Modeling of photon density dynamics in random lasers

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The dynamics of stimulated emission in random lasers is studied using the system of rate equations for population inversion and density of emitted photons. In order to model the behavior of random lasers with nonresonant feedback, no coherence effects are intentionally taken into account. It has been shown that the feedback in the system is necessary for the realization of the regime of relaxation oscillations and spatial confinement of the stimulated emission to the interior regions of the pumped volume. The model also predicts the possibility of localization of stimulated emission in several spatially separated subvolumes of the random laser medium. Finally, it has been demonstrated that two adjacent random laser volumes can strongly enhance the stimulated emission in each other (the "critical mass" effect in random lasers).

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I. INTRODUCTION

Random lasers are the simplest sources of stimulated emission without cavity, with the feedback provided by scatterers. After their first theoretical prediction [1,2] and experimental observation [3,4], a variety of random lasers has been reported in the literature. This includes random lasers based on highly scattering dielectrics materials doped with Nd^{3+} [5–9], Pr^{3+} [10], Ti^{3+} [11] and other ions, ZnO random lasers [4,12,13], random lasers based on scattering polymers [14–16], etc. Due to the easy manufacturing, low price, small size, and robustness in operation, random lasers are very attractive for potential applications, which include express testing of novel laser materials [5], markers [17,18], high brightness laser displays [19], etc. In spite of a large number of studies and publications on random lasers, the mechanisms of their operation are not completely understood, which hinders optimization of random laser performance and use of these unique sources of stimulated emission.

In this work, we theoretically study the dynamics of stimulated emission in random lasers with nonresonant feedback, such as neodymium random lasers, which are characterized by low coherence [8,20]. Therefore, we intentionally neglected coherence and interference effects in our model. That makes this work different from Refs. [21,22], where coherence effects were claimed to play a determinant role in the random laser behavior. (It is obvious that the model neglecting coherent effects is not designed to describe multiple narrow lines in the emission spectra of random lasers with resonant feedback.)

II. MODEL AND COMPUTATION PROCEDURE

The system of rate equations for the population inversion n and the density of emitted photons E, which has been proven to adequately describe the dynamics of a neodymium random laser both qualitatively and quantitatively [9], is used to model the dynamics of stimulated emission in this work,

$$\frac{dn}{dt} = \frac{\frac{P(t)}{Sl_p}}{h\nu_{\text{pump}}} - \frac{n}{\tau} - \frac{E}{h\nu_{\text{em}}} c\sigma_{\text{em}} n,$$
$$\frac{dE}{dt} = -\frac{E}{\tau_{\text{res}}} + \frac{n}{\tau} h\nu_{\text{em}} + Ec\sigma_{\text{em}} n. \qquad (1)$$

Here $P(t)/S$ is the pumping power density, l_p is the penetration depth of pumping (determined by absorption and scattering), σ_{em} is the emission cross section at the wavelength of stimulated emission, $h\nu_{\text{pump}}$ is the photon energy at the pumping wavelength, $h\nu_{\text{em}}$ is the photon energy at the emission wavelength, τ is the luminescence lifetime of the upper laser level $^4F_{3/2}$ (which depends on both radiative and nonradiative decay processes), τ_{res} is the effective residence time of a photon in the pumped volume (in conventional lasers, a similar term represents the lifetime of the photon in a cavity), and c is the speed of light.

The spectroscopic parameters used in the calculations were similar to those in $NdAl_3(BO_3)_4$ powder, $\sigma_{\text{em}}=1\times10^{-18}$ cm^2, $h\nu_{\text{abs}}=4\times10^{-19}$ J, $h\nu_{\text{em}}=2\times10^{-19}$ J, and $\tau=20$ μs [9]. The pumping pulse was assumed to have Gaussian form with the full width at half maximum (FWHM) equal to 10 ns.

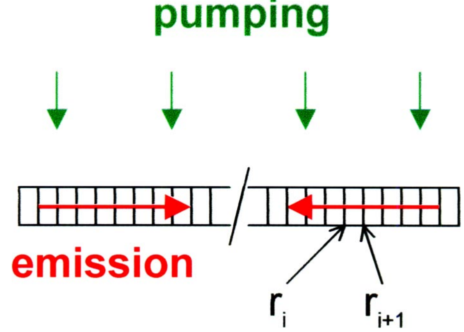

FIG. 1. (Color online) Schematic diagram of a one-dimensional strip of lasing volumes.

