Amplifying volume in scattering media

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We investigate the influence of the excitation spot diameter on the laser threshold of a scattering amplifying medium. Fluorescence spectra are recorded from a suspension of TiO_2 scatterers in Sulforhodamine B dye. The threshold pump intensity becomes larger by a factor of 70 if the excitation beam diameter gets close to the mean free path l. This increase is explained by use of a simple model describing diffusion out of the amplifying volume and is confirmed by a Monte Carlo simulation.

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Disordered systems that both scatter and amplify light have proved to be an interesting subject of study. Since these systems were rediscovered four years ago,[1,2] many reports about these so-called random lasers have been published. There have been substantial efforts from the experimental as well as the theoretical side to unravel the mechanism that gives rise to the surprising behavior of these systems. Intensively studied features have included the occurrence of a threshold in the output power and spectral width[2] and a marked shortening of the output pulse on crossing this threshold.[3] These are characteristics that are common to ordinary lasers but are not usually observed in random systems, in which the light propagates diffusively.

We present experiments that demonstrate that the threshold of the system depends on the size of the pumped volume. The transport of light in a scattering medium depends strongly on the length scales of the system. The relevant scales are the transport mean free path l, the distance after which the propagation direction of light is randomized, and the sample thickness L. If $L \leq l$, the transport of light is not diffusive, and only single scattering may be important.[2,4] If $L \gg l$, however, the propagation of light in the system cannot be explained without a role for multiple-scattering processes.

The threshold of a laser depends on the balance between gain and loss of light in the system. The total amount of gain is the product of the amplification per unit path length and the path length traveled through the amplifying medium.[5] In a random laser the feedback (conventionally provided by mirrors) is supplied by multiple scattering. Incomplete feedback gives rise to loss. The spatial distribution of gain is governed by the spreading of pump light in the system. The path length involved in the gain analysis is the length of diffusive paths through the gain volume. The gain volume is cylindrical; the diameter is set by the diameter of the excitation spot. The thickness d is related to the diffuse absorption length $L_a = \sqrt{ll_a/3}$. In the absence of saturation, $d = L_a$; otherwise (as in this study) $d > L_a$. Owing to multiple scattering, the loss as well as the amplified path lengths, and hence the lasing threshold, depends on the gain volume.[6]

We have recorded fluorescence spectra from a TiO_2-dye suspension as a function of excitation spot diameter and pump-pulse energy. The scatterers have a diameter of 220 ± 70 nm. They are suspended in 1.0 mM [40 parts in 10^6 (ppm)] of Sulforhodamine B dye dissolved in methanol. The TiO_2 volume fraction is 1×10^{-3}, resulting in a transport mean free path $l = 100 \pm 20$ μm; l was obtained from coherent backscattering, measured without the pump beam. The inelastic length of the dye solution is calculated to be $l_a = 100$ μm. The thickness of the sample cell is $L = 1$ cm. During the experiments the suspension is stirred continuously to prevent sedimentation.

The pump source is a Spectra-Physics DCR-2A frequency-doubled Q-switched Nd:YAG laser that produces 9-mJ pulses with a pulse duration of 6 ns at a repetition rate of 20 Hz. The pulses are attenuated by use of a pair of Glan prisms, one of which we can rotate to vary the pump-pulse energy from 0 to 9 mJ. The pump beam is incident upon the sample through a lens ($f = 6$ cm) that sits upon on a translation stage. By movement of the lens along the pump beam the diameter (beam waist at $1/e^2$) of the spot on the sample cell is varied from 80 μm (in the focus) to 2 mm. The excitation and detection directions are at an angle of $\approx 20°$. The fluorescence is imaged upon the entrance slit of a $f = 250$ mm single-stage grating spectrometer. The spectrum is recorded with a Princeton Instruments 1412 diode array.

In Fig. 1, three spectra are shown. Curve A is the fluorescence spectrum obtained from the sample when it is pumped with an excitation pulse energy of 4 μJ. Curve A resembles the emission of the neat dye solution, shown as spectrum C for reference. The difference between A and C is caused by reabsorption of the emitted light by unexcited dye molecules.[7] Curve B is the fluorescence spectrum of the same suspension, pumped with a pulse energy of 0.1 mJ. When the excitation pulse energy is increased, the spectrum narrows strongly. The spectra are obtained by use of a large pumped spot of 2-mm diameter.

