

Optimal concentration of light in turbid materials

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In turbid materials it is impossible to concentrate light into a focus with conventional optics. Recently, it has been shown that the intensity on a dye-doped probe inside a turbid material can be enhanced by spatially shaping the wavefront of light before it enters a turbid medium. Here we show that this enhancement is due to concentration of light energy to a spot much smaller than a wavelength. We focus light on a dye-doped probe sphere that is hidden by an opaque layer. The light ($\lambda = 532$ nm) is optimally concentrated to a focal area smaller than $0.037 \mu\text{m}^2$. The focus can be substantially smaller than the used probe. We use a comparison between the emission and excitation intensity to show the light is concentrated to a spot below the resolution of our oil-immersion objective. The results are in good agreement with an optimal concentration of linearly or elliptically polarized light. © 2011 Optical Society of America

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1. INTRODUCTION

In turbid materials such as white paint, biological tissue, and paper, spatial fluctuations in the refractive index cause light to be scattered. Scattering is seen as a huge vexation in classical imaging techniques, where it degrades the resolving power [1]. This decrease in resolution is caused by the fact that light carrying information about the fine spatial details of a structure has to travel further through the medium than the light carrying low spatial frequency information [2]. Because of the importance of imaging inside turbid materials, many researchers are trying to suppress turbidity [3–8].

Although light scattering is detrimental to imaging, it is recently shown that scattering can be exploited to increase the amount of light energy deep inside turbid materials [9]. By spatially shaping the wavefront of the incident light, the emission of a small dye-doped probe sphere hidden inside a turbid layer was strongly enhanced, proving an increase of excitation intensity at the probe position. No information was obtained about the spatial distribution of the excitation light. It was shown experimentally in the related field of time reversal for ultrasound [10] and microwaves [11] that a system completely surrounded by scatterers is able to focus waves down to an optimal spot whose spatial profile is equal to the intensity correlation function [12].

The goal of our work is to experimentally show that spatial wavefront shaping can be used to focus and concentrate light to an optimal small spot *inside* a turbid material. We find that the size of the focal spot can only be understood if the vector nature of light is taken into account.

2. EXPERIMENT

Figure 1 shows the principle of our experiment. (a) Ordinarily a positive lens focuses an incident plane wave to a spot with a size that is limited by the numerical aperture (NA) of the lens. (b) A strongly turbid material behind the lens scatters the light so that no focus is formed. By matching the incident wave-

front to the scattering sample, we force constructive interference at a target position inside the sample. At this position multiple scattered light arrives from all angles, significantly increasing the NA of the system. We observe that the focal size is no longer limited by the original lens, an effect that was also seen in a related experiment where light was focused far behind an opaque sample using direct feedback of the laser intensity [13]. Here the focus is created on a nanosized fluorescent probe sphere hidden *inside* a strongly scattering layer by using the emission from the probe sphere as a feedback signal to synthesize the wavefront.

Our experiments are performed on opaque layers of strongly scattering zinc oxide (ZnO) pigment sprayed on top of a low concentration of dye-doped polystyrene spheres that will act as local intensity probes. We used probe spheres with a radius of $R = 80$ nm and $R = 150$ nm with a fluorescent emission peak at $\lambda = 612$ nm. At the back of the sample, the probes are directly visible, which allows us to monitor the excitation intensity at their position. ZnO is one of the most strongly scattering materials known and shows no fluorescence in the spectral region where our probes emit. The scattering layer ranges between 7.5 ± 1 and $25 \pm 4 \mu\text{m}$ in thickness and has a mean free path of $\ell = 0.7 \pm 0.2 \mu\text{m}$. By measuring

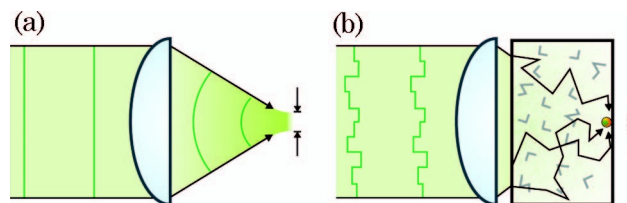


Fig. 1. Principle of the experiment. (a) A positive lens focuses a plane wave to a spot that is limited in size by the NA of the lens. (b) A strongly turbid material behind the lens scatters the light so that no focus is formed. By matching the incident wavefront to the scattering sample, we force constructive interference at a target position inside the sample. The light now arrives from all directions at the target position, significantly increasing the NA of the system.

the angular resolved transmission through the ZnO layers [14], we determined their effective refractive index n_{eff} to be 1.35 ± 0.15 [15,16].