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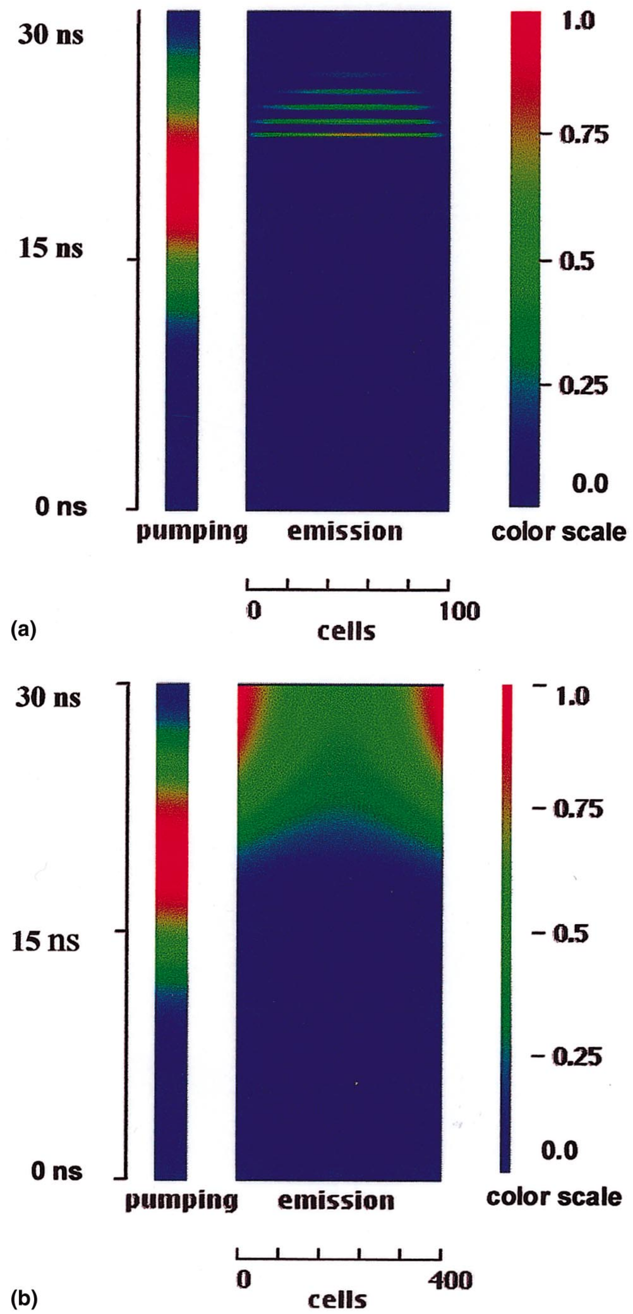

FIG. 2. (Color) Calculated emission dynamics in (a) a strip of 100 cells; pumping intensity corresponds to 1 J/cm^2 of absorbed energy, $r_{aver}=8\%$, the residence time τ_{res} is calculated to be 0.43 ps, the maximum emission intensity is equal to 24 566 rel. units. (b) A strip of 400 cells pumped with the same intensity, with no reflection at the boundaries of the cells; the residence time is calculated to be 0.67 ps, the maximum emission intensity is equal to 16 rel. units. Loss is neglected in both (a) and (b).

We calculated the dynamics of stimulated emission in a one-dimensional strip of amplifying volumes (cells) separated by partially reflective walls, Fig. 1. The cell size was equal to 1 μm, which corresponded to a typical particle size in neodymium random lasers. All cells were assumed to be pumped uniformly. In our model, emission could propagate only along the strip, one photon flux going to the right and one to the left. Reflection coefficients of the cell walls r were calculated with the help of a random function generator and were randomly distributed between $r=0$ and $r=2r_{aver}$, where r_{aver} was the average reflection coefficient. In a similar way, the photon loss l (caused, for example, by absorption) was calculated for each cell, with the value of l randomly distributed between $l=0$ and $l=2l_{aver}$, where l_{aver} was the mean value of the loss. In the computation procedure, instead of direct introduction of the residence time τ_{res} to the system, two neighboring cells were allowed to "communicate" with each other, exchanging with photons, in accordance with the reflection and transmission coefficients assigned to each intercell boundary. The bouncing of reflected photons back and forth determined the photon residence time in a strip.

