Dynamics of stimulated emission from random media

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The spatial and spectral evolution of emission from gain media with homogeneously distributed scatterers is modeled using a Monte Carlo simulation of the random walk of pump and emitted photons. We find a sharp transition to ultrafast, narrow-linewidth emission for a 10-ps incident pump pulse and a rapid approach to steady state for longer pulses. The threshold for this transition is raised significantly when the transverse diffusion of photons out of the excited region is substantial. The quantitative agreement of the results with recent observations of emission from random gain media lends further support to the interpretation of ''random lasers'' in terms of amplified spontaneous emission within a multiply-scattering medium. [S1063-651X(97)06211-9]

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I. INTRODUCTION

The conjunction of lasing and disorder has been of interest since shortly after the advent of the laser [1,2]. Recent observations of lasing action in rare earth doped powders [3] and in colloidal suspensions in dye solutions [4] have renewed interest in amplifying random media. A dramatic narrowing of the spectrum [4,5] and shortening of the emission time [6] is observed above a threshold in pump energy. These results have raised the prospects of utilizing the phenomenon for a variety of display, sensing, and switching applications [7], particularly if the corresponding threshold can be significantly reduced. The colloidal-dye systems are of special interest for understanding emission in random media because the gain and scattering can be varied independently so that computations can be compared with measurements over a wide range of sample properties.

There has been considerable debate regarding the origin of enhanced stimulated emission in random media. Some authors have suggested that lasing action can be explained by considering propagation along specific paths in the medium in analogy with lasing in homogeneous media. Balachandran and Lawandy [8] have proposed a ring laser model. Coherent backscattering of a probe laser beam in opaque amplifying random media, proposed by Zyuzin [9], has been observed [10] and found to be consistent with an intensity distribution on the surface of the medium obtained by solving the diffusion equation for the probe photons with gain. In recent work, we have used a Monte Carlo simulation to understand the emission dynamics in a colloidal dye solution for the case of short pulse excitation studied by Siddique *et al.* [6]. Recently John and Pang have solved the steady-state diffusion equation with nonlinear gain and loss, but without saturation for colloid-dye systems [11] and Wiersma and Lagendijk have solved the spatial diffusion equation with gain to obtain the time dependent response of powdered lasing crystals to excitation by a Q switched laser [12]. In both studies the coupled nonlinear diffusion equations were solved numerically for plane wave excitation. Here we explore the evolution of emission by following the migration of photons and molecular excitation as determined purely by local probabilities. We demonstrate that a Monte Carlo simulation of the random walk of photons, taking into account the nonlinear relationships among local molecular energy-level populations and photon densities within the frequency band of spontaneous emission, predicts behavior that is consistent with experimental observations. This approach is equivalent to solving the coupled nonlinear diffusion equation and molecular rate equations. By treating the essential spatial, temporal, and spectral aspects of the problem, we are able to study the dynamics within the gain medium in detail.

The simulations are carried out in homogeneously scattering samples that are excited by a short incident laser pulse. We consider emission from the front of a sample with transport mean free path ℓ and absorption length ℓ_a because the exponential attenuation length $L_a=\sqrt{\ell \ell_a/3}$ of the pump light is much smaller than the sample thickness L. We have modeled emission for excitation by a plane wave and by a Gaussian beam. Differences between these cases will be discussed, but our primary focus will be the results of simulations for plane wave excitation. This simpler case exhibits the essential features of lasing in the presence of disorder and enables a full study of the dependence upon various parameters of the problem in a moderate computation time. Furthermore, it allows us to visualize the simultaneous spatial and spectral evolution of the molecular excitation and electromagnetic energy within the medium.

