting the front surface of the sample is imaged onto the chip of a CCD camera. A polarizer (*POL*) and a quarter wave plate ($\lambda/4$) are placed in the reflection path to filter the direct reflection from the sample surface. *M* are dielectric mirror.

A schematic of the experimental setup is shown in Fig. 6.2. The setup is similar to the one used in Chapter 4, up until the sample stage. The light source is a continuous wave laser, which emits at a wavelength $\lambda = 561$ nm. The laser beam passes through a polarizing beam splitter (*PBS*) and a half wave plate ($\lambda/2$) to obtain linearly polarized light. The beam is expanded and demagnified

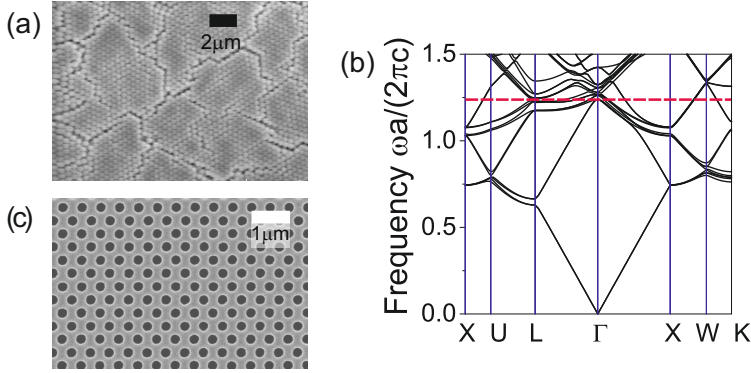


Figure 6.3: (a) Scanning electron micrograph of the fcc (111) top surface of a 3D opal crystal of silicon dioxide spheres with radius $R = 349$ nm grown on a silicon wafer. The opal crystal consists of a lot of small crystal domains separated by grain boundaries. (b) Band structure (black lines) of a silica opal calculated by using the plane-wave expansion method. The horizontal red dashed line indicates the frequency of the illuminating monochromatic light. (c) Scanning electron micrograph of a top-down fabricated 2D silicon photonic crystal with centered rectangular lattice with pore radius of $R = 153$ nm.

to completely fill the back aperture of a microscope objective (Nikon: Infinity corrected, 100 $\times$, NA = 0.9). The microscope objective focuses the beam onto the surface of the sample, to obtain a focus diameter of about 300 nm. In order to observe the flux exiting the front surface of the sample, a combination of the microscope objective and lens L2 images the surface of the sample onto the chip of a CCD camera (Dolphin F-145B). A polarizer and a quarter-wave ($\lambda/4$) plate are placed in the reflection path to suppress the surface reflection from and single scattering inside the sample. The key to this polarization filtering method is that the surface reflections and single scattering retain the same handedness as the incident circularly polarized light from the ($\lambda/4$) plate, while multiple scattering changes the handedness of the light [168,169]. The sample is mounted on a three-axis piezoelectric translational stage (PI NanoCube P-611.3S), which has a travel range of 100 μ m by 100 μ m scanned with a step size $(\Delta x, \Delta y) = 100$ nm in the (x, y) plane perpendicular to the optical axis of the objective. For focusing and alignment purpose the objective is mounted on a manually operated stage with an accuracy $\Delta z = 0.1$ μ m.

In the experiments, we studied light propagation in synthetic opal photonic crystals, which are made of silica colloidal spheres [155, 170]. The colloidal spheres (radius $R = 349$ nm) are assembled on a silicon wafer substrate by vertical controlled drying and are ordered in a hexagonal arrangement, *i.e.*, the fcc (111) plane. Fig. 6.3(a) shows a scanning electron microscope image of an opal crystal grown on a silicon wafer. The opal crystal consists of small grains with an average diameter of 5 μ m, which are created during the growth process. In Fig. 6.3(b), we

show the band structure of the opal photonic crystal and it was calculated using the plane-wave expansion method [80]. The opals have a first stop gap of only 5% relative band width in the $\Gamma - L$ direction. The photon energy of the illumination monochromatic light at $\lambda = 561$ nm is well above the first stop gap and near the second stop gap, in the $\Gamma - L$ direction. The incident light illuminates the opal with a cone of angles that has its center perpendicular to the fcc (111) plane. Light couples into the propagation bands with the same photon energy as the incident light (see red line in Fig. 6.3).

For comparison, we also perform measurements on a two-dimensional (2D) silicon crystal with centered rectangular lattice as shown in Fig. 6.3 (c). The 2D crystal was fabricated by etching pores using reactive ion etching in a silicon wafer [171, 172]. The large single crystal is defined by deep UV step scan lithography [172]. The 2D photonic crystal has a pore radius of $R = 153$ nm and a lattice parameter 691 nm. The measurements on 2D crystals are done in the perpendicular out of plane direction with zero in-plane wavevector where the crystal has a stop gap at the Γ point. The photon energy of the illumination light has a higher energy than the stop gap of the 2D crystal and couples to high-up propagating bands*. As a reference sample, we used an ensemble of disordered of zinc oxide (ZnO) nanoparticles with an average grain size of 200 nm. The ZnO disordered sample has a thickness of 10 μ m, corresponding to 17 transport mean free paths 17ℓ (see Section 4.2.1).

6.3 Distribution of reflected intensity

In Fig. 6.4(a), we show the reflected intensity from a part of the opal with a large crystal domain. The reflected intensity from this part of the crystal has distinct hexagonal peaks. These hexagonal peaks are attributed to the (111) periodic arrangement of the silica spheres in this part of the crystal and to a large contribution of Bloch waves. In contrast to the reflected intensity from a completely disordered ZnO sample shown in Fig. 6.4(b), there is no distinct structure due to the effect of random wave interferences, which results into a speckle pattern. Figs. 6.4(c) and (d) show the images recorded at two different positions on the opal with small crystal grains. The reflected intensities from the two positions, separated by a distance of $\Delta x = 100$ nm, both have some bright spots, which do not show any hexagonal peaks (see Fig. 6.4(a)). In this part of the crystal, we expect that there is a strong contribution of the scattered waves E_s due to the disorder, which result in multiply scattering. Therefore, the reflected intensity from this part of the crystal is speckle-like, as a result of a high contribution of E_s .

We analyze the reflected intensities by studying the histogram, which is expected to reveal the nature of the most dominant waves reflected from the crystal. In a disordered medium, the amplitude and phase of the scattered fields

*The band structure of the 2D silicon photonic crystal needs to be calculated to include wavevectors k_z along z direction, which is not typical of band structures calculated for 2D crystals.

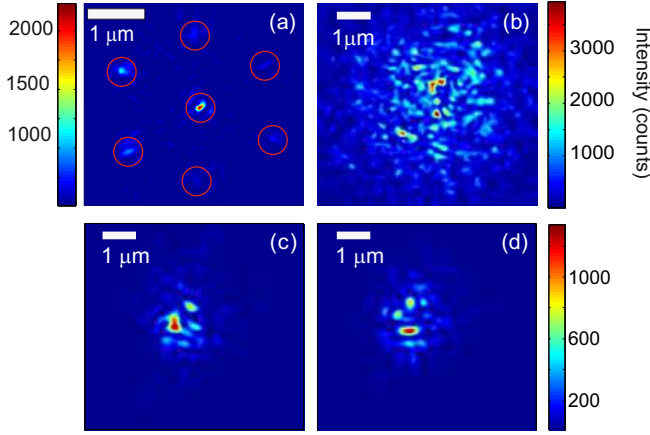


Figure 6.4: CCD camera images of the reflected light from opal and ZnO samples. (a) Reflected intensity from opal with a large crystal grain. The reflected light has hexagonal peaks, which are indicated by the red circles. (b) The reflected light – is a speckle pattern – from a disordered ZnO sample. In (c) and (d) are the measured reflected intensities at two different positions on the opal with a small crystal grain. The distance between the two positions is 100 nm.

is completely randomized due to multiple scattering. The scattered fields are independent and uncorrelated. As a result of the central limit theorem, the joint probability distribution of the real and imaginary parts of the resultant scattered field is a circular Gaussian function [167]. Therefore, the probability distribution of the resultant scattered intensity follows a Rayleigh distribution, which is a single exponential. The intensity of any two points separated by more than a speckle spot radius is uncorrelated [13,15].

In order to investigate the correlation of the reflected intensity, we plot in Fig. 6.5 the histogram of the intensity reflected from the samples and compare with the Rayleigh distribution. The histogram was obtained from the occurrence of intensities within the full-width at half maximum of the beam and the occurrence was normalized to the total occurrence in a single image. As a figure-of-merit to compare different models, we use the reduced χ_R defined as [173]

$$\chi_R = \frac{1}{N-p} \sum_i^N \frac{(O_i^m - O_i^c)^2}{O_i^m} \quad (6.1)$$

where O_i^m is the measured occurrence, O_i^c is the fitted occurrence, N is the total number of data points, and p is the number of adjustable parameters. Assuming Poisson statistics, $\chi_R^2 = 1$, if there is a perfect agreement between data and a fitted distribution, and χ_R^2 is normalized to be independent of the degrees of freedom ($N-p$). As expected, the distribution of intensity from the disordered sample follows the Rayleigh distribution, with $\chi_R^2 = 2.8$, which is

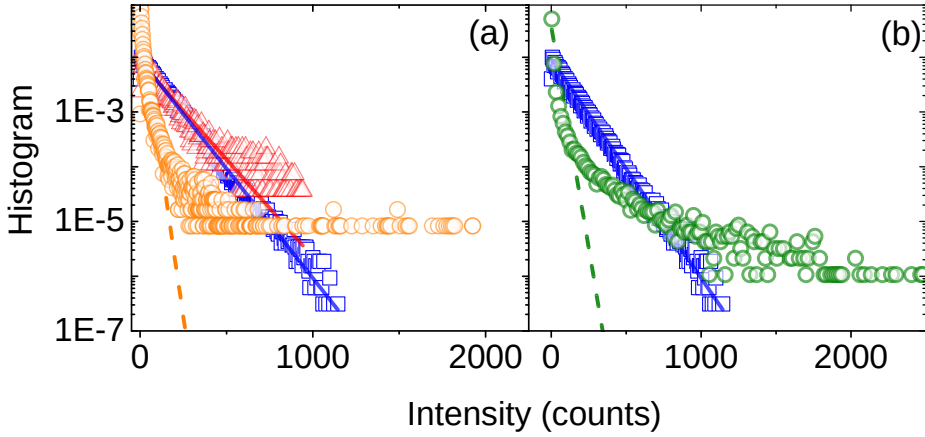


Figure 6.5: (a) Histogram of speckles intensity reflected from the disordered ZnO sample (blue squares), an opal photonic crystal with small crystal grains (red triangles), and with a large crystal (orange circles) grain. The blue solid line, the red solid line, and the dashed orange line are the Rayleigh distributions fitted for the ZnO sample, opal with small, and with large crystal grains, respectively. (b) Probability distribution of light reflected from the 2D silicon photonic crystal (green circles) and the green dashed line is a Rayleigh distribution fit to the data. The blue squares and the blue solid line are the data and fit for ZnO sample respectively, the same as in (a).

an excellent agreement. Interestingly, for the large-domain opal crystal, there is a huge deviation ($\chi_R^2 = 10^{44}$) of the reflected intensity from the Rayleigh distribution. Similarly, we show in Fig. 6.5 (b) the probability distribution of intensity reflected from the 2D photonic crystal, which also has a large crystal domain. For the 2D crystal, there is as well a strongly non-Rayleigh distribution of intensity with $\chi_R^2 = 10^{41}$. These strong non-Rayleigh distributions implies that the reflected fields are strongly correlated, which is the result of a large contribution of Bloch waves. In a perfect crystal, all intensities are completely correlated as they are given by the crystal structure factors [75, 76]. In these large-domain crystals, scattered waves E_s have minimal contribution, since the disorder hardly affect light propagation. There is a slight deviation ($\chi_R^2 = 6.0$) of a single exponential fit from the intensity reflected from the opal photonic crystal with small crystal grains. The deviation from the Rayleigh distribution implies that the reflected intensities are correlated, which is due to the contribution of the Bloch waves E_B . At low intensities, there is however an agreement of the Rayleigh distribution with the measured intensity. We attribute the low intensities to be from the fields that propagate deep inside the sample. These fields have low intensities due to multiple scattering and have a higher probability to encounter disorder compared to fields with high intensity, which have not been

significantly scattered by disorder. Therefore, there is a noticeable contribution of the scattered waves along side with Bloch waves in the opal with small crystal grains.