Using a wavefront synthesizer similar to the one discussed in [9], we spatially divide a monochromatic laser beam ($\lambda = 532$ nm) into up to 640 square segments of which we individually control the phase. The shaped beam is focused onto our sample using a microscope objective (NA = 0.95). The same microscope objective is used to capture the fluorescence from a probe hidden under the scattering layer. At the back of the sample, we use an oil-immersion microscope objective (NA = 1.49) to directly image the excitation and emission light at the probe. A digital feedback system that monitors the amount of fluorescence tailors the wavefront to maximize the emission of a probe sphere hidden by the scattering layer using a pixel by pixel optimization method [17] that effectively inverts the transmission matrix [18].

3. RESULTS AND DISCUSSION

In Fig. 2 we see a typical result of the experiment. When we illuminate the sample with a focused plane wave, we measure a low fluorescence response from a $R = 150$ nm probe (a), and we see a speckle pattern in the excitation light (b). Nothing in the speckle pattern reveals the position of the probe. If a shaped wave, created to maximize the fluorescent emission, is focused onto the sample, we measure an intensity enhancement of the emission (c), and we see a sharp focus of excitation light at the position of the probe (d). It is surprising to see that the dimensions of the probe sphere do not confine the focal size, because the created focus is noticeably smaller than the probe.

The radial intensity profile in the focus is shown in Fig. 3 together with the speckle correlation functions (SCFs) measured through both the illumination and the imaging microscope objective. The SCF is defined as

$$C(\Delta x, \Delta y) \equiv \frac{\langle I(x, y) I(x - \Delta x, y - \Delta y) \rangle}{\langle I(x, y) \rangle \langle I(x - \Delta x, y - \Delta y) \rangle} - 1, \quad (1)$$

where $\langle \cdot \rangle$ is a spatial average over all speckles. We fitted the SCFs with a Gaussian function. The SCF is equal to the point spread function of an optical system [19–21] giving the resolution limit of the illumination and imaging optics. The peak intensity of the focus is 32.1 times the average speckle background intensity. The measured intensity profile has a HWHM of 108 ± 5 nm resulting in a focal area of $0.037 \pm 0.003 \mu\text{m}^2$. The size of the created spot is substantially smaller than the measured illumination SCF, because the scattering layer effectively increases the NA. We also see that the measured spot is equal to the SCF of the imaging system, meaning that the formed spot must be smaller than or equal to the resolution limit of our oil-immersion microscope objective.

To further investigate the focus, we compare the intensity enhancements of the emission η^{em} and excitation η^{ex} . For the excitation intensity, the enhancement is defined as the ratio between the peak intensity of the focus and the average diffusive background intensity. The diffusive background intensity is determined by averaging the intensity at the probe sphere over 100 random realizations of the incoming wavefront. The emission intensity enhancement is defined as the total emission for an optimal wavefront divided by the average emission during the reference measurement. In Fig. 4 we have plotted the measured enhancements for (a) $R = 80$ nm and (b) $R = 150$ nm probe spheres. The number of control segments was varied to create a large spread in enhancements without changing the width of the focus [13]. In some measurements the imaging microscope objective might be slightly defocused, introducing asymmetric error bars in the observed excitation enhancement. In Fig. 4 we see that, for the same probe size, the emission enhancements η^{em} are proportional to η^{ex} . From a linear regression to the data points (solid