II. MONTE CARLO SIMULATION

We model the gain medium as a four level system. For the pump radiation considered, intersystem crossing and excited state absorption can be neglected. The populations in the fourth and second energy levels are negligible, due to rapid nonradiative transitions to the third and first levels, respectively. The density of molecules in the first energy level is then given by $N_1(\mathbf{r},t)=N-N_3(\mathbf{r},t)$, where N is the total density of dye molecules and N_3 is the density in the third level. Thus, the rate equation for N_3 alone suffices to describe the degree of excitation of the dye molecules. It is

$$\frac{dN_3(\mathbf{r},t)}{dt} = n_p(\mathbf{r},t)N_1(\mathbf{r},t)v\sigma_a(\nu) - \frac{N_3(\mathbf{r},t)}{\tau} + \int d\nu \overline{n(\mathbf{r},t,\nu)}[N_1(\mathbf{r},t)v\sigma_a(\nu) - N_3(\mathbf{r},t)v\sigma_e(\nu)], \quad (1)$$

where n_p is the pump photon density and $\overline{n}$ is the density of emitted photons per unit frequency, σ_e and σ_a are the emission and absorption cross sections, respectively, $v=c/n$ is the speed of light in the medium, and τ is the lifetime in the absence of stimulated emission. The four terms on the right-hand side correspond to the rates of absorption of pump and emitted photons, and to the spontaneous and stimulated emission of photons. They are all represented in the simulation where a sum over frequency intervals is used to approximate the integral over the continuous frequency spectrum.

The simulations are carried out for a 1-mm-thick sample of Rhodamine 640 dye with suspended scattering particles. The absorption cross section of the dye of 1.2×10^{-16} cm^2 at the pump wavelength of 527 nm is determined from the absorption measurements of [6]. In that reference, an effective emission cross section was used, which was determined from the lifetime of 4 ns and bandwidth of 40 nm for emission measured in [4]. Here we compute the emission using the actual spectral distribution of spontaneous emission measured in [4]. The peak emission cross section is 5.3×10^{-16} cm^2. Probabilities of reabsorption of the emitted light were determined from the absorption spectrum given in [14].

We assume that the transport mean free paths of the pump and emitted photons are the same. For a dye dissolved in methanol within a glass cuvette, the internal reflectivity R [14] due to the methanol-glass-air interface integrated over all scattering angles is approximately 0.5. The corresponding value for the methanol-glass interface is ≈0.15. While the former value was used in the simulations of plane wave excitation in which transverse diffusion does not play a role, the latter value is a more realistic assumption in the case of Gaussian beam excitation. We find that the emission properties for these two values of R are not very different.

Because the average energy density in the medium is uniform in the transverse direction for plane wave excitation, simulations for this case can be performed by treating only longitudinal diffusion. The average longitudinal displacement for photons moving randomly in three dimensions with displacements of one mean free path ℓ is $\ell/2$. We divide the sample into discrete strips of width $\ell/2$ and assume the photon is displaced by $\pm\ell/2$ in each mean free time $\tau=\ell/v$. The diffusion of photons thus reduces to a random walk in one dimension on a lattice with lattice constant of $\ell/2$. The evolution of the densities of molecular excitation, pump photons, and separately photons that were produced by spontaneous and stimulated emission are each followed in discrete time intervals of τ. We assume the incident photons are randomized at a distance $\ell/2$ into the sample because the value of the transmission is then consistent with the results of diffusion theory. We accounted for the spectral distribution of the emitted photons by dividing the Rhodamine perchlorate emission spectrum into twenty sections of equal weight corresponding to equal values of $\sigma_e\Delta\nu$ and treating each section separately. Whether or not a photon traversing a particular bin at a particular time stimulates emission or is absorbed is decided by drawing a random number and comparing it with the probabilities determined by the rate equations, given the current local values of N_3 and $\overline{n}$. A random number is also used to determine whether a molecule had spontaneously emitted a photon. The populations are updated after each event.

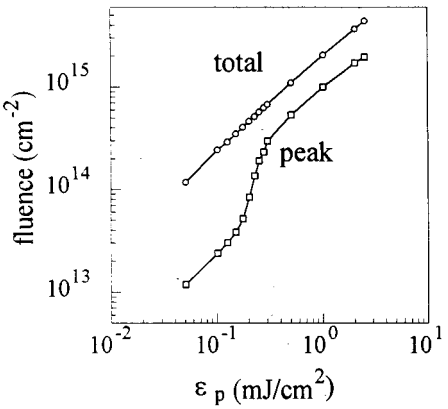

FIG. 1. Log-log plot of the total emission and the peak emission in a band of $\Delta\lambda=5$ nm around 616.5 nm as a function of pump fluence for a sample with $\ell=60$ μm, $\ell_a=300$ μm, and $R=0.5$.