6.4 Spatial correlations of reflected intensity

We proceed by investigating the similarity between the intensity reflected from different positions on an opal photonic crystal using spatial intensity correlations. We performed a two-dimensional raster scan (x, y) on the crystal surface. At each

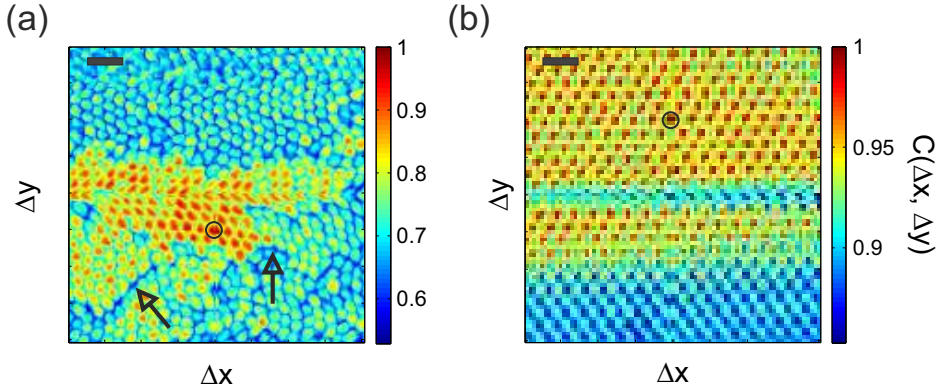


Figure 6.6: A 2D map of correlation of the reflected intensities from an opal photonic crystal (a) and a 2D silicon photonic crystal (b). The intensities reflected at different positions on the samples are correlated using Eq. 6.2. The intensity reflected $I(\zeta, \eta; x_0, y_0)$ from the position indicated by the black circle in both (a) and (b) is the reference position of the correlation. In the correlation map in (a), there are periodic spheres with diameter of $0.6 \mu\text{m}$. The dark blue lines between particles correspond to the grain boundaries in the opal (indicated by black arrows). In (b), the correlation map as well reveals periodic structures over the entire scan area and the periodicity corresponds to the lattice parameters of the 2D photonic crystal. Scale bar in (a) is $1 \mu\text{m}$ and $0.5 \mu\text{m}$ in (b).

position on the sample, we measured the reflected intensity $I(\zeta, \eta; x, y)$, where (ζ, η) are the coordinates on the images. The sample was then translated to another position with a step size $\Delta x = 100 \text{ nm}$ at a fixed y and another image $I(\zeta, \eta; x + \Delta x, y)$ was taken. The raster scan is automated and the scan covered an area of $16 \text{ by } 16 \mu\text{m}^2$. After the data were collected, we chose an image $I(\zeta, \eta; x_0, y_0)$ centered at an arbitrary position (x_0, y_0) as a reference intensity and correlate the other images $I(\zeta, \eta; x_0 + \Delta x, y_0 + \Delta y)$ at positions $(x_0 + \Delta x, y_0 + \Delta y)$ with the reference image $I(\zeta, \eta; x_0, y_0)$. The normalized correlation function

is defined as follows:

$$C(\Delta x, \Delta y) \equiv \frac{\int_0^{\eta_m} \int_0^{\zeta_m} d\zeta d\eta I(\zeta, \eta; x_0, y_0) \cdot I(\zeta, \eta; x_0 + \Delta x, y_0 + \Delta y)}{\left[\int_0^{\eta_m} \int_0^{\zeta_m} d\zeta d\eta I^2(\zeta, \eta; x_0, y_0) \cdot \int_0^{\eta_m} \int_0^{\zeta_m} d\zeta d\eta I^2(\zeta, \eta; x_0 + \Delta x, y_0 + \Delta y) \right]^{\frac{1}{2}}}, \quad (6.2)$$

where ζ_m and η_m are the lengths of the images in the x and y coordinates respectively. Eq. 6.2 enables us to make a map of the correlation over the entire translational area.

Figure 6.6(a) shows the correlation map of the opal crystal. There are at least four interesting features that appear in the correlation map. Firstly, there are periodically-arranged round structures, which have a periodicity of $a_c = 600 \mu\text{m} \pm 100$ that is the nearest-neighbour distance of the periodic structures. The uncertainty stems from the accuracy of the piezo-translation stage. The periodic of the structures a_c matches well with the periodicity of the crystal $a = \frac{\sqrt{3}}{2}(2R) = 0.64 \mu\text{m}$. Naively, one would expect such an agreement but we will see in Section 6.5 a scenario where $a_c \neq a$. Secondly, there are regions close to the reference position that reveal a high correlation of about 0.9. This high correlation implies that the reflected intensity patterns from these positions are very similar to that from the reference position, by at least 90%. The high correlation implies that light reflected from this part of the crystal are mostly due to Bloch waves E_B , which is expected to result into a high correlation. The positions with a lower correlation implies that the reflected intensity are less similar compared to the reference position, which is probably due to a higher contribution of E_s from disorder. Thirdly, it is remarkable that in the highly ordered region, *e.g.*, near the reference point in Fig. 6.6, the correlation varies spatially. This variation is attributed to different spatial in-coupling as well as out-coupling at different positions on the crystal surface, as previously observed in near-field measurements [157, 174]. Fourthly, it is remarkable that the outline of the grain boundaries appear in the correlation map. The grain boundaries are apparent as dark blue lines on the correlation map, two of which are indicated with the arrows. At the grain boundaries, the correlation is the lowest at about 0.6. The low correlation is a result of random multiple scattering events, which result in a different speckle pattern.

Fig. 6.6 (b) shows the correlation map for the 2D silicon crystal. The map shows periodicity in the entire map. From the correlation map, we retrieve the periodicity in the x -direction to be $a = 700 \text{ nm} \pm 100$ and in the y -direction $c = 500 \text{ nm} \pm 100$. From the SEM images, the two lattice constants are $a = 693 \pm 10 \text{ nm}$ and $c = 488 \pm 11 \text{ nm}$ and our observation from the correlation map matches very well with the lattice parameters of the crystal. At some parts in the image the correlation is lower than the other parts. This might be due to some drift in the set-up during the long scan that lasted for about 48 hours. Another possibility is that the sample surface has a small curvature, causing the sample to move out of focus during the scan. Since the correlation scan was

done symmetrically with respect to the $y=0$ line, an instability cannot cause an asymmetric correlation, we therefore attribute the low correlation region to the curvature of the sample surface.

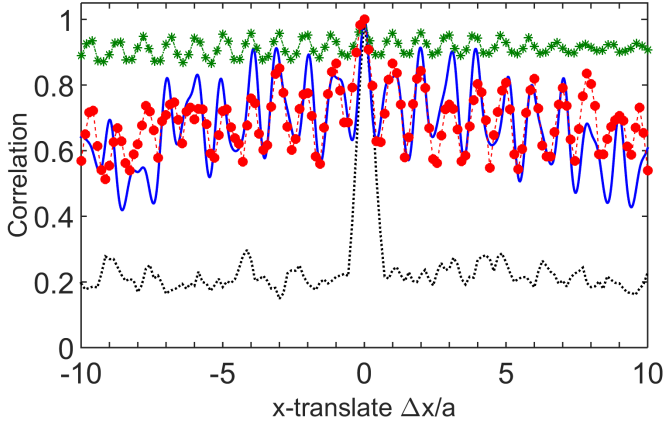


Figure 6.7: Intensity correlation versus x -translation Δx . The green stars and red dots are the data, which are the cross-sections through a fixed y at the reference position in Fig. 6.6 for the 2D silicon and opal photonic crystal respectively. The green and red dashed curve are guides-to-the-eye. The blue straight curve is the correlation model described in Section 6.5. The black dotted line is the correlation from a completely disordered ZnO nanoparticles sample.

In Fig. 6.7, we compare a line profile of the correlation map from the opal photonic crystal with the correlation from a completely disordered ZnO sample. The ZnO sample provides a reference correlation of what to expect from a sample where multiple scattering dominates. The reference ZnO sample decorrelates quickly within $\Delta x = 0.6 \mu\text{m}$ to a level of about 0.2 and there is no periodicity in the spatial correlations, as expected for a random medium. The background correlation level of 0.2 is unexpectedly high for a disordered sample and the reason is probably due to reflections from optical components. For the opal crystal, the correlation reduces to 0.6, and periodicity is seen over the entire range of the translation. This higher level of correlation compared to the ZnO sample is due to a higher contribution of E_B . In the case of the 2D Si photonic crystal, there is a high correlation level of about 0.9. This high correlation level is obtained by the major contribution of E_B , which confirms the result from the strongly non-Rayleigh distribution of intensity from the large-domain crystal. The single crystalline structure and the extreme control of disorder in top-down fabrication method of the silicon photonic crystal result in a nearly perfect crystal structure. The lower correlation in opal compared to the silicon crystal is expected due to grain boundaries and the size and position disorder of the opal.

6.5 Theoretical description of intensity correlation

In this section, we develop a theoretical model to describe intensity correlation of waves reflected from a photonic crystal with disorder. We first state our assumptions and then describe the field exiting the photonic crystal. From the calculated field and then intensity, we obtain the spatial intensity correlation.

6.5.1 Assumptions

1. We consider that a photonic crystal with lattice constant a is illuminated with an incident field $\mathbf{E}_{\text{in}}(\mathbf{r}, \mathbf{k}_{\text{in}})$. We assume that the incident plane waves are converted to Bloch waves $\mathbf{E}_{\text{m}}^{\text{B}}(\mathbf{r}, \mathbf{k}_m)$ with a wave vector $\mathbf{k}_m$ (see Fig. 6.1) after propagating through a distance that is assumed to be of the order of the Bragg length L_B .
2. In the photonic crystal, scattering due to disorder (*e.g.*, grain boundaries of opal, position and size variation) plays a role in the propagation of waves inside the crystal. Here, we consider that spherical waves are generated at the site of the disorder, as shown in Figure 6.1.
3. It is possible that disorder scatter a Bloch wave into another Bloch wave with a different wavevector that is allowed by the band structure of the crystal. We have neglected this possibility since the Bloch wave character becomes apparent after a distance of the order of the Bragg length, which is comparable to the average crystal grain size.
4. We assume that the fields propagating inside the photonic crystal have M wavevectors allowed by the band structure of the crystal. The M wavevectors are obtained from the bands that have the same photon energy as the incident light (see. Fig. 6.3). This assumption might become invalid as the amount of disorder in the photonic crystal tends towards a disordered medium, since the periodic translation symmetry is completely broken in this regime. For our opal photonic crystal, the transport mean free path ℓ is of the order of $10 \mu\text{m}$ [151] and sample thickness $L = 5 \mu\text{m}$, $L/\ell \sim 0.5$ compared to $L/\ell = 17$ of a ZnO scattering medium, which is completely disordered. Therefore, we assume that the disorder in the opal is mild to introduce sufficiently larger number of wavevectors than M .
5. We consider that the reflected field is a coherent sum of Bloch waves and scattered spherical waves, both with wavevectors different from the incident ones.

6.5.2 Total fields exiting a real disordered photonic crystal

We consider that the total field $\mathbf{E}_{\text{out}}(\mathbf{r})$ reflected by the photonic crystal with disorder comprises of Bloch waves $\mathbf{E}_{\text{m}}^{\text{B}}(\mathbf{r}, \mathbf{k}_m)$ and scattered waves $\mathbf{E}_{\text{m}}^{\text{s}}(\mathbf{r}, \mathbf{k}_m)$

both with wavevectors $\mathbf{k}_m$ allowed by the band structure (see Fig. 6.3b). We postulate

$$\mathbf{E}_{\text{out}}(\mathbf{r}_o) = A_{\text{out}} \left[(1 - \beta) \sum_m^M \mathbf{E}_m^B(\mathbf{r}_o, \mathbf{k}_m) + \beta \sum_m^M \mathbf{E}_m^S(\mathbf{r}_o, \mathbf{k}_m) \right], \quad (6.3)$$

where $\mathbf{r}_o$ is the position of observation, M is the number of wave vectors that are supported by the crystal and β is the ratio of the amplitude of the scattered waves to Bloch waves. Considering that intensity rather than field is measured, we define the expected $|\beta_e|^2$

$$|\beta_e|^2 \equiv \frac{1}{1 + \frac{\ell}{L_B}}. \quad (6.4)$$

A photonic crystal without any defect has $\ell/L_B \rightarrow \infty$, and $\beta \rightarrow 0$ while a disordered photonic crystal has $\ell/L_B \rightarrow 0$, and $\beta \rightarrow 1$.

A_{out} normalizes $\mathbf{E}_{\text{out}}(\mathbf{r})$ with condition that the waves are within the crystal

$$A_{\text{out}}^{-2} = \int_0^{\mathbf{L}} |\mathbf{E}_{\text{out}}(\mathbf{r}_o)|^2 d\mathbf{r}_o, \quad (6.5)$$

where $\mathbf{L}$ is a vector that specifies the extent of the crystal in the three directions (x, y, z) . Before we derive an expression for $\mathbf{E}_{\text{out}}(\mathbf{r})$, we first consider a reflected field $\mathbf{E}_m^{\text{out}}(\mathbf{r}, \mathbf{k}_m)$ with a wavevector $\mathbf{k}_m$. We sum over all fields from the different layers (i, j, k) in the x-, y- and z- directions (see Fig. 6.1) to obtain

$$\mathbf{E}_m^{\text{out}}(\mathbf{r}_o, \mathbf{k}_m) = (1 - \beta) \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} \mathbf{E}_m^B(\mathbf{r}_{i,j,k}, \mathbf{k}_m) + \beta \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} \mathbf{E}_m^S(\mathbf{r}_{i,j,k}, \mathbf{k}_m), \quad (6.6)$$

where $\mathbf{r}_{i,j,k} = \mathbf{r}_{i,j,k}^s - \mathbf{r}_o$, $\mathbf{r}_{i,j,k}^s$ is the position of the source of the wave, and N_i , N_j , and N_k are the number of layers in the x-, y-, z- and directions respectively. We assume that β is constant for all wavevectors, which is a first approximation to make the model tractable. Bloch waves are known to be plane waves that are modulated by a periodic function $\mathbf{u}_m(\mathbf{r})$, such that $\mathbf{u}_m(\mathbf{r}) = \mathbf{u}_m(\mathbf{r} + \mathbf{a})$ [24]. Therefore

$$\mathbf{E}_m^B(\mathbf{r}_{i,j,k}, \mathbf{k}_m) = A_B \exp(i\mathbf{k}_m \mathbf{r}_{i,j,k}) \mathbf{u}_m(\mathbf{r}_{i,j,k}), \quad (6.7)$$

where A_B takes care of the normalization of $\mathbf{E}_m^B(\mathbf{r}_{i,j,k}, \mathbf{k}_m)$, using a similar condition as Eq. 6.5. The field of a scattered wave from a layer (i, j, k) $\mathbf{E}_m^S(\mathbf{r}_{i,j,k}, \mathbf{k}_m)$ is

$$\mathbf{E}_m^S(\mathbf{r}_{i,j,k}, \mathbf{k}_m) = \frac{A_s}{\mathbf{r}_{i,j,k}} \exp(i\mathbf{k}_m \mathbf{r}_{i,j,k}), \quad (6.8)$$

where A_s normalizes the scattered field also following Eq. 6.5. Substituting Eqs.