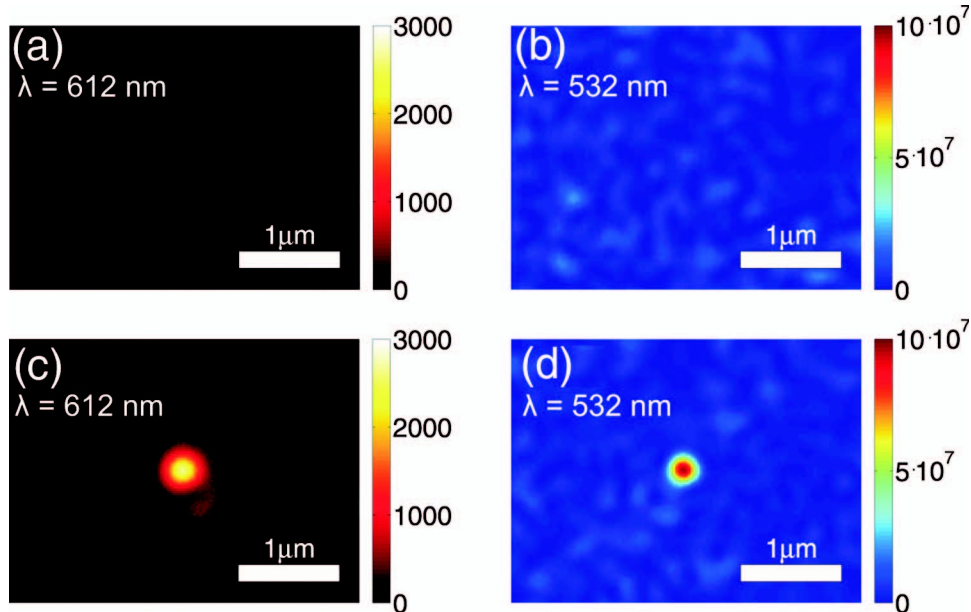


Fig. 2. (a), (c) Fluorescence and (b), (d) excitation images taken at the position of a probe sphere hidden under a layer of ZnO. The probe sphere has a radius of $R = 150$ nm. The images are taken at the back of the sample where the sphere is directly visible. In (a) and (b) we focus a plane wave onto the sample. Scattering creates a random specular pattern of excitation light resulting in a small amount of fluorescent response of the probe sphere. In (c) and (d) we illuminate the sample with a shaped wave that is created to maximize the fluorescent emission. Aside from the fluorescent emission enhancement, we see a sharp focus of excitation light. All intensities are in counts per second.

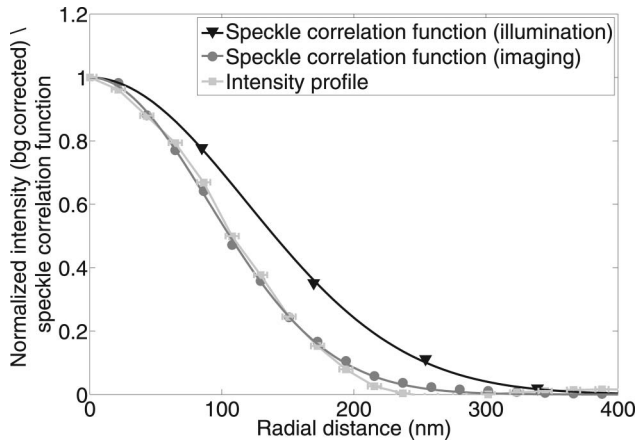


Fig. 3. The measured radial intensity profile of the generated spot (squares), the SCFs of the illumination (NA = 0.95, triangles), and the imaging (NA = 1.49, circles) microscope objectives. We corrected the intensity profile of the focus by subtracting the speckle background (bg corrected) on which it resides.

curves) we extract the proportionality constants $C_{80} = 0.72 \pm 0.07$ and $C_{150} = 0.50 \pm 0.07$ for the $R = 80$ nm and the $R = 150$ nm probe spheres.

We now theoretically investigate the relation between η^{em} and η^{ex} . The emission power caused by a focused excitation field is proportional to the focus intensity integrated over the probe sphere volume. The reference emission power scales with the volume of the probe sphere. On the contrary, the excitation enhancement is independent of the probe volume and is determined by dividing the peak intensity of the focus by the reference speckle intensity. The ratio of the enhancements for a probe sphere with radius R , C_R , is now given by

$$C_R \equiv \frac{\eta^{\text{em}}}{\eta^{\text{ex}}} = \frac{1}{V} \int_0^R \int_0^\pi \int_0^{2\pi} \frac{I(\phi, \theta, r)}{I_{\text{peak}}} r^2 \sin \theta d\phi d\theta dr, \quad (2)$$

where V is the volume of the probe sphere and I/I_{peak} is the position-dependent focus intensity $I(\phi, \theta, r)$ of the excitation light normalized by its peak intensity I_{peak} . We assume that the emission intensity scales linearly with the excitation intensity. From Eq. (2) we see that C_R can be interpreted as the overlap between the normalized intensity of the created focus and the probe sphere.