III. RESULTS OF THE SIMULATIONS

We consider the change of emission properties with increasing pump energy for a system with $\ell=60$ μm and $\ell_a=300$ μm, and for a square 10-ps pump pulse. The number of emitted photons at all wavelengths and in a wavelength interval from 614 to 619 nm are shown in Fig. 1. In this spectral interval, the peak of the emission shifts with pump fluence as shown in Fig. 2. The total emission increases nearly linearly. On the other hand, the intensity at the peak emission frequency exhibits a clear change of slope within a narrow range of pump energies. At these energies,

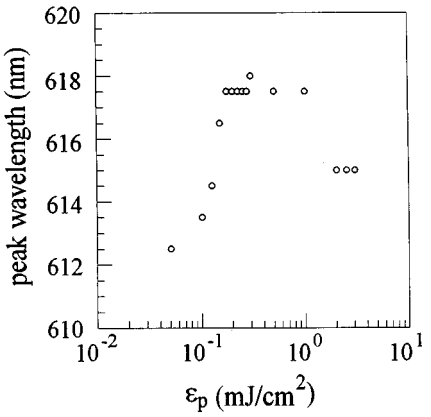

FIG. 2. Semilogarithmic plot of the peak wavelength as a function of pump fluence for the same sample as for Fig. 1.

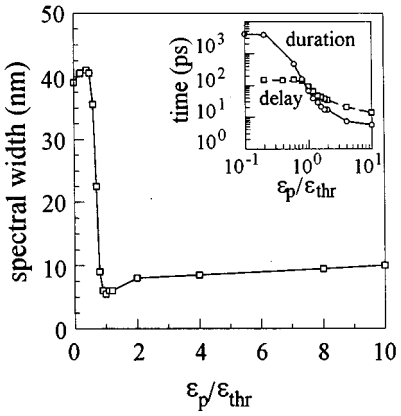

FIG. 3. Spectral width of the emission as functions of pump fluence. The sample parameters used are the same as for Fig. 1. The inset shows a log-log plot of the pulse duration and the delay of the peak emission with respect to the leading edge of the pump pulse as a function of pump fluence. $\epsilon_{thr}=0.26$ mJ/cm^2.

the ratio of the intensity of stimulated to spontaneous emission, R_{em}, begins to exceed unity and stimulated processes lead to marked narrowing of the spectral and temporal emission characteristics [4–6]. It is therefore useful to define $R_{em}=1$ as the threshold for copious stimulated emission. The sharpness of the transition reflected in the spectral and temporal response of the system is shown in Fig. 3. These input-output relations are in agreement with the observations of Lawandy *et al.* of spectral narrowing [4].

In contrast to the peak emission, the total emission shows no change in behavior near threshold; cf. Fig. 1. In contrast to a nonscattering amplifying medium inside an external cavity, spatial mode selectivity is absent in a random system. The change of slope in the peak emission in the transition region simply reflects the narrowing of the emission spectrum, see Fig. 3.

A comparison of emission generated by 10- and 100-ps incident pump pulses at the same flux levels is presented in Fig. 4. The emission following the onset of the pump pulse

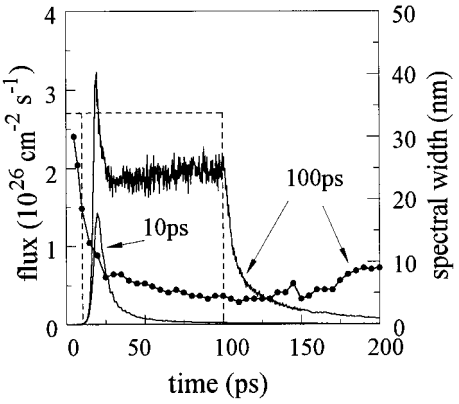

FIG. 4. Emitted flux for two pump pulse durations using the same sample parameters as for Fig. 1. The pump fluence is 1 mJ/cm^2 for the 10-ps pulse and 10 mJ/cm^2 for the 100-ps pulse. The dashed lines indicate the incident pump flux. The dots show the temporal evolution of the spectral width for the 100-ps pulse.