6.7 and 6.8 into 6.6 we get

$$\begin{aligned} \mathbf{E}_m^{\text{out}}(\mathbf{r}_{i,j,k}, \mathbf{k}_m) = & A_{\text{out}} \left[(1 - \beta) \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} A_B \exp(i\mathbf{k}_m \mathbf{r}_{i,j,k}) \mathbf{u}_m(\mathbf{r}_{i,j,k}) \right. \\ & \left. + \beta \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} \frac{A_s}{\mathbf{r}_{i,j,k}} \exp(i\mathbf{k}_m \mathbf{r}_{i,j,k}) \right]. \end{aligned} \quad (6.9)$$

Summing over all the allowed wavevectors M , the total field reflected from the crystal $\mathbf{E}_{\text{out}}(\mathbf{r})$ is

$$\begin{aligned} \mathbf{E}_{\text{out}}(\mathbf{r}_{i,j}) = & A_{\text{out}} \left[\sum_m^M \left[(1 - \beta) \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} A_B \exp(i\mathbf{k}_m \mathbf{r}_{i,j,k}) \mathbf{u}_m(\mathbf{r}_{i,j,k}) \exp(i\phi_{i,j,k}) \right. \right. \\ & \left. \left. + \beta \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} \frac{A_s}{\mathbf{r}_{i,j,k}} \exp(i\mathbf{k}_m \mathbf{r}_{i,j,k}) \exp(i\phi_{i,j,k}) \right] \right]. \end{aligned} \quad (6.10)$$

Here $\phi_{i,j,k}$ is a random phase acquired due to scattering and as a result of reflection and weak scattering, we assume that $\phi_{i,j,k}$ is centered at π , with a negligible spread: $\phi_{i,j,k} = \phi \approx \pi$. The function $\mathbf{u}_m(\mathbf{r})$ is the periodic part of the Bloch function and it is written as

$$\mathbf{u}_m(\mathbf{r}) = \cos(\mathbf{G}_m \mathbf{r}), \quad (6.11)$$

where $\mathbf{G}_m$ is the reciprocal vector, which is equal to $\mathbf{G}_m = 2\pi/\mathbf{R}$, with $\mathbf{R}$ a lattice vector. Substituting Eq. 6.11 into Eq. 6.10 and expressing the exponential terms as cosines and sines, we have

$$\begin{aligned} \mathbf{E}_{\text{out}}(\mathbf{r}) = & A_{\text{out}} \left(\sum_m^M \left[(1 - \beta) \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} A_B [\cos(\mathbf{k}_m \mathbf{r}_{i,j,k} + \phi) \cos(\mathbf{G}_m \mathbf{r}) + \beta \frac{A_s}{\mathbf{r}_{i,j}} \cos(\mathbf{k}_m \mathbf{r}_{i,j,k} + \phi)] \right. \right. \\ & \left. \left. + iE_0 \sum_m^M \left[(1 - \beta) \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} A_B [\sin(\mathbf{k}_m \mathbf{r}_{i,j,k} + \phi) \cos(\mathbf{G}_m \mathbf{r}) + \beta \frac{A_s}{\mathbf{r}_{i,j,k}} \sin(\mathbf{k}_m \mathbf{r}_{i,j,k} + \phi)] \right] \right] \right). \end{aligned} \quad (6.12)$$

Since $\mathbf{E}_{\text{out}}(\mathbf{r})$ is complex, it can therefore be expressed as

$$\mathbf{E}_{\text{out}}(\mathbf{r}) = \Re[\mathbf{E}_{\text{out}}(\mathbf{r})] + i\Im[\mathbf{E}_{\text{out}}(\mathbf{r})] \quad (6.13)$$

The total intensity exiting the crystal is

$$I_{\text{out}}(\mathbf{r}) = \mathbf{E}_{\text{out}}(\mathbf{r}) \mathbf{E}_{\text{out}}^*(\mathbf{r}) = \Re[\mathbf{E}_{\text{out}}(\mathbf{r})]^2 + \Im[\mathbf{E}_{\text{out}}(\mathbf{r})]^2. \quad (6.14)$$

On substituting Eq. 6.12 into Eq. 6.14 and using trigonometry identities, we

obtain an expression for the intensity exiting a real and finite photonic crystal

$$\begin{aligned}
I_{\text{out}}(\mathbf{r}_o) = & \frac{A_{\text{out}}^2}{4} \left(\sum_m^M \left[(1 - \beta) \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} [A_B \cos[(\mathbf{k}_m + \mathbf{G}_m)\mathbf{r}_{i,j,k}] + \cos[(\mathbf{k}_m - \mathbf{G}_m)\mathbf{r}_{i,j,k}]] \right. \right. \\
& + \beta \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} \frac{A_s}{\mathbf{r}_{i,j,k}} \cos(\mathbf{k}_m \mathbf{r}_{i,j,k}) \left. \left. \right] \right)^2 \\
& + \frac{A_{\text{out}}^2}{4} \left(\sum_m^M \left[(1 - \beta) \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} [A_B \sin[(\mathbf{k}_m + \mathbf{G}_m)\mathbf{r}_{i,j,k}] + \sin[(\mathbf{k}_m - \mathbf{G}_m)\mathbf{r}_{i,j,k}]] \right. \right. \\
& + \beta \sum_k^{N_k} \sum_j^{N_j} \sum_i^{N_i} \frac{A_s}{\mathbf{r}_{i,j,k}} \sin(\mathbf{k}_m \mathbf{r}_{i,j,k}) \left. \left. \right] \right)^2. \tag{6.15}
\end{aligned}$$

6.5.3 Model of the intensity correlation

Using Eq. 6.15, the normalized cross correlation $C(\Delta\mathbf{r})$ of the intensity from a reference point $\mathbf{r}_0$ and intensity at position $\mathbf{r}_0 + \Delta\mathbf{r}$ is obtained as

$$C(\Delta\mathbf{r}) \equiv \frac{\int_0^{L_t} I(\mathbf{r}_0) I(\mathbf{r}_0 + \Delta\mathbf{r}) d\mathbf{r}}{\left[\int_0^{L_t} I(\mathbf{r}_0) I(\mathbf{r}_0) d\mathbf{r} \int_0^L I(\mathbf{r}_0 + \Delta\mathbf{r}) I(\mathbf{r}_0 + \Delta\mathbf{r}) d\mathbf{r} \right]^{\frac{1}{2}}}, \tag{6.16}$$

where $\Delta\mathbf{r}$ is the translation from the reference position $\mathbf{r}_0$ and L_t is the transverse length until which the intensity is detected. $C(\Delta\mathbf{r})$ is normalized following Ref. [48]. In Fig. 6.8, we plot the correlation C from Eq. 6.16 as a function of translation along x-direction Δx for different values of β . To determine the allowed wavevectors by the opal crystal for Eq. 6.15, the bands with the same photon energy as the incident light were considered. We ignore propagation bands with nearly flat dispersion as these bands have a high impedance (reflectance $> 90\%$), which is a result of the high effective group refractive index. Therefore, they do not contribute to the light propagating inside the crystal and they also contribute negligibly to the measured intensity.

From Fig. 6.8, we observe that when $\beta = 0$, *i.e.*, the reflected field is only due to Bloch waves, the periodicity of the correlation is $a_c = a/2$. The $a/2$ periodicity of the correlation stems from the interference of the Bloch waves with each other, and therefore the intensity, which is the square modulus of the Bloch field with a period a , has a periodicity of $a/2$. As β increases, *i.e.*, the contribution of the scattered waves increases, the correlation periodicity a_c tends towards the periodicity of the crystal $a_c \rightarrow a$. The reason is because in this regime, the resultant field inside the crystal is an interference between the periodic field E_B and E_s , which is a slowly varying field. Therefore, the resultant field has mostly positive values, which when squared to obtain the intensity, still retains the periodicity a . From the observed periodicity of the correlation a_c , a lower

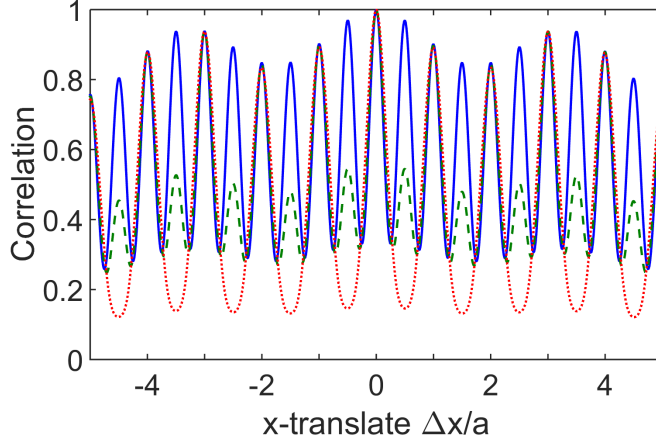


Figure 6.8: Spatial correlation as a function of reduced x-translation $\Delta x/a$ calculated using Eq. 6.16 for different values of β . β is the ratio of the amplitude of the scattered waves to Bloch waves. The values of β are $\beta = 0, 0.4$ and 0.7 for the blue solid, green dashed, and red dot curves respectively. The wavevectors used in the plots are $\mathbf{k} = (0.07, 0.12, 0.37, 0.39, 1.63) \times 2\pi/a$, determined from the band structure in 6.4. The finite size of the crystal is equal to $N_i = 7$, $N_j = 20$, $N_k = 20$ and $\phi = \pi$.

limit of β can be deduced.

In Fig. 6.7, we compare our theoretical model of the correlation with the experimental result. From the experimental data, the periodicity of the correlation $a_c = a$. As described above, since $a_c = a$, we can already conclude that the total reflected field has some contribution from the scattered waves and not only Bloch waves. In our correlation model, the only adjustable parameter is β and with $\beta = 0.8$, the model agrees well with our experimental result. To compare with the expected value of $|\beta_e|^2$ in Eq. 6.4, we use $\ell \approx 10 \mu\text{m}$ determined from enhanced backscattering cone measurement [151] and $L_B = 4.7 \mu\text{m}$, which was calculated using $L_B = 2d/(\pi S)$. The lattice spacing between two crystal planes $d = \frac{a}{\sqrt{h^2 + k^2 + l^2}}$, for the (111) plane, $d = 0.37 \mu\text{m}$. The photonic parameter $S = 0.05$ was obtained from the relative width of the first stop gap. Substituting these values of ℓ and L_B into Eq. 6.4, we obtain $|\beta_e|^2 = 0.32$, which is remarkably very close to $|\beta|^2 = 0.36$ retrieved from our model, after accounting for the 0.2 offset, due to the spurious reflections observed in Fig. 6.7. This value of $|\beta|^2$ that is close to the mid value between 0 and 1 implies that the contribution of the Bloch and scattered waves is of the same order of magnitude.

6.6 Conclusion

We have shown a new method to probe the nature of waves propagating inside a real photonic crystal with disorder. We employ optical correlation of the waves

reflected from synthetic opal photonic crystal and 2D silicon photonic crystal as a probe of the wave structure. We find out that the reflected wave from the synthetic opal has a comparable contribution of Bloch waves and scattered waves. Our method is capable of probing deep inside the crystal and finding the structural wave correlations in photonic crystals and is relevant for numerous applications of photonic crystal devices.

CHAPTER 7

Controlling the intensity of light in large areas at the interfaces of a scattering medium

7.1 Introduction

Wave interference in disordered scattering media results in speckles through the coherent addition of multiple waves, which are independent and have random amplitudes and phases [36]. Between these interfering waves, there exist short-, long-, and even infinite-range correlations [13–20]. These correlations have provided enriching information about mesoscopic transport as well as a deeper understanding of fundamental phenomena such as enhanced backscattering [10, 11] and Anderson localization [9].

In 1990, using speckle correlations, Freund predicted that an opaque scattering medium can be used as a lens and other optical elements by designing an appropriate incident wavefront [48]. Only recently, this prediction was confirmed by the advent of innovative wave-shaping methods such as wavefront shaping [49, 83–87], time reversal [51–53], phase conjugation [54–56], and transmission-matrix-based control [57–59]. In wavefront shaping, an optimization algorithm receives as a feedback the intensity in a target area, typically one speckle spot with an area $A = \lambda^2/2\pi$. The algorithm then modifies the spatial phase of the incident field on the scattering medium, such that the intensity in the target spot is maximized. These wave-shaping methods have led the way for exciting applications such as non-invasive biomedical imaging [60–62], advanced optics [63–68], and cryptography and secure communication [69, 70].

