

Spatial threshold in amplifying random media

Ramy G. S. El-Dardiry,^{1,*} Allard P. Mosk,² and Ad Lagendijk¹

¹FOM-Institute for Atomic and Molecular Physics (AMOLF), Science Park 104, 1098 XG Amsterdam, The Netherlands

²Complex Photonic Systems, MESA+ Institute for Nanotechnology and Department of Science and Technology, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands

*Corresponding author: dardiry@amolf.nl

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We study experimentally as well as numerically the transport and generation of light in multiple scattering media with optical gain. By imaging the spatial distribution of light escaping from the side of the sample, the propagation depth is analyzed. Far below and far above random laser threshold, the spatial profile of emission light is independent of pump intensity, while around threshold, the spatial distribution of emission light changes profoundly. The experimental results are explained by interpreting the numerical solutions to a set of coupled time-dependent diffusion equations on a nonuniform spatial grid. Our studies provide a new and easily accessible method for observing the random laser threshold. © 2010 Optical Society of America

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Understanding the transport of light inside multiple scattering media is of paramount importance for both life scientists [1] and physicists [2]. Recently, the study of light sources inside disordered media has attracted much interest [3,4]. In random lasers, sources, multiple scattering, and optical gain are combined. The majority of experimental random laser studies have focused on spectral analysis [5–7]. The spatial distribution of random laser light inside the scattering medium is largely uncharted territory. New side imaging techniques enabled the determination of important transport parameters in passive random media, such as the transport mean free path and the extrapolation length [8], and were essential in explaining the directionality of lasing in weakly scattering active media [9].

In this Letter, we exploit these new techniques to study the effect of gain on the spatial profile of emission light inside scattering samples. The results are shown to be in quantitative agreement with diffusion theory.

Samples were fabricated by suspending TiO₂ particles (R900, DuPont, median particle size 410 nm, volume fraction 1%) in an acidic solution of 1 mM Rhodamine 640 P in methanol. The experimental cell was a quartz cuvette (Hellma, inner size 10 mm × 10 mm × 45 mm, wall thickness 1.25 mm). Experiments were performed within 1 h after treatment in an ultrasonic bath, before significant sedimentation of TiO₂ particles occurred. The transport mean free path ℓ was determined via enhanced backscattering [10] to be $5 \pm 2.5 \mu\text{m}$ at $\lambda = 600 \text{ nm}$.

In Fig. 1 the experimental apparatus is schematically shown. Excitation light generated by an optical parametric oscillator (Opolette, 20 Hz, 5 ns, $\lambda = 563 \text{ nm}$) is focused onto the sample by a microscope objective (Leitz, 0.25 NA) to a spot with a diffuse fluorescence FWHM diameter of $60 \pm 4 \mu\text{m}$. The emission collected in reflection is spectrally analyzed using a spectrograph (Oriel MS-257, resolution 0.5 nm) connected to an EMCCD camera (Hamamatsu, C-9100). We extended the conventional random laser setup with the sample side imaging technique shown by the lower arm in Fig. 1. An aspherical lens (f -number = 1.5) collects the light exiting the sample from the side and another lens images

the sample surface onto a CCD camera (Mightex). Color filters were used in both detection paths to remove excitation wavelengths. The sample was placed on a translation stage in order to set the distance between the focus spot and the edge of the sample to $30 \mu\text{m}$. The aspherical lens was also placed on a translation stage to ensure the sample surface remained in focus on the CCD camera.

The photoluminescent emission spectrum centered at 610 nm was measured for various values of the pump fluence. Figures 2(a) and 2(b) show the peak spectral radiance and the width of the output spectrum versus the averaged pump power. The peak spectral radiance and the spectral width show a clear threshold around $20 \mu\text{W}$.

The side imaging technique makes it possible to study the propagation of light in active random media directly. Figure 3(a) shows the 50% intensity contour of the spatial profile below ($0.14 \mu\text{W}$) and above threshold ($465 \mu\text{W}$). The spatial profile of the emission light expands around threshold in both the y and z directions. The maximum of the emission profile starts to lie deeper within the sample around threshold.

To explain the experimental results, we model the transport of light by three coupled differential equations [11] in a cylindrical system. The three equations describe the diffusion of laser photon density $W_l(z, r, t)$, pump photon density $W_p(z, r, t)$, and the density of molecules in the upper laser level $n_1(z, r, t)$:

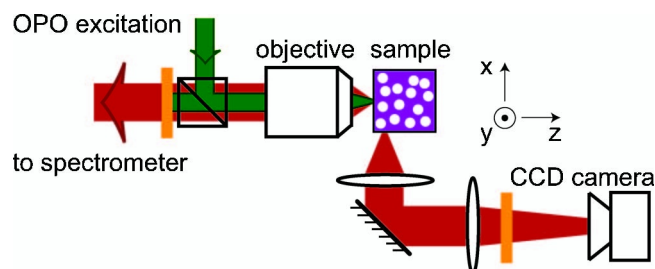


Fig. 1. Schematic top view of the experimental apparatus. Green, pump beam; red, emission light. The orange bars represent color filters.

