

LASER ACTION IN CONDENSED DISORDERED MEDIA OF ACTIVE DYE-STAINED ANIMAL TISSUES AND SANDY COLLOIDAL SCATTERING WALLS

Masood Siddique, Quan Zhen Wang, and R. R. Alfano[†]

[†]The City College and The Graduate School of The City University of New York, Institute for Ultrafast Spectroscopy and Lasers, New York State Center for Advanced Technology for Ultrafast Photonic Materials and Applications, Dept. of Electrical Engineering and Department of Physics, New York, New York 10031

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ABSTRACT

Significant narrowing of both the spectral and temporal profiles of emission radiation from optically pumped dyes was observed in discrete and continuously disordered media such as dilute colloidal dye solutions and densely packed forms of sandy powders and animal tissue treated with rhodamine 640 dye solution. The narrowing of the spectral and temporal response is attributed to laser action arising from feedback of the emission radiation from the surrounding scattering walls into the photoexcited dye regions of the animal and sandy colloidal disordered media. © 1996 Society of Photo-Optical Instrumentation Engineers.

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1 INTRODUCTION

Laser amplification occurs when stimulated gain overcomes the losses in an excited medium.¹ In these systems, scattering is generally kept to a minimum since it contributes to the loss of the pump and emission radiation, and adds to the destruction of the spatial coherence of the laser action. It is well known that scattering increases the laser threshold, where the gain overcomes the losses. Laser action in highly scattering media in nonresonating cavities was considered theoretically by Letokhov in the 1960s² and demonstrated experimentally in the 1980s by Markushev et al.³⁻⁸ In these experiments, Markushev et al. produced stimulated emission in optically pumped powdered laser crystallites of grain sizes ranging from approximately 1 to 200 μm . Most recently, laser action was observed in optically excited colloids of discretely scattering dielectric particle suspensions in dye solutions.⁹⁻¹¹ Under certain conditions, the excitation threshold for the generation of laser action in colloidal dye solutions was lowered as the density of scatterers was increased. The mechanisms of laser action in colloidal dye solutions, reviewed by Genack et al.,¹² were inconclusive at that time. The questions arise as to what the mechanisms of this effect are and whether such laser action would take

place in continuously random scattering media such as biomedical tissues and disordered solids treated with dyes.

This paper reports on a systematic study of laser action in discretely disordered condensed media and continuously disordered animal tissues in an optically active dye host using temporal and spectral measurements to support a feedback mechanism from the surrounding scattering walls of the host medium.

2 EXPERIMENTAL METHOD

Rhodamine 640 perchlorate dye in methanol solution of 10^{-3} M concentration was used as the optically active medium in all measurements. Three classes of disordered host media were prepared: (1) discretely disordered dilute colloidal solutions, (2) densely packed powders, and (3) continuously disordered animal tissues. The discretely scattering dilute colloidal samples were made by adding TiO_2 particles coated with Al_2O_3 (average diameter 0.25 μm) to the active dye host solution in varying densities. The wet, densely packed powders in the active dye host were prepared by adding powdered Al_2O_3 (5, 0.3, and 0.03 μm diameter) and TiO_2 (0.25 μm diameter), in a cuvette of the host dye and allowing them to settle until maximum packing density was reached at the bottom. Two kinds of these

Address all correspondence to R. R. Alfano. E-mail: alfano@scisun.sci.cny.cuny.edu

sandy densely packed powder samples were prepared: (1) wet powders which were obtained by adding the powders to the dye host in a cuvette and (2) a dried paste, which was obtained by mixing the powders with dye solution and allowing them to dry. The animal tissues were chicken tissue and pig fat, sliced into 1×1-cm pieces and stained with the active dye solution. All of the above samples were placed in a 1×1×3-cm glass cuvette and excited from one end, except for the animal tissues, which were excited directly from the exposed top of the cuvettes.

For temporal measurements, the sample was excited by a 10-ps pulse at 527 nm, obtained from a frequency-doubled single shot Nd-glass laser. The pump pulse intensity was varied from 450 μJ to about 1 μJ by inserting neutral density filters in the path of the excitation pulse. The pulse was focused to a spot size of 0.5 mm onto the slightly off-axis (approximately 5 deg) sample surface. The portion of the pulse transmitted through the beam splitter was collected by an energy meter to monitor the pulse energy. The backscattered light from the front surface of the sample surface was collimated and the pump pulse then filtered out by longpass filters to cut off wavelengths shorter than 540 nm. The collimated light was then focused onto the slit of the streak camera along with a small portion of pump pulse (reference pulse) timed to arrive about 50 ps earlier than the signal light. The streak camera was a Hamamatsu C979 with a SIT tube (C1000) and a temporal analyzer to measure and record the signal with a temporal resolution of about 10 ps.