In an earlier experiment [83], it was observed that there is not only an intensity enhancement in the target speckle spot but also in an area outside the target speckle spot. Consequently, the total transmission is enhanced, even though only the intensity in a single speckle spot is monitored. An intuitive explanation for this observation is that there is a redistribution of energy between reflection and transmission channels, since absorption is negligible in the scattering samples used. Here, we take a step further by investigating how the enhancement of the total transmission depends on the optimization area. In the absence of absorption, we expect to observe a concomitant effect in the total reflected intensity. Moreover, we expect to find the effect of long-range correlations,

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especially of the form that exists between the reflected and transmitted speckles, as recently predicted in Ref. [175]. An optimization of the total intensity transmitted through a scattering medium, which is the extreme case of our study, has been performed in Ref. [84] where the optimization area was not systematically varied. The dependency that we seek will give insight to the intensity redistribution between the transmitted and reflected speckles. Such a fundamental understanding is useful for applications of wavefront shaping in efficient energy harvesting in solar cells [82,93,94], multi-channel quantum secure communications [68,176,177], imaging [62,178,179], and complex beam delivery through a scattering medium [180].

In this chapter, we experimentally study how the optimized intensity on both interfaces of a scattering medium depends on the optimization area on the transmission side. We imaged the transmitted intensity onto the chip of a camera, and thus, there is a freedom to control the optimization radius. We observe that as the optimization radius increases, the enhancement of the total transmitted intensity increases. We find a concomitant decrease of the total reflected intensity, which implies that there is a redistribution of intensity from reflection to transmission. In addition, our result reveals qualitative evidence of the long-range reflection-transmission correlation.

7.2 Experimental details

7.2.1 Experiment set-up

The experimental set-up is shown in Figure 7.1. The light source is a Helium Neon (HeNe) laser, which emits at a wavelength $\lambda = 632.8$ nm and has an output power of 5 mW. A combination of a half-wave plate ($\lambda/2$) and a polarizer (P) controls the incident power and also fixes the polarization of the beam. A beam expander with a magnification of $20\times$ expands the beam to fill the active area of the spatial light modulator (SLM). The spatial light modulator is a Holoeye Pluto (6010 - NIR - 011), which has 1920×1080 pixels and controls the horizontal polarization. A beam splitter (BS) separates the incident and reflected light from the SLM. The reflected light is focused by a lens L1 (focal length $f = 250$ mm). Aperture A, placed at the focal plane of the lens L1, filters the higher diffraction orders of the SLM's pixels and transmits only the 0^{th} order. With a telescope consisting of lenses L1 and L2 ($f = 250$ mm), the SLM is imaged onto the pupil of a microscope objective MO1 (Zeiss: Infinity corrected, $63\times$, NA = 0.95), which focuses the light onto the surface of the sample. The sample is an ensemble of disordered zinc oxide nanoparticles that are spray-painted on top of a glass cover slide. The sample thickness is $17 \pm 0.2 \mu\text{m}$ and the transport mean free path ℓ of similar samples was reported in Chapter 4 and Ref. [138] to be $\ell = 0.6 \pm 0.2 \mu\text{m}$. The intensity transmitted through the sample is imaged onto the chip of a charged-coupled device (CCD) camera (CCD1) using a combination of an oil-immersion objective MO2 (Olympus: Infinity corrected, $60\times$, NA = 1.42) and lens L3 ($f = 500$ mm). The calculated magnification of imaging the back surface of the sample (M1) is $167\times$. Similarly, a combination of the focusing objective MO1 and lens L2

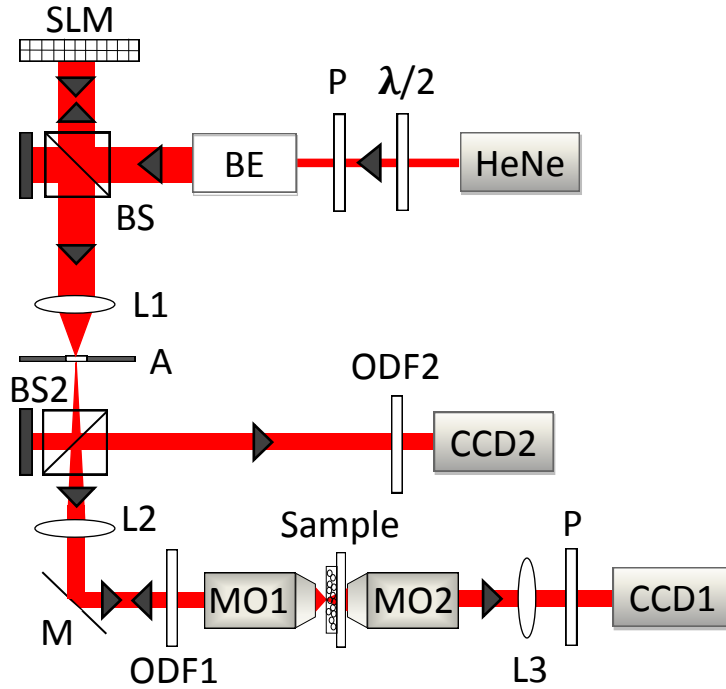


Figure 7.1: Experimental set-up. A Helium Neon (HeNe) laser beam is expanded and modulated by a spatial light modulator (SLM). The light is directed towards the microscope objective (MO1) (numerical aperture $NA = 0.95$) and then focused onto a multiple scattering sample. The sample is made of disordered zinc oxide (ZnO) particles. The intensity transmitted through the sample is imaged onto the chip of a charged-coupled device (CCD) camera (CCD1) through an oil-immersion objective ($NA = 1.42$) and lens L3. The reflected intensity is also imaged through a focusing objective and lens L2 and detected by a CCD camera (CCD2). P: polarizer, $\lambda/2$: half-wave plate, BE: beam expander, BS: beam splitter, A: aperture, M: mirror.

images the reflected intensity exiting the front surface of the sample onto the chip of a CCD camera (CCD2). The calculated magnification on the reflection side is $95\times$. The cameras CCD1 and CCD2 are both of the same type (AVT Dolphin 145B), with a pixel pitch of $6.45\ \mu\text{m}$. Using the calculated magnifications, the pixel pitches on the front and back surfaces of the sample are $0.068\ \mu\text{m}$ and $0.038\ \mu\text{m}$ respectively. The optical density filter ODF1 (Thorlabs NE05A), with a measured attenuation factor $AF = 0.33$, attenuates the incident intensity on the sample, in order to prevent saturation of the cameras. The reflected intensity is further attenuated by placing ODF2 (Thorlabs NE10A), with a measured $AF = 0.10$ in the reflection path. The reflected intensity is in total attenuated by a

factor of 0.033.

In the set-up of Ref. [84], it is impossible to control the optimization area since the scattering sample was directly attached to a photodetector. Moreover, as a result of the refractive index contrast (approximately a factor of 2) between the sample and the detector, the detected signal in Ref. [84] is limited by significant surface reflections between the sample and the photodetector. With our set-up, we have the freedom to control the optimization area. A further advantage of our study is that surface reflections are suppressed since there is an index match between the sample substrate and the immersion oil.

7.2.2 Experimental procedure and parameters

In order to optimize multiple speckle spots, we used the partitioning algorithm, which is described in Ref. [50] rather than the piecewise algorithm, which is typically used in previous wavefront shaping experiments [49, 83, 84, 100, 181]. We find that the partitioning algorithm out performs the piecewise and genetic algorithms for optimizing multiple speckle spots (see Appendix). In the optimization, the number of segments used is 5000. We systematically increased the number of transmission channels by increasing the optimization radius r_o . The number of transmission channels M scales linearly with the probed area $A (= \pi r_o^2)$ following the equation

$$M = \frac{2\pi A n_e^2}{\lambda^2} \frac{\ell}{L}, \quad (7.1)$$

where n_e is the effective refractive index of the scattering medium [114, 115]. For a specific optimization radius, we repeated the wavefront shaping experiment for about 3 to 5 times at a fixed position on the sample. As a reference phase pattern, we displayed 100 randomly generated patterns, with the same number of segments as the optimized pattern. Compared to the optimized pattern, these randomly generated phase patterns have diffraction losses and a power incident on the sample that is larger by only 5%, which underestimates the intensity enhancement by this insignificant amount.

To quantify the enhancement η_{targ} of the total intensity in the target area, we define $\eta_{\text{targ}} \equiv \frac{P_{\text{targ}}^o}{\langle P_{\text{targ}}^u \rangle}$ following Refs. [49, 136]. P_{targ}^o and P_{targ}^u are the power in the target area with the optimized and unoptimized patterns, respectively. $\langle \rangle$ denotes an ensemble-averaged power over the 100 different random phase patterns. We also quantified the enhancement $\eta_{\text{tr, re}}$ of the total transmitted intensity and the total reflected intensity using $\eta_i \equiv \frac{P_i^o}{\langle P_i^u \rangle}$. Here, $i = \text{tr}$ for transmitted light, $i = \text{re}$ for reflected light, P_{tr}^o and $\langle P_{\text{tr}}^u \rangle$ are the total transmitted power through the sample with the optimized and unoptimized patterns respectively and P_{re}^o , and $\langle P_{\text{re}}^u \rangle$ are the total reflected power through the sample with the optimized and unoptimized patterns respectively. The enhancement of the intensity outside the

optimization area is quantified as

$$\eta_{\text{out}} \equiv \frac{P_{\text{tr}}^o - P_{\text{targ}}^o}{\langle P_{\text{tr}}^u - P_{\text{targ}}^u \rangle}. \quad (7.2)$$

7.3 Results

7.3.1 Radial distribution of transmitted intensity

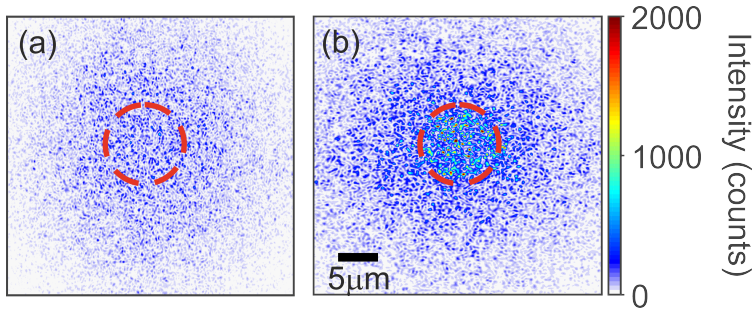


Figure 7.2: Camera images of transmitted intensity at the back surface of a zinc oxide sample. In (a) and (b), unoptimized and optimized wavefronts were displayed respectively on the spatial light modulator (SLM). The optimization radius $r_o = 4.72 \mu\text{m}$, which is indicated by the red dashed circle.

In Figs. 7.2 (a) and (b), we show the CCD camera images of the transmitted intensity for the unoptimized and optimized incident wavefronts, respectively. In the wavefront shaping experiment shown in Fig. 7.2 (b), the optimization radius is $4.7 \mu\text{m}$, which corresponds to 121 pixels. The intensity in the optimization area increases significantly compared to the unoptimized intensity and the intensity outside the target area increases as well. For a better visualization of the intensity increase, we plot the radial distributions of the transmitted intensity in Fig. 7.3. The radial distribution is obtained by summing the intensities within a ring with a width δr and an inner radius of r_r (see inset in Fig. 7.3). Averaging with angle helps to reduce the intensity fluctuation from the speckle pattern. There is a significant intensity increase in the optimization area for both optimization radii $r_o = 15.2 \mu\text{m}$ and $r_o = 4.7 \mu\text{m}$. This intensity increase is expected since the intensity in the optimization area is the feedback to the partitioning algorithm. The intensity outside the optimization area remarkably increases as well. This intensity increase agrees with the observation by Vellekoop and Mosk in Ref. [83], where the intensity outside the optimization area was observed to increase as well. We quantify the intensity enhancement inside and outside optimization areas, and the total transmitted intensity in the next sections.

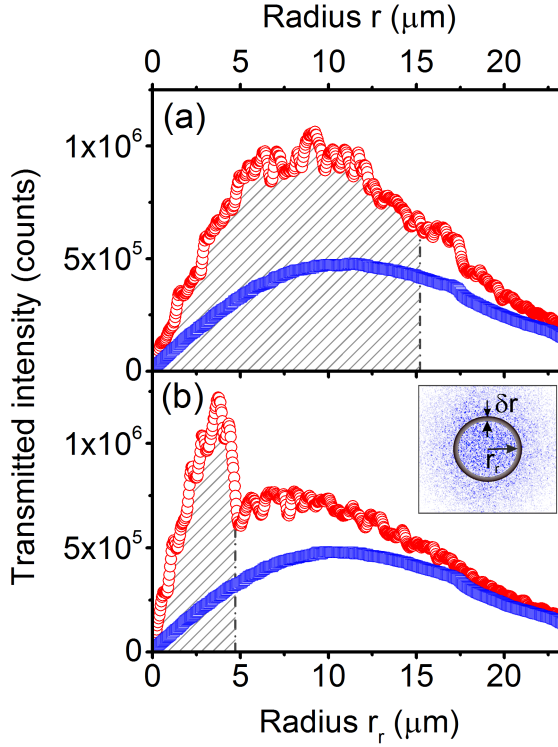


Figure 7.3: Transmitted intensity versus radius r . The transmitted intensity is the total intensity within the circumference of a ring, which has an inner radius r and ring width δr (see inset). The optimization radii r_o are (a) $r_o = 15.2 \mu\text{m}$ and (b) $r_o = 4.7 \mu\text{m}$. The hatched area under the curves is the optimization area. The red circles and blue squares are the intensities for the optimized and unoptimized incident wavefronts respectively.