$$\frac{\partial W_\ell}{\partial t} = D \nabla^2 W_\ell + (\sigma_e c n_1 - \sigma_r c n_0) W_\ell + \frac{\beta}{\tau} n_1, \quad (1)$$

$$\frac{\partial W_p}{\partial t} = D \nabla^2 W_p - \sigma_a c n_0 W_p + \frac{1}{\ell} I_{\text{in}}, \quad (2)$$

$$\frac{\partial n_1}{\partial t} = \sigma_a c n_0 W_p - (\sigma_e c n_1 - \sigma_r c n_0) W_\ell - \frac{1}{\tau} n_1. \quad (3)$$

Here D is the diffusion constant; σ_e , σ_r , and σ_a are the emission, reabsorption, and absorption cross sections of the dye molecules, respectively; τ is the spontaneous emission lifetime; and n_0 is the density of molecules in the ground state. The source term for the pump light, $\ell^{-1} I_{\text{in}}(z, t)$, is a pulse with width τ_p and decays exponentially in z with an extinction length $\ell_{\text{ext}}^{-1} = \ell_a^{-1} + \ell^{-1}$ (with ℓ_a as the absorption length). In our system, scattering is responsible for most of the extinction ($\ell \ll \ell_a$), and hence the source term is treated as power independent. The term β is the spontaneous emission factor and enables us to take into account spectral narrowing without making the equations explicitly spectrally dependent.

Partial differential Eqs. (1) and (2) are converted into ordinary differential equations in time by the method of lines. The set of equations is then solved numerically using MATLAB [12]. For that purpose, a nonuniform cylindrical grid is introduced to allow the calculation of a relatively large system in two dimensions and, at the same time, to have detailed information close to the origin [13]. Defining $h_+ \equiv x_{i+1} - x_i$ and $h_- \equiv x_i - x_{i-1}$, the finite difference form of the first and second derivatives read

$$\frac{\partial f}{\partial x} \rightarrow \frac{f(x+h_+) - f(x-h_-)}{h_+ + h_-}, \quad (4)$$

$$\frac{\partial^2 f}{\partial x^2} \rightarrow \frac{h_- f(x+h_+) - (h_+ + h_-) f(x) + h_+ f(x-h_-)}{0.5 h_+ h_- (h_+ + h_-)}. \quad (5)$$

The two differential equations describing the laser light and the pump light, Eqs. (1) and (2), obey mixed boundary conditions: the photon densities are assumed zero at a characteristic length scale (the extrapolation length) outside the sample. The sample is modeled as a cylinder with dimensions $r = 150\ell$ and $z = 150\ell$. The time taken into consideration spans 40 ns. Realistic experimental situations [11] ($\ell = 5 \mu\text{m}$, $\rho = n_1 + n_0 = 6 \cdot 10^{23} \text{ m}^{-3}$, $\sigma_a = 1.6 \cdot 10^{-20} \text{ m}^2$, $\sigma_r = 0.073 \cdot 10^{-20} \text{ m}^2$, $\sigma_e = 4 \cdot 10^{-20} \text{ m}^2$, $\beta = 0.07$, $\tau_p = 5 \text{ ns}$) were calculated on a $40 \times 50 \times 60$ ($t \times r \times z$) grid.

In Fig. 3(b) we show the 50% contour of the numerically determined time-integrated diffusive emission photon density profile below and above threshold in the r - z plane. The profile above threshold is expanded compared to the profile below threshold. We compare experiment and theory by analyzing cuts from the numerical data at $r = 30 \mu\text{m}$ in the r - z plane. In Fig. 3(c), the values for z at which the intensity profile reaches its maximum and the width of the intensity profile are plotted versus the pump power for both the numerical

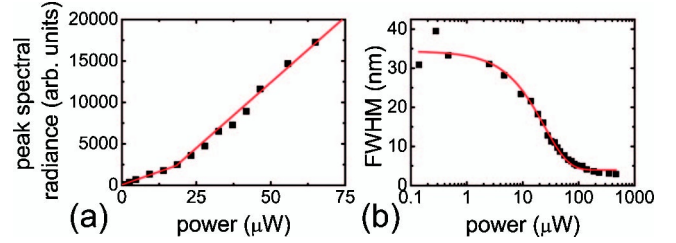


Fig. 2. (a) Peak spectral radiance versus the pump power. (b) Spectral width versus the pump power on a semilog scale. The red solid curve is a sigmoidal fit and serves as a guide to the eye.

calculation and the experiment. In agreement with experimental observations, the spatial width of the laser light profile increases around threshold but remains constant far above threshold. The inclusion of reabsorption is important in order to make the results quantitatively correct.

To understand why the spatial profile is fixed far below and far above threshold, we analyze the contribution of spontaneous and stimulated emission to the total emission in time. Figure 4 shows calculated contours of the population of the upper laser level for pump powers below and above threshold in the z - t plane. Far below threshold, the origin of the emitted light is spontaneous emission. In this regime, the set of equations is linear; an increase in the pump fluence simply leads to more spontaneous emission events everywhere in the sample, but does not change the normalized spatiotemporal profile of the excited molecules.