For spectral measurements, a frequency-doubled Nd-Yag laser providing 2-ns pulses at 532-nm radiation was focused on the biomedical sample surface to a spot size of 2 mm diameter to optically excite the samples. The radiation emitted in the backscattered direction was collected and focused onto a spectrometer after the exciting radiation had been filtered out by two longpass filters.

The absorption length for the neat dye solutions at $10^{-3}M$ concentration was measured to be 140 μm . The transport mean-free-paths at the excitation wavelengths of the dilute colloidal dye solutions of titania particles, calculated using Mie theory, varied from approximately 1800 μm for a particle density of $5 \times 10^9 \text{ cm}^{-3}$ to 18 μm for $5 \times 10^{11} \text{ cm}^{-3}$ density. The volume content of titania in these solutions did not exceed 0.5%. The time-resolved stimulated emission from these colloidal dye solutions was measured in a wide range of samples for: $l_{tr} \gg l_a, d$ to $l_{tr} \ll l_a, d$ domains, where l_{tr} is the transport mean-free-path length, l_a is the absorption length of the neat dye solution (both at the excitation wavelength), and d is the diameter of the spot size of the input pulse. The transport mean-free-paths in the case of densely packed powders are about 4 μm in the case of TiO_2 powders and approximately 2.5 mm for chicken tissue and 0.35 mm for fat.

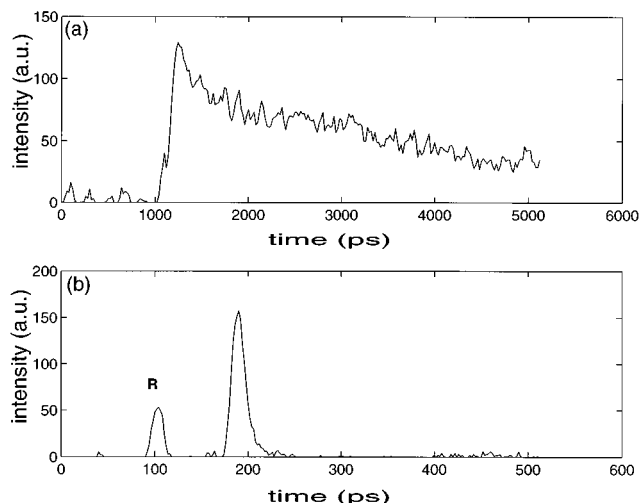


Fig. 1 Time-resolved profile of the emission radiation from optically excited active colloidal solution of rhodamine 640 in methanol solution at $10^{-3}M$ concentration with TiO_2 particles. (a) Spontaneous emission from neat dye by an excitation pulse energy of 18 μJ and (b) emission of a short pulse by excitation pulse energy of 16 μJ in the presence of a scatterer density of 10^{11} cm^{-3} . R is the input reference pulse. The amplitudes of the reference and signal profiles are not to scale.

3 RESULTS

Temporal measurements were conducted to observe the dependence of emission profile on the input excitation intensity. The lasing threshold was reached when the emission lifetime shortened from the typical spontaneous lifetime, on the order of nanoseconds, to the picosecond domain.

For dilute colloidal solutions with dye concentration of $10^{-3}M$, the introduction of scatterers decreased the threshold energy compared with the neat dye solution. The time-resolved emission profiles from excited neat dye and scattering solutions are presented in Fig. 1. The measured time-resolved emission, when the lasing threshold had been reached, was of the same duration as the input pulse of 16 ps duration (10 ps emission duration convoluted with 10 ps response time of the streak camera). What makes this type of laser action peculiar is the fact that the short pulse emission was observed for input energies much lower than the threshold energy for the corresponding neat dye. The threshold energies for colloidal dye solution of varying l_{tr} are summarized in Table 1.