7.3.2 Enhancement of the intensity in the optimization area

We plot in Fig. 7.4 the intensity enhancement η_{targ} in the optimization area versus the optimization radius r_o . We obtained a maximum enhancement of $257\times$ for an optimization radius of about $0.3 \mu\text{m}$, which corresponds to one speckle spot. As the optimization radius increases, η_{targ} decreases drastically down to $2\times$ at the largest radius $r_o = 15.2 \mu\text{m}$, which corresponds 1716 open transmission channels. In Ref. [144], the enhancement for a multiple channel optimization was found to be inversely proportional to the number of speckle spots M in the optimization area. In the model, it is assumed that the optimized intensity is distributed equally to all optimized channels, which are considered statistically independent. Therefore, the enhancement is expected to depend inversely on the square of the optimization radius r_o ,

$$\eta_{\text{targ}} = \frac{K}{r_o^2} + 1, \quad (7.3)$$

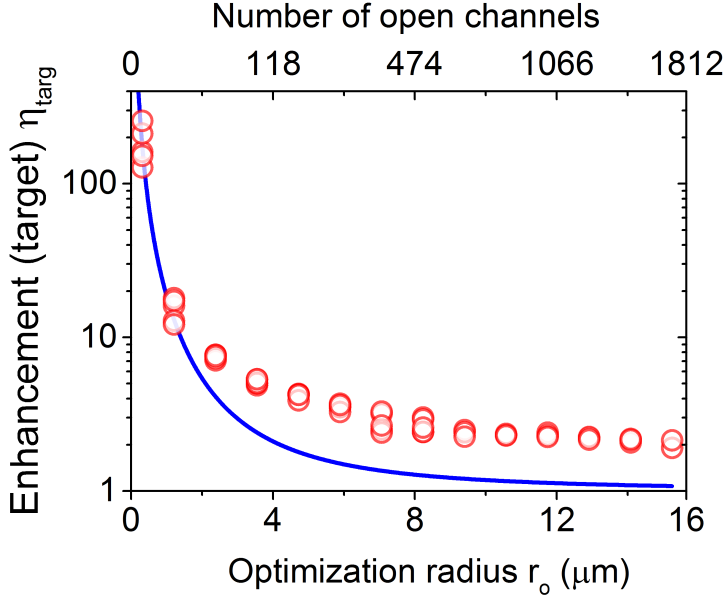


Figure 7.4: Enhancement η_{targ} in the target area versus optimization radius r_o . The red circles are the experimental data and the blue curve is an inverse square law fit following the prediction of Ref. [144].

where K is a constant that depends on the number of effectively controlled input channels on the sample. In Fig. 7.4, we show a fit to the experimental data using Eq. 7.3 and K is the only adjustable parameter. The fit agrees well with the first two optimization radii $r_o = 0.3 \mu\text{m}$ and $1 \mu\text{m}$ that have 1 and 7 transmission channels respectively. Strikingly, the fit deviates from the experimental data by about a factor of 2 at large radii. This deviation signifies that there is more intensity available in the optimization channels, than that predicted according to Eq. 7.3, especially at large radii.

We discuss three possible reasons for the deviation of the theory from the experimental data. First, it is known that noise has a significant effect on the single-speckle optimization [136]. Our wavefront shaping experiments are in the regime where shot noise is much higher than the camera and laser noise that are about 1% and 2% respectively. According to Ref. [136], in this shot noise regime the enhancement of a single-speckle optimization is expected to scale linearly with the total intensity in the optimization area P_{targ} . Extending this model to the optimization of multiple channels, we derive

$$\eta_{\text{targ}} = \frac{K P_{\text{targ}}}{r_o^2} + 1 = K' + 1. \quad (7.4)$$

The constant $K' \equiv KC$, where C is a pre-factor in the relationship $P_{\text{targ}} = C r_o^2$. A constant enhancement with radius obviously does not describe our

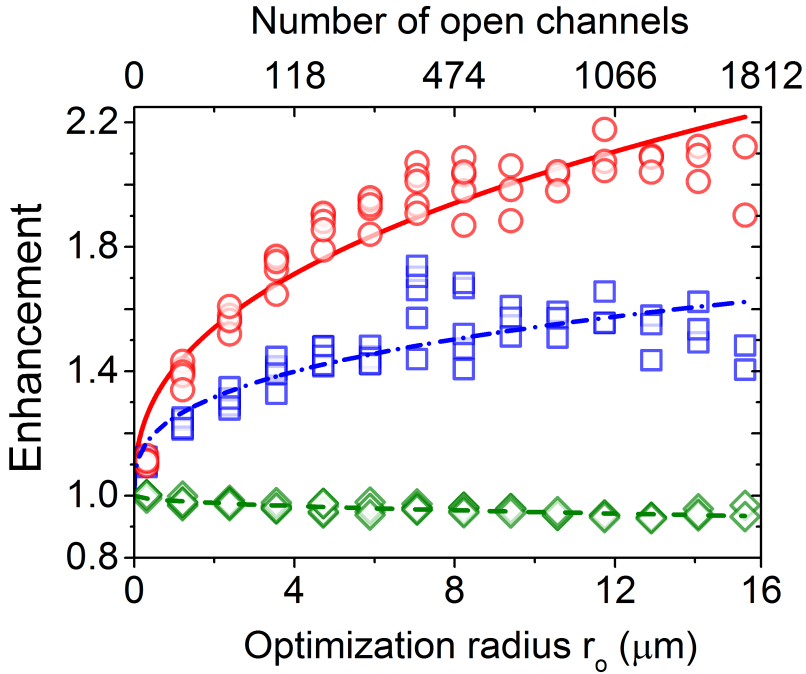


Figure 7.5: Enhancement versus optimization radius r_o . The red circles are the experimental data of the enhancement of the total transmitted intensity. The blue squares are the enhancement of the intensity in the area outside the target area and the green diamonds are the enhancement of the total reflected intensity. The red solid curve, blue dash-dot curve and the green dash curve are fits to the experimental data using Eq. 7.5 and the parameters are given in Table 7.1.

experimental data.

Second, the observed increased enhancement might be due to intensity redistributed from the speckles outside the optimized area to speckles inside the optimized area. If this is the case, then the total transmitted intensity would be constant for all optimization radii. A third hypothesis is that there is a redistribution of intensity from the reflected speckles to the transmitted speckles. In this case, the effect of enhancing the transmitted intensity is expected to be noticeable on reflection as a reduction of the reflected intensity. We will also check the validity of these two hypotheses in the next section.

7.3.3 Change of both transmitted and reflected intensities

The enhancements of the total transmitted intensity η_{tr} , total reflected intensity η_{re} and intensity outside the optimization area η_{out} versus the optimization radius are shown in Fig. 7.5. The enhancement of the total transmitted intensity gradually increases from about 1.1 to 2 at the largest radius of $15.2 \mu\text{m}$, which

	B	n
η_{tot}	0.42	0.4
η_{re}	-0.02	0.5
η_{out}	0.25	0.35

Table 7.1: The values of the adjustable parameters B and n obtained by fitting Eq. 7.5 to the experimental data shown in Fig. 7.5 for the total transmission enhancement η_{tot} , total reflection enhancement η_{re} and the enhancement of the intensity outside the target area η_{out} .

is close to the $17 \mu\text{m}$ large size of the detected speckle pattern. There is also an enhancement of the intensity outside the optimization area and it is about $\eta_{\text{out}} \approx 1.5$ at large radii. The enhancements of both the total transmitted intensity and the intensity of area outside the optimization nullifies the second hypothesis. In contrast, the enhancement of the total reflected intensity slowly decreases to $\eta_{\text{re}} \approx 0.93$ as the optimization radius increases. The decrease of the η_{re} is not as rapid as the increase of η_{tot} because the reflected intensity is about nine times higher than the transmitted intensity. Therefore, a large intensity enhancement on transmission corresponds to a small intensity enhancement on reflection.

In order to accurately compare the decrease of η_{re} with the increase of η_{tot} , we need to know the dependence of both terms on the optimization radius. The dependence of the enhancements on the optimization radius is unknown and we find that a power-law fit following

$$\eta = \frac{B}{r_o^n} + 1 \quad (7.5)$$

describes the experimental data. Here, B and n are the adjustable parameters. The fits to the experimental data are shown in Fig. 7.5 and the values of B and n obtained from the fits are shown in Table 7.1. We obtained $n = 0.4$ and $n = 0.5$ for the enhancement of the total transmitted and reflected intensities respectively and these values agree remarkably. In Fig. 7.6, we plot the absolute changes in the transmitted and the reflected intensities after accounting for the attenuation factor of the ND filters. The absolute changes in the transmitted and reflected intensities both collapse to the same curve, fitted with $n = 0.45$. Therefore, the enhancement of the transmitted intensity corresponds to a decrease in the reflected intensity. This validates the third hypothesis that there is a redistribution of intensity from the reflected speckles to transmitted speckles of the scattering medium.

7.3.4 Radial distribution of reflected intensity

At this point, one question arises: how does the reflected intensity spatially change? Is the change global, *i.e.*, the overall reflected intensity decreases uniformly, or local, *i.e.*, the intensity decreases more at the area where the

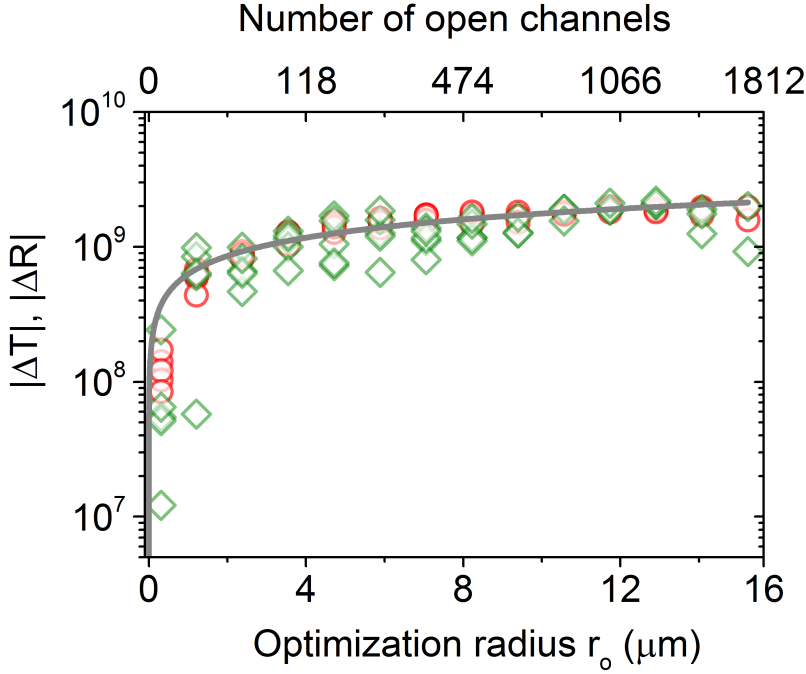


Figure 7.6: Measured absolute change in the total transmitted intensity (ΔT) (red circles) and total reflected intensity (ΔR) (green diamond) versus optimization radius r_o . The solid gray curve is a fit using Eq. 7.5, with $B = 6.3 \times 10^8$ and $n = 0.45$.

transmission is enhanced? As illustrated in Fig. 7.7, a global change of the reflected intensity implies that the redistributed intensity to transmission is deducted equally from all the spatial channels. This is expected if all input spatial channels contribute equally to all the output spatial channels. On the other hand, a local change implies that the intensity of the spatial input channels maps one-to-one with that of the spatial output channels. The local change is expected as a result of the reflection-transmission long-range correlation predicted in Ref. [175].

In order to observe the type of change, we plot the radial distribution of the reflected intensity in Figs. 7.8 (a) and (b) for optimization radii of 15.2 μm and 8.4 μm respectively. In both optimization radii, the optimized (red circles) and unoptimized (blue squares) intensities matches quite well from $r = 0$ to about $r = 5 \mu\text{m}$. The optimized intensity asymmetrically deviates from the unoptimized intensity between $r = 5 \mu\text{m}$ and 10 μm and at $r > 10 \mu\text{m}$, both intensities become equal and decrease in the same way.