Based on the observation that the focus can be smaller than the resolution of our microscope objective, we formulate the hypothesis that it has the same profile as a focus created with a high-NA lens with an acceptance angle of 90° . To correctly compute the intensity profile $I(\phi, \theta, r)$, we have to take the vectorial nature of light into account, as a scalar-based approach underestimates the focal volume (if we neglect the vectorial nature of light, the spot size is calculated using a plane-wave decomposition of the contributing wave vectors [22] resulting in the dotted-dashed lines in Fig. 4). The polarization in our focus will be a combination of linear polarizations (in general elliptic), because the feedback in our experiment is provided by the emission of the probe sphere, which has no preference for a specific polarization. By observing the development of the focus in the plane perpendicular to the sample surface we ruled out the possibility that the focus is the result of radially polarized light. As the time integrated overlap of the electric field with the fluorescent sphere is independent of the ellipticity, we are free to choose the polarization as linear in our calculations. Using the focus intensity profile from [23], we find an elliptical focal area of $0.048\pi\lambda^2/n^2$. For our system where the light is optimally concentrated into the center of a polystyrene sphere ($n = 1.60$), this results in a focal area of $0.02 \mu\text{m}^2$ with the HWHM of the two axes 110 and 67 nm. We use the intensity profile of the focus to calculate the overlap integral in Eq. (2). From these calculations we find that $C_{80} = 0.82$ for the 80 nm spheres and $C_{150} = 0.51$ for the 150 nm spheres (dashed lines in Fig. 4) are in good agreement with the experimental values. This data therefore supports our hypothesis and leads to the conclusion that the light is being optimally concentrated.

The area of the generated focus inside the scattering material lies far into the sub- μm^2 regime, yielding exciting opportunities. In biological imaging, for example, selective illumination of fluorescent areas with high resolution is highly desirable. The position of the illuminated area could then be recovered by measuring the diffusive spot of emitted light at the surface of the sample [9]. Several imaging techniques using turbidity have already been demonstrated in the last year [24–26]. These techniques would directly benefit from the generation of a small focus. The efficient light delivery to places inside scattering materials might also be used to study fundamental properties of light transport in both ordered and

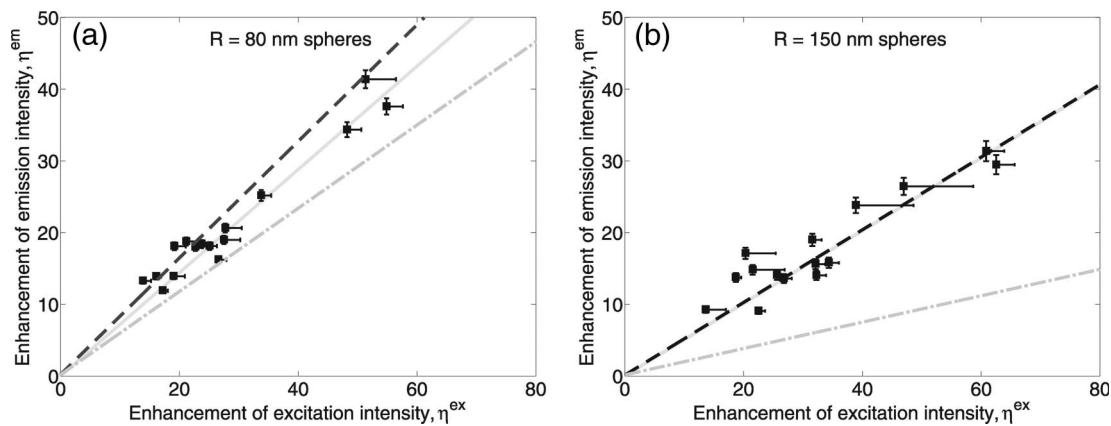


Fig. 4. Measured enhancements of the excitation and emission intensity for spheres with a radius of (a) $R = 80$ nm and (b) $R = 150$ nm. The solid curves indicate the linear regression of the data points. We show the expected regression for light that is optimally concentrated to the center of the probe using a vector theory (dashed curves) and a scalar approximation (dotted-dashed lines).

disordered structures. Furthermore, we foresee important implications of our work for recent results in particle trapping [27] using opaque materials.

Our experimental apparatus is currently not able to detect near-field effects. Expanding the setup with a scanning near-field probe would allow for direct imaging of the focus. Furthermore, such a setup could use measured evanescent waves as a feedback signal to generate the focus. It would be interesting to see if evanescent waves could be coupled to a focus, which are known to influence the spatial correlations in the near field of disordered samples [28–30].

4. CONCLUSIONS

In conclusion, we have focused light onto fluorescent probes hidden by a strongly scattering layer of ZnO. We studied the shape and dimensions of the created focus. We found that the light is optimally concentrated to a focal area of at most $0.037\ \mu\text{m}^2$, which is smaller than some of the probe spheres. A study of the intensity enhancements of both the fluorescence and excitation, performed on different probe sizes, supports the conclusion of optimal light concentration.

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