All samples of active densely packed wet powders exhibited stimulated emission as indicated by their ultrafast temporal profiles. The temporal profiles of stimulated emission in TiO_2 powder in dye solution are displayed in Fig. 2. Samples with smaller grain sizes of Al_2O_3 (0.03 μm) had higher thresholds than those with larger grain sizes. In the dried-paste form, an ultrafast time-resolved profile of the emissions could only be obtained from the alumina powders of 5 and 0.3 μm at a lowered

Table 1 Comparison of input excitation thresholds for TiO₂ particles in rhodamine 640 dye solution (10⁻³ M) in methanol.^a

I_{ire} (μ)	Threshold energy (μ J) 10 ⁻³ M; $l_0=140 \mu$	$R_{eff} \sim l - (I_{ire}/L)$
Neat dye	45	0.00
1400	35	0.86
700	25	0.93
140	17	0.986
70	15	0.993
14	13	0.9986

^a The particle density was varied to change the value of I_{ire} . The samples' corresponding reflectivity, R_{eff} , was calculated for a sample length, $L=1$ cm. I_{ire} is the transport mean free path at the emission wavelength of 620 nm. l_0 is the absorption length of the excitation radiation in neat dye, and R_{eff} is the effective reflective parameter.

threshold. In the samples where stimulated emission was observed, the duration of the emission was of the same order as the input pulse. The lasing thresholds for the range of samples tested are given in Table 2.

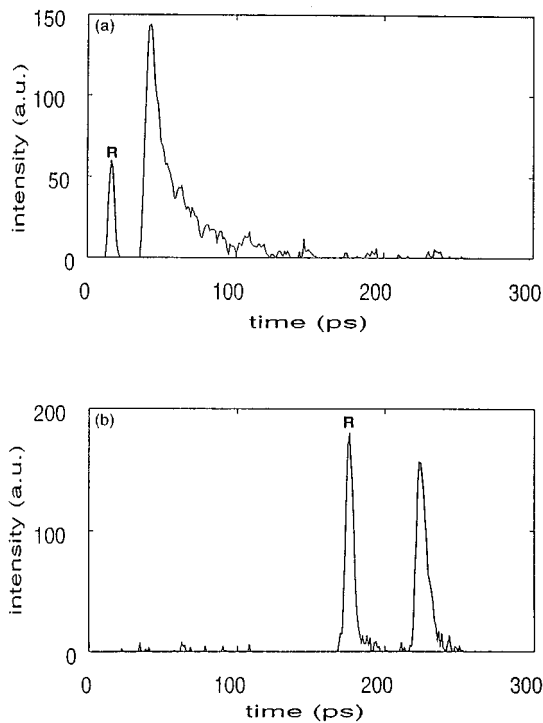


Fig. 2 Time-resolved emission profile of densely packed TiO₂ particles in rhodamine 640 solution. (a) Emission profile at threshold due to excitation by pulse energy of 1.5 μ J and (b) emission of short pulse at excitation pulse energy of 4 μ J. R is the input reference pulse. The amplitudes of the reference and signal profiles are not to scale.

Table 2 Energy threshold measurements from dense powders and Rh640 perchlorate dye from excitations by 10ps at 527 nm pulses.

Powder material	Particle size (diameter, μ)	Threshold energy (μ J)	
		Wet/mucky	Dry paste
Al ₂ O ₃	5	5	10
Al ₂ O ₃	0.3	7	10
Al ₂ O ₃	0.03	20	100
TiO ₂	0.25	1.5	Nonlasing

The active biological tissues were measured for both temporal and spectral shortening of input intensity-dependent emission profiles.¹³ At lower input intensity, a broad bandwidth fluorescence emission spectrum was observed which narrowed when the input intensity was increased. The narrowing of the spectrum indicated the onset of laser action. When the freshly exposed tissue was excited, a narrowing of the emission spectrum was readily observed when the input intensity was above a threshold value. The results of these measurements are displayed in Fig. 3 for chicken tissues. The bandwidth of the fluorescence at low input intensity was measured to be about 30 nm, which narrowed to less than 9 nm when the input intensity was increased to the threshold energy of 20 mJ/pulse. The intensity-dependent temporal profiles from excited chicken tissues with rhodamine 640 dye in methanol are plotted in Fig. 4. A long decay time was observed, which is characteristic of spontaneous emission of the dye; it was more than 4 ns and shortened to about 20 ps when the input intensity was increased beyond the laser action energy threshold. These results are consis-

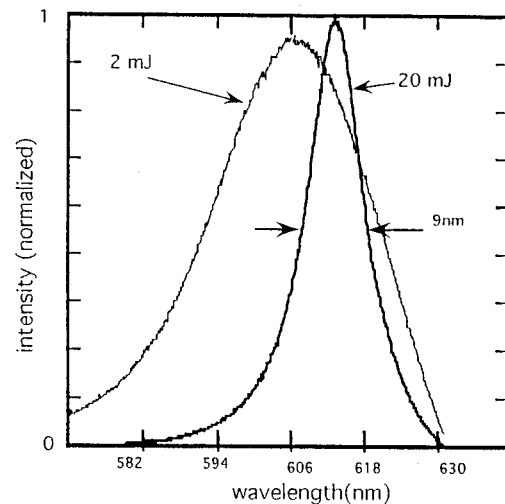


Fig. 3 Spectral profiles of emission radiation from optically excited rhodamine 640 dye in chicken tissue for excitation levels of 2 mJ and 20 mJ pulses.