Above threshold, stimulated emission is responsible for most ($>99\%$ for $P/P_{\text{th}} > 1$, where P_{th} is the pump power threshold) of the light emitted by the dye molecules. Gain compensates for the losses in the system, but the gain can never, except for temporal fluctuations, actually exceed the losses. The main losses in a random laser are due to the diffusive loss term, $\nabla^2 W_\ell$, which is

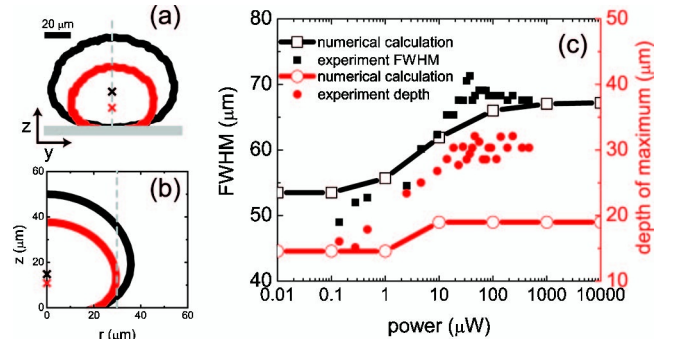


Fig. 3. (a) Measured 50% intensity contour of emission light exiting the sample from the side below (red) and above threshold (black) in the y - z plane. Gray line, boundary of the sample; crosses, the maxima of the profiles. (b) Time-integrated calculated 50% intensity contour of emission light in the r - z plane below (red) and above threshold (black); crosses, maxima of the profiles. (c) Theoretical and experimental characteristics of the spatial profiles along the dashed curves in (a) and (b) versus pump power. Black open squares ($\square$), numerically calculated values of the FWHM of the cut of the spatial distribution of laser light; black solid squares ($\blacksquare$), experimental values from the profile cut; red open circles ($\circ$), numerically determined position of the laser light maximum in depth; red solid circles ($\bullet$), experimental values.

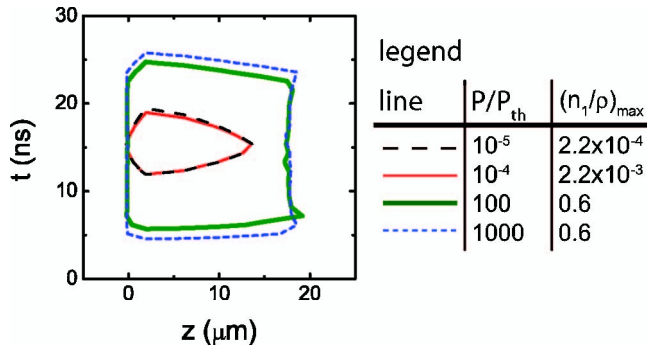


Fig. 4. Calculated 50% contours of the excited state profile in the z - t plane for different pump powers far below and far above threshold.

only nonzero close to the origin. As a consequence, the gain length in a random laser is strongly spatially dependent. Once above threshold, the stronger the system is pumped, the deeper inside the sample reabsorption losses are compensated for. The cancellation of reabsorption hardly changes the diffusive laser light profile, since the required gain is small compared to the gain needed to compensate the diffusive losses. Analogous to conventional laser systems the gain saturates above threshold [14], the maximum density of excited molecules, n_1/ρ , is fixed for all pump powers above threshold. Furthermore, for higher pump powers the time the population inversion lasts becomes longer. The expansion of the spatial laser light profile around threshold thus visualizes the transition from spontaneous emission to stimulated emission as the dominant mechanism of radiation.

The numerical calculation succeeds in explaining the main experimental features. Yet Fig. 3(c) shows that the changes in the numerical data are smaller than in the experiment. We attribute these differences to two limitations in the model. First, the system is modeled in cylindrical coordinates, whereas, in the experiment, the sample configuration is not radially symmetric. Taking all boundaries into account properly would imply a three-dimensional model, which is too computationally intensive. Second, population of the dye triplet state is ignored in our model. Accumulation of molecules in the triplet state reduces the density of molecules available for absorption of pump light, leading to an extra shift of the emission maximum not considered by the model.

In conclusion, we have studied the transport and generation of light originating from sources inside a multiple scattering medium. Our side imaging method is a novel experimental way to determine and study the random laser threshold. Around the random laser threshold, the spatial distribution of emission light expands. Far below threshold, each pump photon increases the number of

excited molecules following a pump power independent spatial profile. Far above threshold, each pump photon eventually results in a stimulated emission event following again a pump power independent spatial profile. Our analysis emphasizes that the gain length in a diffusive system is a spatially dependent variable [15]. Moreover, above threshold, the minimal gain length is determined by the diffusive loss inside the system and is not easily tuned by the excitation power. Our observations relate to random laser systems in which spectral spikes are observed. The expansion of the lasing volume rather than a decrease in gain length is an important factor not yet taken into account for explaining the observation of an increasing number of spikes with increasing pump power [16].

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