In Fig. 2 the FWHM of the spectra is plotted versus the pump-pulse intensity. There is a difference in FWHM below threshold for the two curves shown

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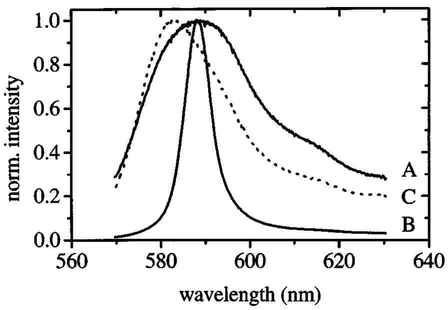

Fig. 1. Fluorescence of the TiO_2-dye suspension at A, 4 μJ mm^{-2} and B, 0.1 mJ mm^{-2} pump-pulse energy compared with C, the fluorescence of the neat dye solution.

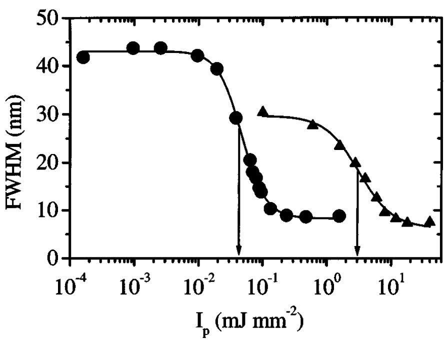

Fig. 2. Linewidth versus excitation pulse intensity for (●) 2-mm and (▲) 80-μm pump diameters. The curves are fits to the data. The threshold of each curve, indicated by an arrow, depends on spot size.

in Fig. 2, owing to a small difference in spectral shape. The change of FWHM within one series can, however, be reliably used to determine the threshold. From these data we define the laser threshold as the inflection point of a sigmoidal fit through the data points. This procedure is repeated for a number of different excitation spot sizes. When the pump-beam diameter is decreased, the threshold intensity shifts to values that are 70 times higher than those measured with the largest spot sizes. The diameter of the pump beam strongly influences the threshold intensity, as is evident from Fig. 3. A consequence of this result is that an experiment on random lasers will yield different results depending on whether a focused pump beam or plane-wave excitation is used. Note that we vary the beam size independently of l. In earlier work,[2,3] l was varied and (occasionally) $l > L$, so necessarily l was also greater than the pump-beam size. A change in the properties of the system must then be attributed to the strongly differing transport in the diffusive ($l \ll L$) and single-scattering regimes.

We asserted above that a change in the threshold pump intensity owing to a change in excitation spot size would be a multiple-scattering effect. To verify this claim we repeated the experiment on a neat dye solution and on a suspension with $l \approx 2$ cm ($>L$). We found that there is no clear threshold in either situation, unless the fluorescence is collected from the direction of amplified spontaneous emission. The FWHM of the fluorescence spectrum depends on the excitation intensity but not on the excitation spot size. Neither does the amplified spontaneous emission threshold for these transparent systems depend on spot size. We conclude that the relation between pump-beam size and threshold is due to multiple scattering.

The increase of the threshold for small excitation regions is explained in the following way: Light is emitted in the pumped region of the sample, from where it starts to diffuse. If the pumped area is large, the amplifying volume is large. Light that is emitted in the pumped volume can travel a long path inside the part of the system that has gain; the light is amplified strongly. If a path reaches the passive (unexcited) part of the system, there is a large probability that it will return to the amplifying region because of the large pumped area. For a small excitation beam diameter the light paths will very probably leave the amplifying region after a short time, with a small chance of returning. That this is so means that one needs a larger gain to compensate for losses, i.e., the threshold is higher.

To check this explanation quantitatively we performed a Monte Carlo simulation of random walks in the geometry,[8] as depicted in Fig. 4. The left-hand side shows a schematic of the system. The vertical line is the sample interface, and the pump light is incident

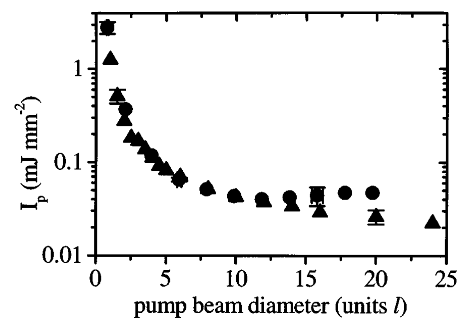

Fig. 3. Threshold excitation intensity versus pump-beam diameter in units of $l = 100$ μm from (●) the experiment and (▲) the simulation. The simulation data are multiplied by 2.

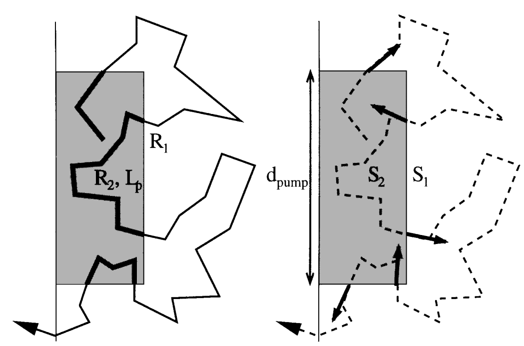

Fig. 4. Left, model geometry of the system: the box is the amplifying volume, and the wiggly line represents a path. Right, the implementation of this path that was used in the simulation. The return probabilities R_1 and R_2 are calculated for S_1 (outside the gain volume) and S_2 (inside). For S_2 we also evaluate the path length L_p in the gain volume.

from the left-hand side (not shown). The shaded box is the pumped volume. The wiggly line is a light path, which is amplified if it is the gain volume (thick line segments).