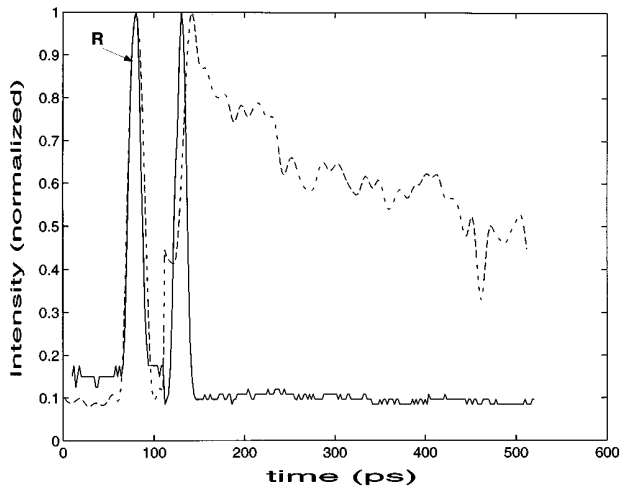


Fig. 4 Time-resolved emission from optically excited rhodamine 640 dye in chicken tissue. Dashed line, long duration spontaneous emission from excitation by a 30 μ J pulse; solid line, ultrafast pulse emission when excited by a 400 μ J pulse. *R* is the input reference pulse. The reference and signal profiles have been normalized.

tent with previously reported observations¹⁴ at different excitation parameters, demonstrating the same physics.

4 DISCUSSION

To explain the laser action in disordered media, a simple scattering walls feedback model, as shown in Fig. 5, is envisioned.¹⁵ The scatterers act as a feedback conduit inside the media by forming photon-confining walls around the directly pumped excited region. The directly pumped excited region is shaped like a thin disk whose diameter is the exciting pulse's spot size and the thickness L_a is the absorption length, modified by the presence of scatterers, of the excitation wavelength, into the scattering medium given by diffusion theory¹⁶ is:

$$L_a = \left(\frac{l_{tr} l_a}{3} \right)^{1/2} ; \text{ for } l_a \gg l_{tr},$$