For a proper interpretation of these results, we describe the global and local change in the reflected intensity as follows. First, we model the unoptimized

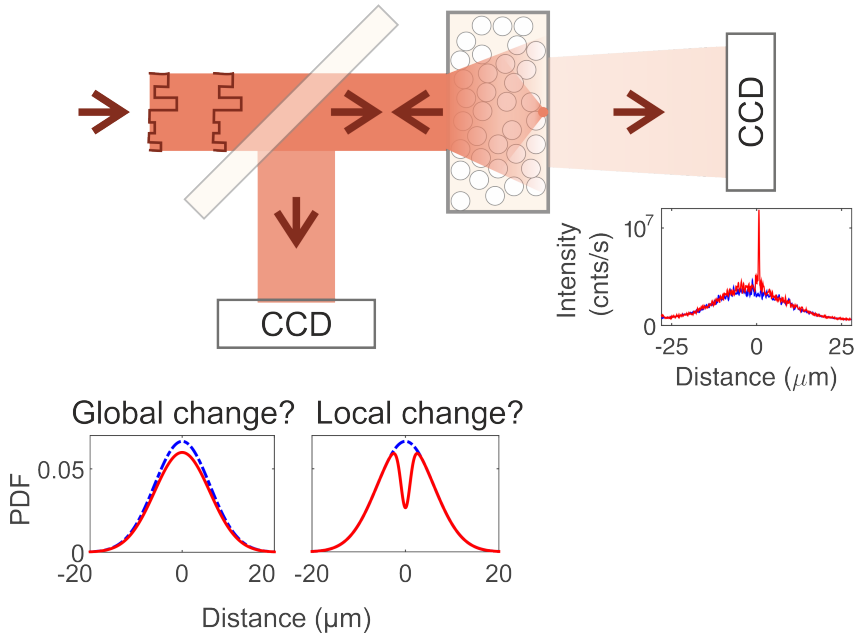


Figure 7.7: (Color online) An illustration of the question about the type of change happens spatially on when the reflected light is decreased to enhance the transmitted light. The wavefront-shaped light is incident on the sample to obtain an optimized focus at the back surface of the sample. The imaged intensity data at the back surface shows an enhanced peak (top inset). The question is how does the reflected intensity spatially re-distribute? A global change (cartoon on the bottom left inset) means that there is a uniform decrease in the amplitude of the Gaussian envelope of the reflected intensity and a local change (cartoon on the bottom right inset) means there is a local dip in the Gaussian envelope.

reflected intensity I_u using a 2D circular Gaussian function equal to

$$I_u(\boldsymbol{\rho}) = \frac{A_1}{2\pi\sigma_1^2} \exp\left(-\left(\frac{|\boldsymbol{\rho} - \boldsymbol{\rho}_0|^2}{2\sigma_1^2}\right)\right). \quad (7.6)$$

Here, A_1 is the amplitude, $\boldsymbol{\rho} = (x, y)$, x and y are the spatial coordinates, $\boldsymbol{\rho}_0 = (x_0, y_0)$, x_0 and y_0 are the coordinates of the center, and σ_1 is the standard deviation that defines the spread of the function. In order to model both the global and local change, we define a second 2D circular Gaussian function I_o

$$I_o(\boldsymbol{\rho}) = \frac{A_2}{2\pi\sigma_2^2} \exp\left(-\left(\frac{|\boldsymbol{\rho} - \boldsymbol{\rho}_0 - \boldsymbol{\rho}_c|^2}{2\sigma_2^2}\right)\right), \quad (7.7)$$

where $\boldsymbol{\rho}_c = (x_c, y_c)$, x_c and y_c are the displacements of the center of I_o from x_0 and y_0 respectively and σ_2 is the standard deviation of I_o . We model the

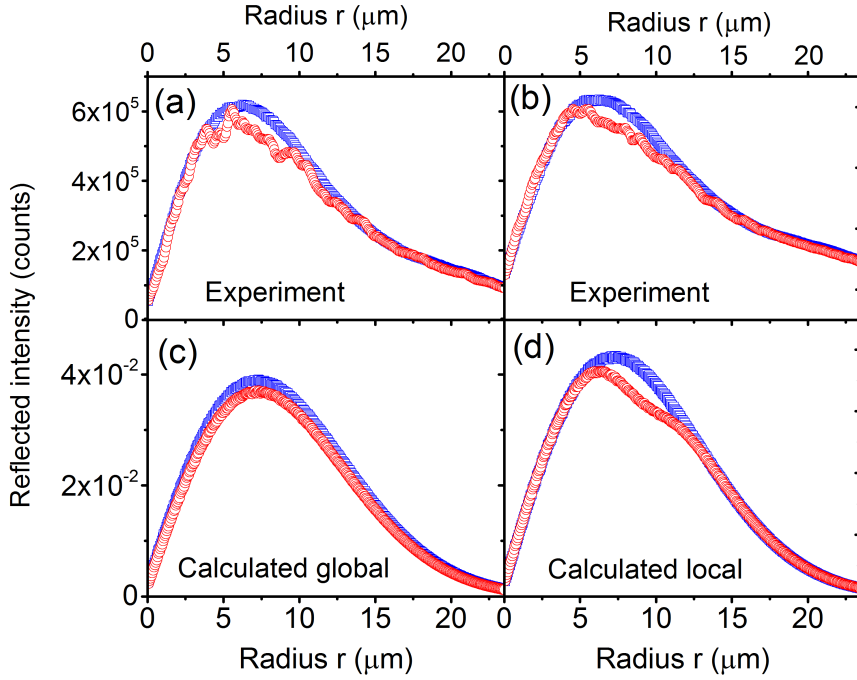


Figure 7.8: Total reflected intensity integral along the circumference of a ring (see inset of Fig. 7.3). The blue squares are the experimental data with unoptimized wavefront displayed on the SLM and the red circles are the experimental data with the optimized wavefront. The optimization radii are 15.2 μm and 8.4 μm in (a) and (b) respectively. The calculated global and local change are plotted in (c) and (d).

optimized intensity I_g , in the case of the global change, as the difference between I_u and I_o , when $\sigma_1 = \sigma_2 = \sigma_g$ and $A_1 > A_2$, to get

$$I_g(\boldsymbol{\rho}) = \frac{A_2}{2\pi\sigma_g^2} \exp\left(-\left(\frac{|\boldsymbol{\rho} - \boldsymbol{\rho}_0 - \boldsymbol{\rho}_c|^2}{2\sigma_g^2}\right)\right) - \frac{A_1}{2\pi\sigma_g^2} \exp\left(-\left(\frac{|\boldsymbol{\rho} - \boldsymbol{\rho}_0|^2}{2\sigma_g^2}\right)\right). \quad (7.8)$$

To model the optimized intensity I_l , in case of a local change, we use the difference between I_u and I_o , when $\sigma_1 > \sigma_2$ and $A_1 > A_2$, to get

$$I_l(\boldsymbol{\rho}) = \frac{A_2}{2\pi\sigma_2^2} \exp\left(-\left(\frac{|\boldsymbol{\rho} - \boldsymbol{\rho}_0 - \boldsymbol{\rho}_c|^2}{2\sigma_2^2}\right)\right) - \frac{A_1}{2\pi\sigma_1^2} \exp\left(-\left(\frac{|\boldsymbol{\rho} - \boldsymbol{\rho}_0|^2}{2\sigma_1^2}\right)\right). \quad (7.9)$$

Using Eqs. 7.6, 7.8 and 7.9, 2D Gaussian functions were calculated for the unoptimized light, and optimized light with either global or local changes respectively. A projection of the generated functions onto the x-axis is shown in the bottom left and right insets of Fig. 7.7. Following the same procedure for the analysis of the experimental data, we obtain the radial distribution of the

calculated functions by integrating the intensity along the circumference of a ring of width of δr and inner radius of r_r .

In Figs. 7.8 (c) and (d), we plot the calculated intensity versus radius for optimized and unoptimized light for both the global and local change. To obtain the curves in Figs. 7.8 (c) and (d), we used these parameters: $\sigma_1 = \sigma_g = 7.5 \mu\text{m}$, as obtained by fitting a Gaussian function to the unoptimized reflected intensity; $A_1 = 1$ and $A_2 = 0.05$. For both global and local changes, the values of A_1 and A_2 are chosen such that the ratio of the total area under the numerical optimized and unoptimized light is 0.95, which corresponds to a comparable enhancement on reflection in Fig. 7.5. x_0 and y_0 were chosen to be both $45.5 \mu\text{m}$, which is exactly at the center of the generated 2D Gaussian function. The adjustable parameters are $\sigma_2 = 2.1 \mu\text{m}$, and $x_c = y_c = 6.4 \mu\text{m}$ and we will comment on these values shortly below. For the global change shown in Figs. 7.8 (c), there is a symmetric deviation of the reflected intensity from the unoptimized light and the deviation is centered at the peak position that is around $7 \mu\text{m}$. At radial positions between $r = 0$ and $r = 5 \mu\text{m}$ and $r = 12 \mu\text{m}$ and $r = 26 \mu\text{m}$, optimized light agrees well with unoptimized light. These features of the calculated global change does not correspond with the features of the experimental data shown in Figs. 7.8 (a) and (b). For the local change shown in Fig. 7.8 (d), there is interestingly an asymmetric deviation of optimized light from unoptimized light between $r = 5 \mu\text{m}$ and $r = 12 \mu\text{m}$. This asymmetric deviation is very similar to what is observed for the two optimization radii in Figs. 7.8 (a) and (b). Our experiment results therefore indicate that there is a local change in the reflected intensity rather than a global change.

We now comment on the values of the adjustable parameters. In the experiment, $x_c \approx 0.5 \mu\text{m}$ and $y_c \approx 2.4 \mu\text{m}$ and we have used $x_c = y_c = 6.4 \mu\text{m}$ in Figs. 7.8 (c) and (d), in order to have a similar asymmetric deviation in the local change. It should be noted that the global change is almost independent of ρ_c , since $|\rho_c| \ll |\rho_0|$ (see Eq. 7.8). The discrepancy between the experiment values of (x_c, y_c) and the adjusted values might be the result of the thermal drift of the SLM, laser, mechanical stage onto which the sample is mounted and other apparatuses. From the experimental and adjusted values of (x_c, y_c) , we estimate that the beam is displaced by approximately $7 \mu\text{m}$, which can happen since the measurements took several days. Consequently, due to the beam displacement of $7 \mu\text{m}$, the optimization area mapped onto the reflection light is almost outside the reflected intensity, which has $\sigma_1 = 7.5 \mu\text{m}$. Therefore, the large optimization radii on the transmission side does not have a significant effect on the reflection side. This explains why $\sigma_2 = 2.1 \mu\text{m}$, rather than $15.7 \mu\text{m}$ and $8.4 \mu\text{m}$, shows in Fig. 7.8 a feature that is comparable to the feature from the experiment. Despite these imperfections in the experiment, our data qualitatively shows that there is a local change in the optimized intensity.

7.4 Summary

We have experimentally shown that as the optimization radius increases, the enhancement of the total transmitted intensity increases, which corresponds to a decrease of the total reflected intensity. We also find that the enhancement of intensity in the optimization area decreases as the radius increases. From the radial intensity distribution of the reflected intensity, we found an indication that there is a local decrease rather than a global decrease in the reflected intensity. The local decrease confirms that the transmitted and reflected intensities are spatially correlated as predicted by Fayard *et al* [175]. Our results have prospects in extending the applications of wavefront shaping to increase the total transmitted intensity through the rough layer on top of the silicon absorber in a solar cell. Our results are also relevant for multi-channel quantum secure communication [68, 176, 177], where enhanced intensities are desired in multiple transmission channels; for transmitting arbitrary intensity distribution through a scattering medium [180]; and imaging through an opaque medium [62, 178, 179].

7.A Comparing wavefront shaping algorithms for large areas optimization

For optimization of intensity in large areas, we investigated three different wavefront shaping algorithms: stepwise sequential algorithm, partitioning and genetic algorithms. The details about how these algorithms work are well described in Refs. [50, 182]. First, the sequential algorithm modulates the phase of the segments of the SLM one by one and combines them at the end of optimization. Second, the partitioning algorithm modulates the phase of 50% of the segments simultaneously and keeps the optimized phases on the SLM. The modulated segments are chosen randomly at each step. A better performance of the partitioning algorithm is expected because a larger number of segments is controlled simultaneously, and this is expected to yield a significant change in the target signal compared to the sequential algorithm. Third, the genetic algorithm begins by creating a population of random phase masks, which are ranked using the measured enhancement. The phase masks are combined using a weight proportional to the enhancement and then further mutated to create new phase masks. The new phase masks are measured and replace the low ranking members of the population. As the whole steps are repeated, the average enhancement of the population increases and finally converges.

We performed wavefront shaping experiments to compare the enhancement of the three algorithms. In Fig. 7.9, we plot the enhancement η_{targ} in the optimization area versus the optimization radius r_o . The enhancement decreases with increasing radius for all algorithms, which is expected. With the partitioning algorithm, the enhancement is on average approximately 85% higher than the sequential algorithm. The partitioning algorithm outperforms the sequential algorithm because of the larger modulation signal in the optimization area. Furthermore, the partitioning algorithm has an enhancement that is 80% higher

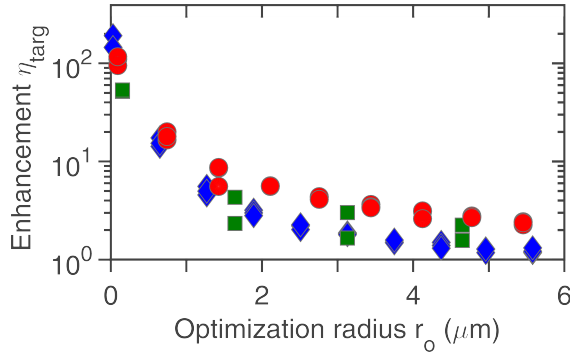


Figure 7.9: Enhancement η_{targ} in optimization target versus the optimization radius r_o for different algorithms. The red circles, green squares, and blue diamond are the data point for the partitioning, genetic and stepwise sequential algorithm algorithms respectively. The partitioning algorithm outperforms the other algorithms.

than the genetic algorithm. For the genetic algorithm, we expected a performance comparable to the partitioning algorithm, since a comparable number of segments is simultaneously controlled in the two algorithms. We attribute the lower performance to the fact that the genetic algorithm requires a large number of experimental parameters, which might differ for different optimization radii. A further detailed study of using genetic algorithm for large areas optimization is needed. We have therefore used the partitioning algorithm, since it shows a better performance than the other two algorithms.