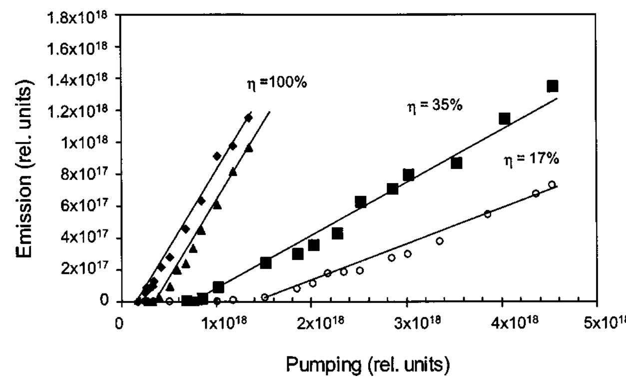

FIG. 3. Input/output curves calculated for a strip of lasing volumes. Diamonds: $r_{aver}=10\%$, $\tau_{res}=0.50$ ps, no loss; squares: $r_{aver}=10\%$, $\tau_{res}=0.50$ ps, 1% loss; circles: $r_{aver}=10\%$, $\tau_{res}=0.50$ ps, 2% loss; triangles: $r_{aver}=5\%$, $\tau_{res}=0.35$ ps, no loss. The units on the axes correspond to the numbers of emitted or absorbed photons.

III. CALCULATION RESULTS

Only spontaneous emission and relatively weak amplified spontaneous emission (ASE) with relatively low intensity were calculated in the system at low pumping energies. With increased pumping, after reaching the threshold, the first short relaxation oscillation pulse occurred. The peak intensity above the threshold was ~1000 times greater than that just below the threshold. With the further increase of pumping intensity, the number of short stimulated emission pulses increased, as well as their intensity. The pulses were getting shorter, they were shifted toward the beginning of the pumping pulse, and the frequency of relaxation oscillations was getting higher. This type of behavior was similar to that observed experimentally in many neodymium random lasers [5,8,9]. The typical calculated two-dimensional (time/cell number) kinetics of emission above the threshold is shown in Fig. 2(a).

In the calculation presented in Fig. 2(a), the strip consisted of 100 cells, r_{aver} was equal to 8%, and the mean residence time of the photon in the system was equal to $\tau_{res}=0.43$ ps. As follows from this figure, in the system with feedback the emission is getting localized (confined) in the central part of the pumped volume. In a number of theoreti-

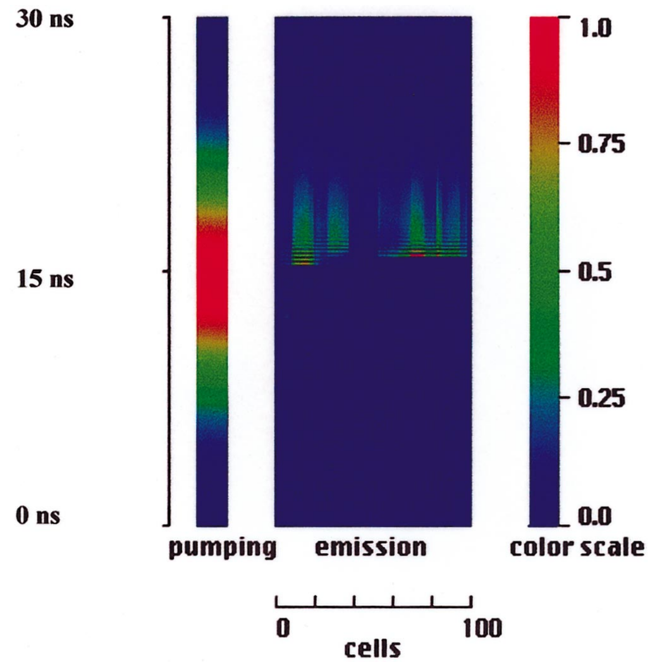

FIG. 4. (Color) Calculated emission dynamics in a 100-cell strip with loss introduced to the system, $l_{aver}=4\%$, pumping intensity corresponds to $10\ \mathrm{J/cm^2}$ of absorbed energy, and the maximum emission intensity is equal to 29 270 rel. units. Multiple regions of light localization are seen in the figure.