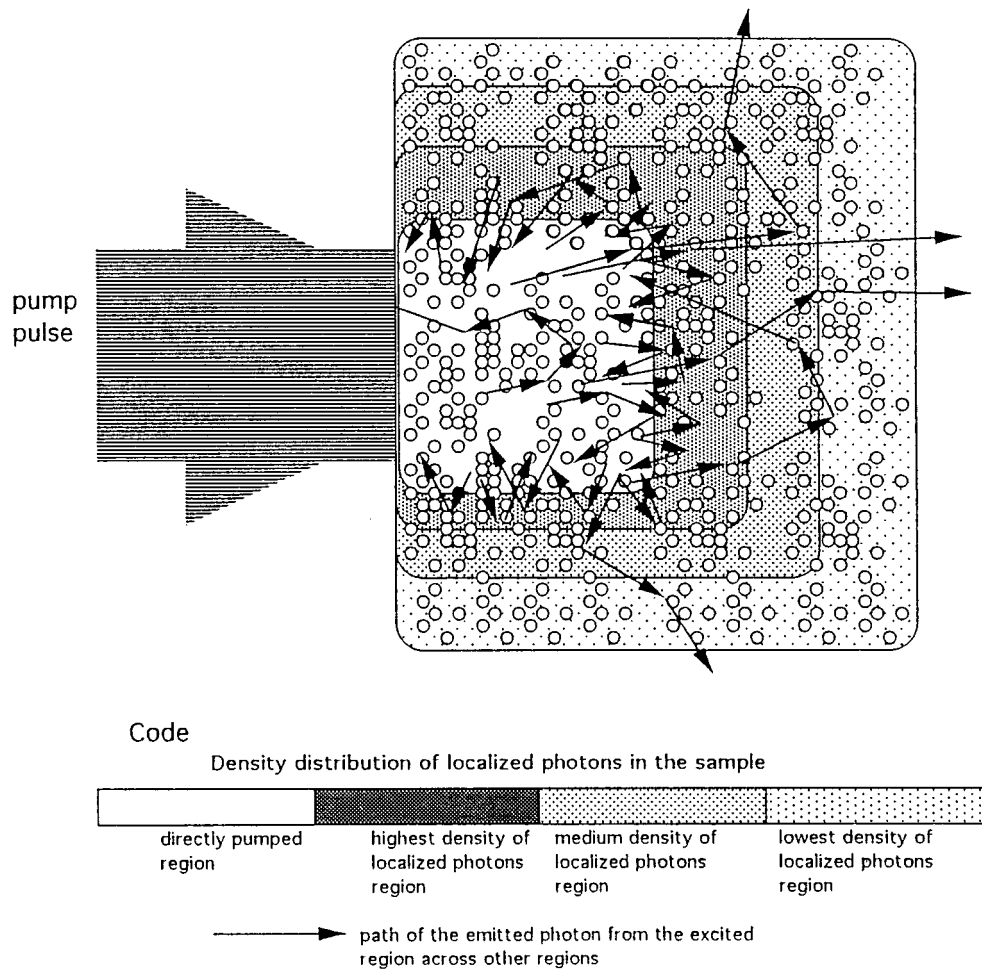


Fig. 5 Pictorial model of feedback from the photon-confining scattering walls. The photons are reflected back into the optically excited disk by the reflecting walls formed by the scattering media surrounding the active disk formed by the excitation pulse. The reflectivity of the surrounding scattering walls is dependent on the scatterer density.

and for the case of $l_a \ll l_{tr}$, $L_a \sim l_a$. In either case, the excitation radiation is absorbed near the surface to a maximum depth of l_a (140 μm , in this case) or less. When the flux of photons spontaneously emitted from the excited disk and scattered back from the surrounding wall of the host medium into the excited medium reaches a critical limit, the energy deposited in the medium by the pump pulse is extracted by the stimulated emission, which is dominated by the wavelengths with the highest gain.

The amount of feedback is dictated by the reflectivity of the scattering walls around the directly pumped region, where the confined photons reach a density maximum.¹⁷ The reflectivity of these scattering walls is dependent on the transport mean-free-path at the emission wavelength, l_{tre} . A qualitative measure of the amount of feedback induced by these scattering walls back into the excited region can be considered as the effective reflectivity of the scattering medium given by^{18,19}:

$$R_{\text{eff}} \propto 1 - \left(\frac{l_{tre}}{L} \right),$$

where L is the transverse dimension of the scattering region, l_{tre} , is the transport mean-free-path and R_{eff} is the effective reflectivity of the medium at the emission wavelength.

The amount of energy extracted by stimulated emission is dependent on the feedback due to reflectivity. Higher reflectivity implies more feedback, thus lowering the required excitation intensity to reach lasing threshold. When the scatterer density is made progressively lower (increasing the l_{tre}), the reflectivity of the scattering walls around the excited region decreases, which in turn increases the required input excitation intensity to reach the critical feedback level needed for laser action. To support this model, the effect of reflectivity of the scattering host on input threshold energy is presented in Table 1. The feedback reflectivity is shown to increase with a decrease in l_{tre} along with its corresponding decrease in input threshold energy. This result supports the scattering-feedback model.¹⁴ In the case of densely packed powders, as presented in Table 2, the R_{eff} for TiO_2 is in the order of 0.9996, which is much higher than the dilute colloidal sample case. The threshold is accordingly lower. In this region, one has to keep in mind that in the case of densely packed powders, the value of the host dye concentration is altered because the volume content of the powders is more than 50% and photon localization effects may come into play to enhance the feedback effects.

The case of laser action in animal tissues is quite novel since the structure of the tissue is dramatically different from the discretely scattering media. The animal tissues contain cells up to 10 μ in size, within which is an assortment of finer structures as well as regions of varying refractive indices, such as the nuclei, making such tissues a truly heteroge-

neous continuously disordered medium. The scattering in these structures will give rise to photon paths different from the discretely scattering powders and colloidal solutions of liquid dyes. By the same measure, the relative R_{eff} in chicken tissue will be 0.75 and 0.965 in fat, which shows relatively higher thresholds compared with the discretely scattering hosts. The higher threshold is accounted for by the lower reflectivity and the inherent difference in scattering. These measurements support the scattering wall model of lasing action arising from feedback from the surrounding scattering reflective medium.

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