For the simulation a path is cut at the edges of the amplifying volume. This procedure results in two sets of paths: one set, S_1, traveling out of the gain volume and one set, S_2, traveling inward (right-hand side of Fig. 4). From Fig. 4 one can see that it is beneficial for the amount of amplification per path to return to the boundary of the excited volume: A path in S_1 returning to the pumped volume will be amplified further; a path in S_2 returning to the boundary remains in the sample (i.e., it is not lost). We calculate the probability of returning to the amplifying or the passive boundary (R_1 and R_2, respectively) for the sets of paths S_1 and S_2, respectively.[8] These probabilities can be compared with the reflectivities of cavity mirrors in an ordinary laser geometry. For paths in S_2 the traveled path length L_p is counted. We calculate the threshold gain γ_{th}, which is the amplification per unit path length needed to compensate for cavity losses: $\exp(\gamma_{\mathrm{th}} L_p)\, R_1 R_2 = 1$. Our simulation is performed for a series of different radii of the gain volume from $0.5l$ to $100l$, resulting in R_1, R_2, and L_p values for each radius. The depth is kept fixed at $d = 3l$, a number estimated from the amount of pump photons per pulse injected into the sample and the resulting saturation. For small beam radii this results in a rod-shaped volume. Different shapes give similar results; the most influential parameter is the ratio of gain volume to boundary area.

To compare the experiment and the simulation we must convert γ_{th} obtained from R_1, R_2, and L_p to a threshold pump intensity. The threshold gain is inserted into a set of laser rate equations,[5] where only cavity losses are considered. We analyze the system in steady state for simplicity, corresponding to a cw situation. Earlier simulations[6] showed that a quasi-steady state develops within 100 ps. The pump pulse in our experiments is much longer, so this is a valid approximation. Isolating the pump intensity I_p in terms of γ_{th} results in the relation $I_p = K(\gamma_{\mathrm{th}}/\gamma_0 - \gamma_{\mathrm{th}})$. Here γ_0 is the maximum attainable gain, $n\sigma_{\mathrm{em}} = n \times \mathrm{QE} \times \sigma_{\mathrm{abs}}(\mathrm{max})$. n is the number density of dye molecules. The quantum efficiency[9] (QE) of Sulforhodamine B is ≈ 0.7, and, using $\sigma_{\mathrm{abs}}(\mathrm{max}) = 4.2 \times 10^{-16}\ \mathrm{cm}^2$, we find that $\gamma_0 = 77\ \mathrm{cm}^{-1} = 0.77 l^{-1}$. The constant $K = \eta\hbar\omega_p/(\tau\sigma_{\mathrm{abs}})$. The excited-state lifetime of the dye in the TiO_2 suspension is measured to be $\tau = 3.2$ ns, $\hbar\omega_p = 2.33$ eV is the pump photon energy, and $\eta = 1.32$ is the refractive index of methanol. The absorption cross section for pump light $\sigma_{\mathrm{abs}}(532\ \mathrm{nm}) = 1.6 \times 10^{-16}\ \mathrm{cm}^2$. The result is shown in Fig. 3.

Comparing the results from the experiment and the simulation, we see that there is excellent agreement in the behavior of the threshold pump intensity as a function of the excitation spot diameter. The upturn at small diameters that is observed in the experiment is correctly reproduced by the simulation. Intensities from the experiment are approximately a factor of 2 larger than those from the calculation; the difference is caused by the simplicity of our model. All losses except those that are due to incomplete feedback are disregarded. We assume that all incident pump light is absorbed; scattering of the pump light out of the system is not taken into account. The threshold intensities resulting from the simulation are monotonically decreasing. The experimental threshold of I_p, however, levels off for the largest beam diameters. This leveling off is attributed to reabsorption loss, which is important for long paths in the passive part of the system.

We have demonstrated that the excitation intensity needed to drive a scattering gain medium to its threshold depends strongly on the beam diameter of the pump. This effect is due to multiple scattering; it is not observed in the absence of scatterers. Light that propagates diffusively through the medium starts from the excited volume and is amplified less strongly if the amplifying region of the sample has a small diameter, of the order of $5l$, giving rise to a threshold that is 70 times higher than if the gain volume is large. The experimental data are accurately reproduced by a random-walk simulation.

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