cal and experimental works (see, for example, Refs. [22–25]), localization of the random laser mode to the volumes with characteristic sizes ranging from a fraction of the wavelength to hundreds of wavelengths has been reported. However, in the references above, localization was treated as a property directly relevant to the coherence of a light wave. As we show in this work, spatial localization of the random laser intensity also takes place in the case of incoherent (nonresonant) feedback.

Note that the relaxation oscillations calculated above are similar to those predicted by the diffusion model [1,2]. However, spatial localization of emission was not explicitly considered in Refs. [1,2].

Figure 2(b) shows the dynamics of stimulated emission in a strip consisting of a larger number of cells, 400, and without reflection at the cell boundaries, $r_{aver}=0$. Because of the larger size of the strip, the mean photon residence time τ_{res} was equal to 0.67 ps, longer than that in the strip of Fig. 2(a). The amount of pumping energy per cell in Fig. 2(b) was the same as in Fig. 2(a). However, despite longer residence time, no short high-intensity pulses of stimulated emission have been predicted in the long strip. Furthermore, no spatial confinement of emission to the central part of the pumped volume is seen in Fig. 2(b). Instead, the emission has its maximum values at the ends of the strip, which is an expected behavior in the case of amplification in open paths. Although we have not proved this rigorously, it appears likely that relaxation oscillations and the spatial confinement accompany each other and can serve as evidence of a feedback in the system.

The majority of known solid-state random lasers, including optically pumped neodymium lasers [5,8,9], ZnO random lasers [22], and random lasers based on scattering polymers [26], operate in the pulsed regime with relaxation oscillations. Based on the calculated result above, we conclude that all types of random lasers listed above do have a feedback, which is necessary for the realization of relaxation oscillations and the confinement of a lasing mode. Simple ASE in photon open paths, which are elongated by scattering, is not enough to cause the stimulated emission dynamics similar to that observed experimentally.