CHAPTER 8

Effect of spatial intensity fluctuations on wavefront shaping

8.1 Introduction

In order to stir the propagation and absorption of light in complex photonic media, wavefront shaping is one of the basic tools [71, 100, 142, 183]. Wavefront shaping involves controlling the incident wavefront on a scattering medium, such that light can be focused anywhere, even deep inside the most strongly scattering materials [49, 86, 87]. The wavefront of light is spatially modulated by a spatial light modulator (SLM) to realise an optimized wavefront. Wavefront shaping has opened up several exciting applications such as non-invasive biomedical imaging [60–62], advanced optics [63–68], and cryptography and secure communication [69, 70].

As with all measurements, wavefront shaping is unavoidably affected by experimental noise, especially because the measurement time is often lengthy: about 30 minutes for 2000 degrees of freedom on the SLM. Previous studies have identified several experimental sources of noise such as laser intensity fluctuations, camera noise, sample drift, unstable environmental conditions, and imperfect phase and amplitude modulations [49, 50, 59, 84, 136, 181, 182]. None of these studies have studied the effect of spatial intensity fluctuations within a laser beam on wavefront shaping. Spatial intensity fluctuations in laser and x-ray beams are well-known to be the result of the presence of higher order transverse laser modes [184–188]. Continuous wave diode-pumped solid state lasers that are commonly used in wavefront shaping are designed to suppress the high order transverse modes by using a longitudinal pumping method, wherein a pump diode laser is longitudinally focused on the active medium [189–191]. The pump beam profile is assumed to be constant inside the active medium. Due to unavoidable diffraction effects, however the beam of the pump laser diverges and higher order transverse modes are excited. These transverse modes are also excited by an instability and asymmetry of the laser resonator, by fluctuations in the power supply, and by a non-uniform temperature distribution in the active medium [192].

In this chapter, we investigate the effect of spatial intensity fluctuation on wavefront shaping. We observe non-uniform temporal intensity fluctuations along the beam profile of a beam of a diode-pumped solid state laser that is commonly used for wavefront shaping. To study the fluctuations, we use a simple and novel

method of correlating the temporal intensity fluctuations across different parts of the beam. We use mode cleaning with a pinhole and a single mode fiber to suppress the spatial intensity fluctuations within the beam. We observe that mode cleaning increases the fidelity of wavefront shaping by over a factor of 2. Our method and result are applicable in experiments, such as wavefront shaping, adaptive optics and interferometry that require sampling of the intensity on the profile of the laser beam.

8.2 Mode cleaning set-ups

The spatial intensity and phase profile of a laser beam fluctuates non-uniformly due to the high order transverse laser modes that appear at different times. Mode cleaners, also referred to as spatial filters, are used to select only the fundamental mode of the laser and they filter the high order modes. Common mode cleaners for low power lasers are a pinhole [193, 194] and a single mode fiber [195, 196], which we both used in our experiments. In Fig. 8.1, we show a scheme for mode

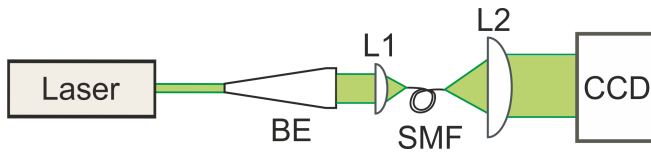


Figure 8.1: Mode cleaning using a single mode fiber. A laser beam is expanded using a beam expander (BE) and focused using a lens (L1) onto the surface of a single mode fiber (SMF). The output of the fiber is collimated using a collimator (L2) and the images of the beam are recorded using a charged coupled device (CCD) camera.

cleaning using a single mode fiber. A laser beam (Cobolt JiveTM, wavelength $\lambda = 561 \mu\text{m}$, beam diameter, $1/e^2 = 0.7 \text{ mm}$) is expanded using a beam expander (BE) with a magnification of $3\times$. With a numerical aperture $\text{NA} = 0.19$, an aspheric lens L1 (Thorlabs C150TME-A, focal length $f = 11 \text{ mm}$) focuses the expanded beam onto the front surface of a single mode fiber SMF (Thorlabs P3-460B-FC-1, $\text{NA} = 0.10 - 0.14$). A fiber collimator (Schäfter and Kirchhoff, 60FC-L-0-M75-26, $f = 75 \text{ mm}$) collimates the output of the fiber to have a beam diameter of 3 mm and a charge coupled device (CCD) camera (AVT Dolphin 145B) takes images of the beam at regular intervals.

The experiment set-up for mode cleaning using a pinhole is shown in Figure 8.2. The beam is focused with a lens L1 ($f = 50 \text{ mm}$) and a pinhole (diameter $= 50 \mu\text{m}$) is placed at the focal plane of the lens L1. At the focal plane, different spatial Fourier components of the original beam are focused at the different points along the Fourier plane. The pinhole in the Fourier plane then filters the off-axis Fourier components of the beam. A second lens L2 (250 mm) is then used to recollimate the beam.

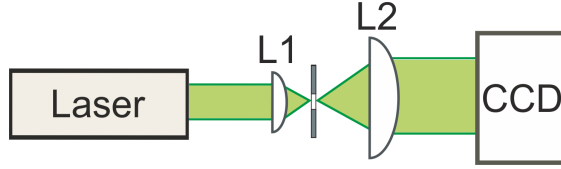


Figure 8.2: Mode cleaning using a pinhole. A laser beam is focused using lens (L1) and a pin hole is positioned at the Fourier plane of lens L1. The pinhole spatially filters the beam. A lens L2 is used to collimate the beam and images of the beam are recorded by a CCD camera.

8.3 Analysis procedure

We took CCD images of the beam over a period of 30 minutes with an interval of 5 seconds and then grouped the camera pixels into macropixels with 400 pixels in each macropixel i . We obtained the intensity averaged over all pixels in each macropixel as I_i and determined the intensity fluctuation $I_{ac,i}(t)$ over time in a macropixel i , as

$$I_{ac,i}(t) = \frac{I_i(t) - \langle I_i(t) \rangle}{\langle I_i(t) \rangle}, \quad (8.1)$$

where $\langle \rangle$ signifies the average over time. Thereafter, we obtained the correlation between the intensity fluctuation $I_{ac,i}(t)$ in each macropixel and the intensity fluctuation $I_{ac,c}(t)$ in a reference macropixel. The normalized correlation C_i of a macropixel i with a reference macropixel c is equal to

$$C_i(\mathbf{r}_i, \mathbf{r}_c) \equiv \frac{\langle I_{ac,c}(t) I_{ac,i}(t) \rangle}{\left(\langle I_{ac,i}(t) I_{ac,i}(t) \rangle \langle I_{ac,c}(t) I_{ac,c}(t) \rangle \right)^{\frac{1}{2}}}, \quad (8.2)$$

$\mathbf{r}_i$ and $\mathbf{r}_c$ are the coordinates of a macropixel i and the reference macropixel c respectively. Calculating $C_i(\mathbf{r}_i, \mathbf{r}_c)$ for all macropixels in the CCD images, we then obtained a 2D spatial correlation function of the beam.

In order to quantify, the fluctuations of the laser power, we use the signal-to-noise ratio (SNR), which is defined as the ratio of the average laser power P_{av} to the standard deviation of the laser power P_{std} ,

$$\text{SNR} \equiv \frac{P_{av}}{P_{std}}. \quad (8.3)$$

8.4 Result

8.4.1 Spatial intensity correlations

In Fig. 8.3(a), we show a CCD image of the expanded beam with a magnification of $5\times$. There are some intensity variations in the beam profile and these are attributed to distortion and aberrations in the magnification optics. The pixels

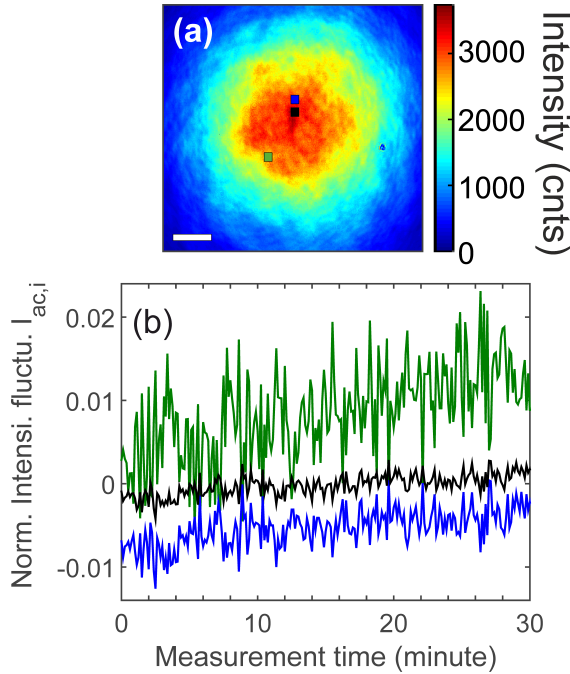


Figure 8.3: (a) CCD image of the intensity profile of the beam. (b) Normalized intensity fluctuation $I_{ac,i}(t)$ of macropixels versus measurement time. The black, blue, and green curves are the intensity fluctuations of macropixels indicated by the black, blue and green squares in (a). The green and blue curves are been shifted by 0.01 and -0.005 respectively. The time interval between the measurements of camera images is 5 seconds. The scale bar is 0.5 mm.

of the images are grouped into macropixels and in Fig 8.3(b), we plot the temporal intensity fluctuations $I_{ac,i}(t)$ in the macropixels indicated with black, blue, and green squares in Fig 8.3(a). The intensity in the black and blue macropixels fluctuates with comparable amplitudes and in phase for most of the times. Interestingly, the intensity fluctuation in the green macropixel has a larger amplitude than the fluctuations in the black and blue macropixels and the fluctuation is out of phase as well. This different intensity fluctuations within the beam is unexpected if the beam is single mode and undesirable for wavefront and spatial intensity sampling experiments.

We now quantify the similarity between the intensity fluctuations in the spatial profile of the beam using correlations. On the CCD image shown in Fig 8.3(a), we choose the macropixel indicated with the black square, which has the highest intensity as the reference macropixel. In Fig. 8.4(a), we show the 2D spatial correlation within the beam, before mode cleaning. There are regions within the beam with different correlation values, ranging from a high correlation close to 1 to low correlation of about 0.2. These different correlation values are because the

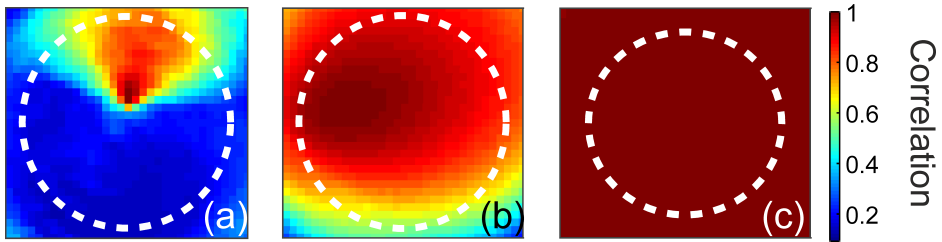


Figure 8.4: Spatial correlation of the beam before mode cleaning (a), after mode cleaning with a pinhole (b), and with a single mode fiber (c). The white dashed circle indicates the $1/e^2$ beam diameter, which is equal to 3.5 mm in (a) and (b) and 3 mm in (c).

temporal intensity fluctuations in some parts of the beam do not correlate with other parts of the beam, as we have shown in Fig. 8.3(b). The non-uniformity in the fluctuation of the different parts of the beam is rather surprising since a uniform spatial correlation is expected for the beam, which is specified to be single mode. Moreover, the non-symmetric correlation within the beam profile is unexpected since the beam is supposedly symmetric. The non-uniform spatial correlation in the beam is undesirable in wavefront shaping experiments and other experiments that spatially samples the intensity or phase in within a beam profile.

We show in Figs. 8.4(b) and (c) the spatial intensity correlation after mode cleaning with a pinhole and a single mode fiber respectively. Mode cleaning significantly improves the spatial correlation in the beam. There is a high correlation in about 90% of the profile beam after mode cleaning using a pinhole. The low correlation in the edges of the beam is attributed to the misalignment of the focusing lens, imperfect circular shape of pinhole, and heating of the metal of the pinhole. For mode cleaning using a single mode fiber, there is a remarkably high and uniform correlation in the whole beam surface and a correlation of 0.98 being the lowest value. A uniform correlation is expected for a single mode beam, which is the output of a single mode fiber. The uniform correlation from the single mode fiber is an improvement for our wavefront shaping experiment because the correlation in the beam fluctuation implies that all the segments on the spatial light modulator will experience the same intensity and phase noise fluctuation and this is very desirable.