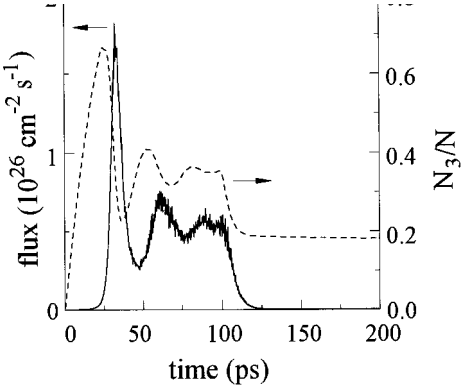

FIG. 5. Emitted flux (solid curve) and normalized excited state population at a depth of ≈ 50 μm in the sample where the gain is strongest (dashed curve). The sample length is 0.18 mm and $R=0$. All other sample parameters are as for Fig. 1. The pump fluence is 5 mJ/cm^2 and the pump pulse duration is 100 ps.

exhibits comparable delays and a short burst of emission. For the longer pulse we observe the approach to quasi-steady-state emission, which is achieved within 30 ps. The evolution of the emission bandwidth shows that the quasi-steady-state emission is dominated by stimulated processes. The rapid approach to steady state after the initial emission burst indicates that the relaxation oscillations that are typical for the transient behavior of many lasers, particularly solid-state lasers [15], are strongly damped. Pronounced oscillations (spiking) have been predicted recently in powdered lasing crystals [12]. Under certain conditions we find that spiking also occurs in colloidal dye systems as seen in Fig. 5. Spiking is achieved by using thin samples to reduce the dwell time of photons in the medium. This raises the modulation frequency of the intensity so that it exceeds the overall decay rate of the modulation.

The results of our simulations are in good agreement with the spectral [4] and temporal [6] properties of emission that have been observed experimentally. However, the thresholds that we obtain are an order of magnitude lower than the experimental values under comparable conditions [6]. In the experiments, transverse diffusion of the photons out of the laterally confined gain region plays a significant role. Transverse diffusion is irrelevant for the simulations with plane wave excitation because the gain is laterally uniform. For a finite gain region produced by a Gaussian beam excitation, transverse diffusion must be taken into account. In the model, the incident beam is divided into radial sections. The simulation keeps track of a network of bins, one bin for each ordered pair (ρ,z) in cylindrical coordinates for which the number densities of photons and the population densities of excited molecules are followed. The angular distribution was neglected since the system is assumed to have cylindrical symmetry.

Figure 6 shows the bandwidth and pulse duration of the emission for excitation with a Gaussian beam with a waist 250 μm for the same sample parameters as for Fig. 3. As expected, the threshold, defined as the pump fluence at which the spectral width is a minimum, rises, from 0.4 to 1.6 mJ/cm^2. For Gaussian beam excitation the ratio R_{em} is not a good measure for the emission characteristics. In con-

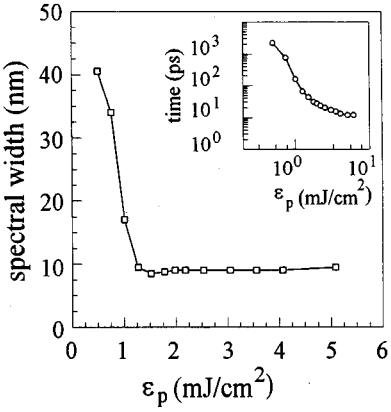

FIG. 6. Spectral width of the emission as functions of pump fluence for Gaussian beam excitation for a sample with $\ell = 100\ \mu$ m, $\ell_a = 500\ \mu$m, and $R = 0.15$. The inset shows a log-log plot of the pulse duration as a function of pump fluence.