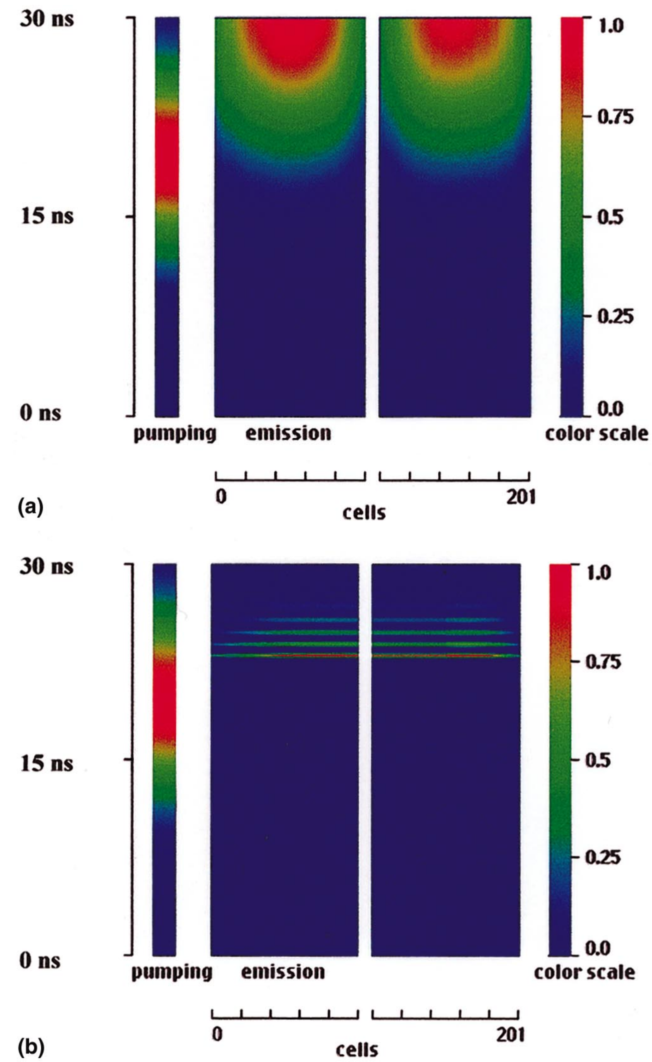

FIG. 5. (Color) Calculated emission dynamics in (a) two adjacent pumped volumes separated by an opaque screen, pumping intensity corresponds to $1\ \mathrm{J/cm^2}$ of absorbed power, and emission maximum is equal to 6.6 rel. units; (b) two volumes separated by an 80% transmissive screen, pumped with the same intensity as in (a); emission maximum is equal to 19 655 rel. units. In both parts, $r_{aver}=3\%$, $l_{aver}=0$.

The diffusion model predicts high quantum slope efficiency of random laser emission, approaching 100% when all incident pumping energy is absorbed in the sample. However, the experimentally measured slope efficiencies in neodymium random lasers are much lower (0.2% in Ref. [9], ~1% in Ref. [27], or 20–25 % in Ref [5]). To describe the reduced slope efficiency in our theoretical model, we introduced a loss to the system. In the one-dimensional model, the loss can have a physical meaning of passive absorption. However, in two-dimensional or three-dimensional models, loss can also be due to the escape of emitted photons from the pumped volume. Figure 3 shows input/output curves calculated in the system with and without loss and with different photon residence times. As follows from this figure, the change of the photon residence time influences the threshold but does not influence the slope efficiency. On the other hand, loss in the system simultaneously increases the threshold and reduces the slope efficiency. Thus, we demonstrate that loss can be the mechanism responsible for a significant reduction of the slope efficiency in random lasers.

In the presence of loss, multiple spatially confined locations of stimulated emission can be predicted in a strip of lasing cells, Fig. 4. (At the threshold, the stimulated emission in a strip occurs only in one localized spot. However, with the increase of the pumping energy, the number of locations of confined stimulated emission increases.) A qualitatively similar result was reported in Ref. [22], where the coherence effects were taken into account and claimed to be principally necessary for the predicted stimulated emission dynamics. As we show in this work, multiple spatial localizations of stimulated emission can also be predicted in random lasers with *incoherent* feedback.

In the last example, we demonstrate stimulated emission in two adjacent pumped laser volumes, which can exchange with photons. First, we show that at a certain pumping density, the intensity of emission in two volumes separated with an opaque screen (no exchange with photons) is low and no relaxation oscillations are observed, Fig. 5(a). No loss is taken into account in this particular calculation. At the same pumping, but with a partially transparent screen (transmittance=80%), stimulated emission characterized by relaxation oscillations appears in the system of two coupled laser volumes, and the maximum emission intensity is increased approximately 3000 times, Fig. 5(b). In contrast with the stimulated emission in a single strip, where the stimulated emission is confined to its central part, the maximum of the emission intensity in a couple of adjacent volumes is close to the gap separating the volumes. Thus, effectively the two volumes operate as a single one. This phenomenon is similar to the critical mass effect in nuclear fusion. This analogy was first suggested by Letokhov in Ref. [2]. In a similar calculation, but with a loss introduced to the system, the volumes did not communicate with each other and the absence of an opaque screen did not cause any significant changes in the stimulated emission.

IV. SUMMARY

To summarize, we have shown that (i) feedback in random lasers is essential for the regime of relaxation oscillations. Elongation of open photon paths by scattering is not enough to produce a train of short stimulated emission pulses in response to one longer pumping pulse (relaxation oscillations). (ii) Relaxation oscillations and the spatial localization of the stimulated emission accompany each other and apparently serve as evidence of a feedback in the system. (iii) The photon residence time influences the stimulated emission threshold and does not affect the output slope efficiency. At the same time, loss in the system increases the threshold and lowers the slope efficiency. This may partially explain low slope efficiency, which is experimentally observed in many random lasers. (iv) In the system with loss, multiple spatially localized volumes of stimulated emission can be predicted in random lasers with nonresonant feedback. (v) We have demonstrated that two adjacent random laser volumes enhance each other's performance when they can exchange with photons. This phenomenon is analogous to the critical mass effect known in nuclear fusion.

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