8.4.2 Signal-to-noise ratio (SNR) of the laser intensity

We investigate the change in the signal-to-noise ratio SNR before and after mode cleaning. In Figs. 8.5 (a), (b) and (c), we show the SNR before mode cleaning, after mode cleaning with a pinhole, and single mode fiber respectively. Before mode cleaning, the macropixel at the center of the beam has the maximum SNR of 1176, while the SNR at the outer part of the beam is on average about 110,

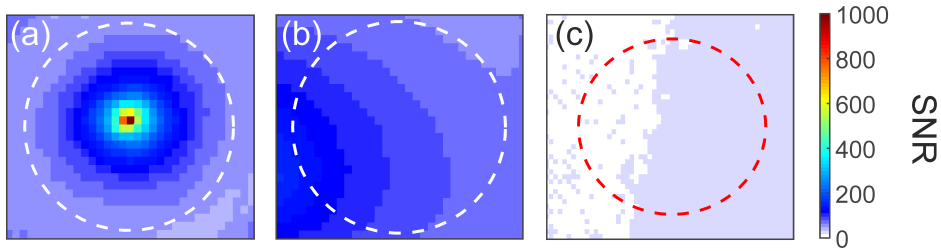


Figure 8.5: Signal-to-noise ratio (SNR) of within the laser beam before mode cleaning (a), after mode cleaning with a pinhole (b), and with a single mode fiber (c). The white and red dashed circles indicate the $1/e^2$ beam diameter, which is equal to 3.5 mm in (a) and (b) and 3 mm in (c).

which is an order of magnitude lower. This large spatial variation of the SNR is attributed to the presence of higher order modes in the beam. Interestingly, after mode cleaning the story changes: the SNR within the beam becomes constant within the whole beam. For mode cleaning with a pinhole, the SNR on the left side of the beam is about 2 times higher than in the other parts and we also attribute this slight non-uniformity of the SNR to the factors already mentioned in Section 8.4.1. As expected for the single mode output of the fiber, the spatial SNR within the beam stays constant at around 15.

It is interesting to note that although the uniformity of the spatial correlation increases, the total SNR within the beam significantly decreases after mode cleaning: a factor of 7 with a pinhole and 50 for a single mode fiber. The reason for this decrease is that the uncleaned beam contains higher order modes that superimpose with the fundamental cavity mode. These unwanted higher modes are spatially filtered out by the pinhole and the single mode fiber. The output of the mode cleaning is therefore a single transversal mode. Since the power of the higher order modes is attenuated, the power fluctuation in the cleaned beam increases and this is the reason for the decrease in the SNR. The decrease in the SNR can be compensated for by normalizing the measured signal by the output of the fiber.

8.5 Effect of mode cleaning on wavefront shaping

Using the experimental set-up described in Section 4.2.1, we performed wavefront shaping experiments before and after mode cleaning with a pinhole, and single mode fiber. To quantify the effect of mode cleaning, we use fidelity ($|\gamma|^2$), which is described in Section 4.2.3. The fidelity quantifies the overlap between the experimentally obtained incident field and the ideal incident field that optimally couples light to the target spot [83,100]. To measure the fidelity of each generated field from wavefront shaping, we exploit the fact that fidelity is well approximated by the ratio of the power in the target spot to the average total transmitted power

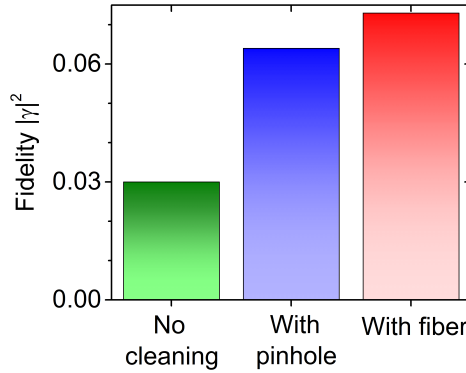


Figure 8.6: The maximum fidelity in wavefront shaping experiments without mode cleaning (MC) (green bar), with mode cleaning using a pinhole (blue bar), and mode cleaning using a single mode fiber (SMF) (red bar). The samples used in the experiments are disordered ZnO samples, which have sample thicknesses $L = 9.1 \pm 3.3 \mu\text{m}$ (green bar), $L = 8.2 \pm 2.0 \mu\text{m}$ (blue bar), and $L = 7.5 \pm 0.5 \mu\text{m}$ (red bar).

for an unoptimized incident field. Here, we report only the maximum fidelities obtained from the experiments, since several factors, (*e.g.*, inhomogeneous sample thickness, experimental noise, and instability in environmental conditions) result into a large fluctuation of the fidelity. In the wavefront shaping experiments, the sample is a disordered ensemble of zinc oxide particles with nearly constant sample thicknesses, as listed in the caption of Fig. 8.6. The fidelity is therefore expected to be comparable, if mode cleaning has a negligible effect. The maximum fidelities reported here are obtained from 20, 5 and 120 wavefront shaping experiments without mode cleaning, mode cleaning with a pinhole, and single mode fiber respectively.

In Fig. 8.6, we show a bar chart comparing the maximum fidelity obtained from wavefront shaping experiments before mode cleaning, after mode cleaning with a pinhole, and single mode fiber. From Fig. 8.6, the fidelity increases from 0.03 to about 0.06 (with a pinhole) and to about 0.07 (with a single mode fiber). It is remarkable to note that the fidelity consistently increases for mode cleaning with both a pinhole and single mode fiber. The fidelity increases after mode cleaning likely because the intensity within the spatial profile of the beam fluctuates coherently after mode cleaning. It is noted that without mode cleaning and a sample with thickness $L = 11.3 \mu\text{m}$, the authors in Ref. [83] obtained a maximum fidelity of about 0.24, which is higher than the maximum fidelity obtained here. The reasons for the lower fidelity are at present unclear and could be a mixture of factors such as fluctuations in environmental conditions, laser noise (a HeNe was used in Ref. [83]), a longer wavelength ($0.63 \mu\text{m}$ compared to $0.56 \mu\text{m}$), which reduces the number of excited sample channels, a higher focusing numerical aperture of 0.95 compared to 0.9 here, and control of two polarizations in Ref. [83], which could increase the fidelity by a factor of two. A systematic

study of the effect of these factors on fidelity is still required including the effect of mode cleaning for comparable number of experiments in all cases. Despite these factors, we observe a consistent increase of the fidelity after mode cleaning with the same wavefront shaping set-up.

8.6 Summary

We observe the presence of non-uniform spatial intensity fluctuation within the beam of a laser. The non-uniform spatial correlations are attributed to the presence of higher order modes and are removed by mode cleaning. We demonstrate that a single mode fiber is more efficient to ensure a uniform spatial correlation than a pinhole. An increase of more than a factor of 2 in the fidelity was observed after mode cleaning. Our result is applicable to experiments that spatially sample intensity or phase profile within a beam such as wavefront shaping, adaptive optics and interferometry.

CHAPTER 9

Summary and outlook

This thesis presents experimental investigations into the propagation of light inside both disordered and ordered complex photonic systems. The experimental results are interpreted using theoretical and numerical models.

One of the main focus of this thesis is to determine experimentally and theoretically the distribution of the energy density inside a scattering medium when the incident wavefront is optimized. As a starting point, we calculated the energy density inside a 2D waveguide with disorder and decomposed the energy density in terms of the eigenfunctions of the diffusion equation. We found that very few eigenfunctions (e.g $M = 1$ for an open transmission channel and $M = 7$ for shaped waves) are remarkably sufficient to reconstruct the energy densities. The fact that the reconstruction can be accomplished with a few number shows that the energy density inside the scattering is similar to the diffusion eigenfunctions. A rigorous theoretical proof of this similarity is, however, still an open challenge at this time. One approach is to derive the energy density as a function of depth based on the principle of conservation of flux and the known total transmission.

We have measured the total stored energy and the depth-dependent energy density inside three-dimensional scattering media. The energy density was probed using fluorescent nanoparticles distributed inside the sample. The experimental data reveal that the total stored energy is enhanced using wavefront shaping in agreement with theoretical prediction and numerical simulations. Our result can be used to control the color of white LED. A white LED has a blue LED that illuminates phosphor particles, which both scatter and absorb blue light and re-emit in the red wavelengths [92]. Optimizing on the blue light will increase the blue content of the white LED as well as the red, since the total energy stored is enhanced. On the other hand, the red content of the white LED can be increase by optimizing on only the red re-emitted.

Having learned a lot about the internal distribution of the energy density, we studied experimentally the intensity changes at the interfaces of a 3D scattering medium as the optimization radius increases. We found that the total transmission increases up to a factor of 2 as the optimization radius increases that is a concomitant of a decrease of the total reflected intensity. A detailed analysis of the spatial distribution of the reflected intensity reveals that there is a local reduction of the intensity rather a global reduction over the entire spatial profile. This observation confirms amazingly the reflection-transmission correlation, which has been recently predicted. In line with the goals of “Stirring” of

light, this result can be applied to increase the intensity transmitted through the scattering layer on a solar cell, thereby increasing the photocurrent [94]. Application of wavefront shaping to a real solar cell involves several practical challenges. One of the challenges is that it is impracticable to realize the complexity of a wavefront shaping set-up on a solar panel. A solution is to have the optimized phase pattern imprinted onto phase masks, which can be deposited as a front layer on the solar cell. A phase mask is also needed for applying wavefront shaping on a white LED. Another challenge is that sunlight is temporally incoherent, which is not an ideal source for wavefront shaping. To avoid this problem, different phase masks can be made for different wavelengths of interest in the spectrum. Thereby, there can be a broadband enhancement of the photocurrent.

Regarding light distribution at an interface, we studied the reflected light exiting the interface of photonic crystals, which have inevitable disorder. The question we asked is whether it is possible to deduce the form of the wave propagation inside the photonic crystal by only measuring the reflected intensity? We model the total field exiting the crystal as a coherent superposition of Bloch waves and spherical waves. We found that the contribution of Bloch waves and scattered spherical waves inside an opal photonic crystal are remarkably comparable. Our approach reveals the form of the waves inside the crystal without perturbing the field. A further investigation (either by numerical simulation or experiment) is still needed to confirm the prediction of the model for different ranges of the ratio of the amplitude of the Bloch waves and spherical waves. An exciting long-time goal is to dope a 3D photonic band gap crystal with intentional point defects (or cavities), as this would be a photonic realization of the Anderson model [9].

As with any experimental method, wavefront shaping is largely influenced by noise [136]. We investigate the effect of uncorrelated spatial intensity fluctuations within the laser beam in our wavefront shaping experiment. The fluctuations are attenuated using spatial mode cleaners: a pinhole and a single mode fiber. We found that the fidelity of optimization depends on these spatial fluctuations and can reduce the fidelity by a factor of 2. Our result therefore shows that a spatial mode cleaner should be applied to a laser beam in a wavefront shaping set-up.

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Nederlandse samenvatting

Dit proefschrift beschrijft onderzoek over het ‘roeren’ en beheersen van de propagatie en absorptie van licht in zowel ongeordende als geordende complexe nanofotonische materialen. Het ‘roeren’ van licht betekent het beheersen van licht met behulp van wavefront shaping en nanostructuren om de lichtabsorptie te verhogen. Dit is erg nuttig voor bijvoorbeeld zonnecellen en witte LEDs.

Een van het hoofddoelen van dit proefschrift is het experimentele en theoretische onderzoek naar de energiedichtheid in een ongeordend medium als het invallende licht wordt geoptimaliseerd. Met behulp van numerieke berekeningen vonden we dat de verdeling van de energiedichtheid in een ongeordend medium kan worden ontleed in termen van de eigenfuncties van de diffusievergelijking. Ook vonden we dat opmerkelijk genoeg slechts een heel klein aantal eigenfuncties (bijvoorbeeld $M = 1$ voor een open transmissiekanaal en $M = 7$ voor geoptimaliseerd licht) voldoende is om de energiedichtheid te reconstrueren. Dat de reconstructie bereikt wordt met weinig eigenfuncties betekent dat de verdeling van de energiedichtheid goed overeenkomt met de diffusie eigenfuncties.

We hebben experimenten uitgevoerd over de totale energieopslag en de diepte-afhankelijke energiedichtheid binnen een ongeordend medium. De energiedichtheid werd probed met behulp van fluorescerende nanodeeltjes die verdeeld waren over het medium. De experimentele data laten zien dat de totale energieopslag in het medium wordt vergroot met geoptimaliseerd licht, is in overeenstemming met de theoretische en numerieke berekeningen.

Nadat we veel hebben geleerd over de interne energiedichtheid in een ongeordend medium, hebben we ons geconcentreerd op de intensiteit aan het oppervlak van het medium. We hebben de optimalisatie radius bij wavefront shaping gevarieerd en de gereflecteerde en uitgezonden intensiteiten gemeten. We vonden dat de uitgezonden intensiteit tweemaal groter is bij de maximale radius terwijl de gereflecteerde intensiteit overeenkomstig afneemt. Interessant is dat de ruimtelijke distributie van de gereflecteerde intensiteit een lokale reductie in plaats van een globale reductie laat zien. Deze waarneming bevestigt de voorspelde reflectie-transmissie correlatie.

Met betrekking tot de licht distributie aan een interface, onderzoeken we het gereflecteerde licht bij fotonische kristallen met wanorde. We willen de vorm van licht propagatie in de fotonische kristallen weten. We modelleren het totale veld gereflecteerd van het kristal als een coherente superpositie van Bloch- en verstrooide golven. We hebben gevonden dat de contributie van Bloch- en verstrooide golven in een opaal fotonisch kristal vergelijkbaar zijn.

Het is welbekend dat in wavefront shaping ruis een belangrijke rol speelt. We hebben daarom het effect onderzocht van ongecorreleerde ruimtelijke intensiteit fluctuaties in een laserbundel bij ons wavefront shaping experiment. De fluctuaties zijn met een pinhole en single-mode fiber gereduceerd. We vonden dat de optimalisatie fidelity van wavefront shaping experimenteel afhankelijk is van deze ruimtelijke fluctuaties en de fidelity kunnen reduceren met een factor van 2. Ons resultaat laat zien dat het essential is om een spatial mode cleaner toe te passen in een laserbundel voor wavefront shaping.

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Femi