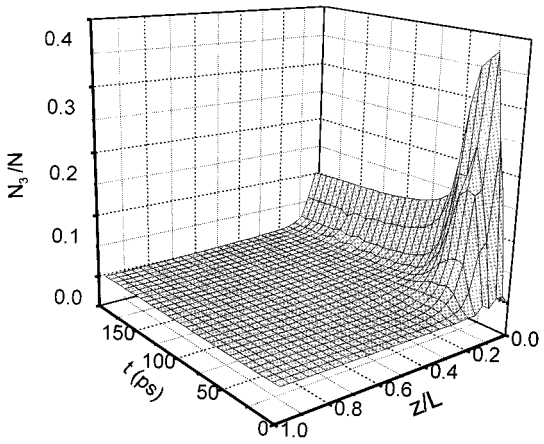

FIG. 7. Degree of molecular excitation N_3/N as a function of depth inside the sample and time. The sample parameters are the same as for Fig. 1 and the pump fluence of 0.56 mJ/cm^2 is approximately twice the threshold value.

trast to plane wave excitation, a substantial amount of excitation remains in the sample after the initial emission burst and decays slowly once the incident pulse is no longer present largely via spontaneous emission. The spectral and temporal emission peaks, however, are primarily determined by the early stimulated emission before transverse diffusion leads to a substantial migration of photons out of the gain region.

IV. DISCUSSION

The spectral and temporal emission characteristics above threshold can be understood in terms of the dominance of stimulated processes resulting from increased path lengths of photons in the region with high gain due to multiple scattering. The spectral features result from the interplay of amplification and absorption of the spontaneous emission in the sample region where the photons spend most of their time. For low excitation, the gain region is confined to the very front of the sample. Amplification and absorption play a minor role and the emission spectrum is that of spontaneous emission. With increased pumping, longer path lengths of emitted photons lead to a shift of the emission peak towards higher wavelengths where the absorption is lower and a spectral narrowing due to amplification at frequencies with the largest gain. If the pumping is increased even further, depletion of the ground state reduces absorption. This results in a shift of the peak emission back towards smaller wavelengths where the amplification is stronger. These features are clearly visible in Fig. 2 and have been observed by Noginov *et al.* [16].

A particularly striking result of the simulation is the short time required for the buildup of stimulated emission and its short duration above threshold. In Fig. 7 we present the distribution of molecular excitation in space and time for excitation at a fluence of twice the threshold level for plane wave excitation. Despite considerable depletion of the ground state, the molecular excitation is highly concentrated near the front surface of the sample in the present system. This is very different from the case of the powdered Ti:sapphire sample with weak absorption studied by Wiersma and Lagendijk [12] where the excitation builds up throughout the sample. The average path length of photons emitted in the gain region is approximately $z^2/\ell \leqslant (120^2/60)\ \mu\text{m} \approx 240\ \mu$m. This length is comparable to ℓ_a [18]. Since the photons traverse this distance in approximately 1 ps, their migration out of the sample does not limit emission dynamics. Significant stimulated emission, indicated by the rapid falloff of N_3 in Fig. 7, occurs only in the short time window in which the photon density and the excited state population density in the front layer of the medium are both large. The resulting emission in a single picosecond burst has been observed by Siddique *et al.* [6].

For longer pumping of the colloid-dye system, laser spiking can be observed if the depletion of the upper-level population by stimulated processes is as efficient as the pumping. This is not the case if the gain region (i.e., the region excited efficiently by the pump laser) is confined to the front layer of the sample while the emitted photons diffuse throughout the whole sample volume. We can make the sample volume more uniformly excited without changing dye-colloid parameters by reducing the sample length. This and the reduction of the internal reflectivity also reduce the lifetime of the photons in the medium. The period of the oscillations of the emission shown in Fig. 5 is $\approx$30 ps, about three times shorter than what one estimates from the approximate analytic solution of the time dependent laser rate equations for a homogeneous medium [15].

While the qualitative features of emission observed from random dye solutions are captured well by the simulations, there remains a difference in the exact value of the threshold, even when transverse diffusion is taken into account in the simulations with Gaussian beam excitation. This remaining difference is probably attributable to our assumption that the transport mean free paths of pump and emitted photons are the same. Our measurements show that for the titania powders used in many experiments ℓ for the pump photons at about 530 nm is approximately half that for the emitted photons at about 620 nm. This should roughly double the threshold fluence since the average pathlength for emitted photons in the gain region would then be halved. Such a threshold would be in the range of thresholds observed in experiments with comparable parameters [6].

In conclusion, we have studied the properties of emission from optically pumped random gain media by Monte Carlo simulation of the random walk of photons. By fully modeling the nonlinear problem we show that the emission from random media can be explained in terms of incoherent excitation and migration.

ACKNOWLEDGMENTS

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